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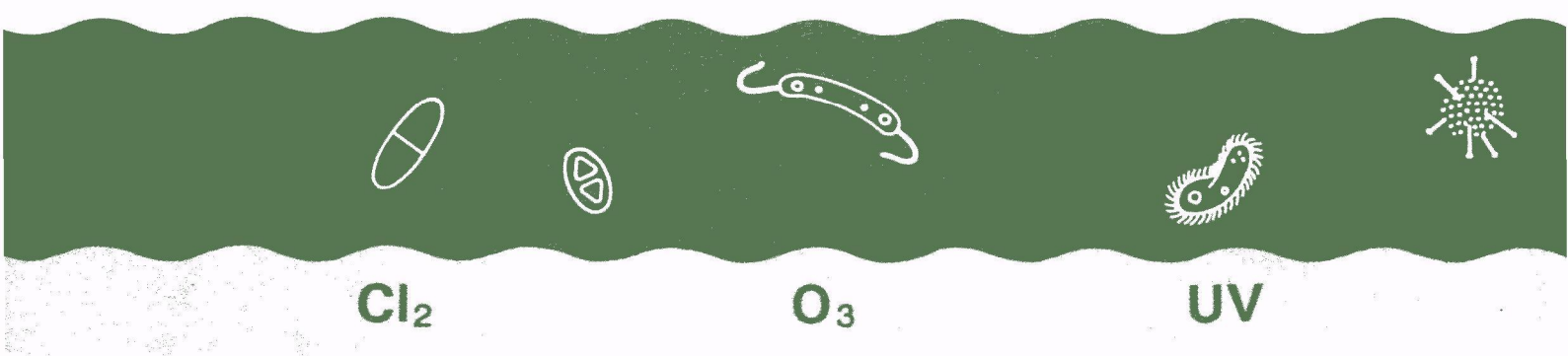
Municipal Wastewater Disinfection

Proceedings of Second National Symposium



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MUNICIPAL WASTEWATER DISINFECTION

Proceedings of Second National Symposium
Orlando, Florida
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Sponsored by the
Municipal Environmental Research Laboratory
and the
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Edited by

A. D. Venosa and E. W. Akin

MUNICIPAL ENVIRONMENTAL RESEARCH LABORATORY
HEALTH EFFECTS RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OHIO 45268

DISCLAIMER

The following papers have been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

Infective Dose of Waterborne Pathogens
Elmer W. Akin

Viral Gastorenteritis Caused by Norwalk-Like Agents
Raphael Dolin

Risk Assessment of Wastewater Disinfection
David W. Hubly

Wastewater Health Effects Studies and the Need for Disinfection
Walter Jakubowski

Fresh Recreational Water Quality and Swimming-Associated Illness
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Ultraviolet Dose Measurement in Wastewater Disinfection
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Pilot Investigation of Ultraviolet Wastewater Disinfection at the New York City Port Richmond Plant
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A Comparison of Analytical Methods for Residual Ozone
Gilbert Gordon

Control of Ozone Disinfection by Exhaust Gas Monitoring
Albert D. Venosa

Ozone-Mass Transfer Coefficients
Edward J. Opatken

The Effects of Operation and Maintenance
on the Performance of Selected Ozone Systems
Randy Junkins

The work described in the remaining papers was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred

FOREWORD

The Environmental Protection Agency was created because of increasing public and government concern about the dangers of pollution to the health and welfare of the American people. Noxious air, foul water, and spoiled land are tragic testimony to the deterioration of our natural environment. The complexity of that environment and the interplay between its components require a concentrated and integrated attack on the problem.

Research and development is that necessary first step in problem solution and it involves defining the problem, measuring its impact, and searching for solutions.

Two major functions of the EPA research and development program are (1) to develop control technologies and systems to protect people from unnecessary and harmful exposure to wastewater pollutants and (2) to determine the health effects of waste treatment and disposal practices. To these ends, the Municipal Environmental Research Laboratory and the Health Effects Research Laboratory in Cincinnati, Ohio have supported research studies in the respective areas.

This report is the result of a combined effort of the two laboratories to transfer relevant information obtained from recent research studies, most of which were funded by EPA. The holding of a research symposium and the publication of the proceedings is a viable mechanism for disseminating the latest results in a research area. This proceedings provides a comprehensive report on what is known concerning the health and technological aspects of wastewater disinfection.

F. Gordon Hueter, Director
Health Effects Research
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Francis T. Mayo, Director
Municipal Environmental Research
Laboratory

PREFACE

This symposium was the sequel to a similar one on the same topic held in Cincinnati, Ohio, in September 1978. It was designed to address many of the questions raised and deficiencies in knowledge identified at the prior meeting and to address an additional subject area, health aspects. The sessions were organized into three scientifically related but topically separate research areas: (1) health effects and epidemiology, (2) alternative disinfection technology, and (3) design and operation/maintenance considerations.

A brief comment concerning organization of the proceedings' contents is in order. The papers are printed in exactly the same order they were presented. Most of the printed material, however, appears in much greater detail than was presented orally. Those papers requiring peer review according to EPA's publication regulations were so treated. All extemporaneous discussions were tape recorded on site. Unfortunately, however, technical difficulties with the microphone and recording equipment were experienced early in the meeting, and consequently the questions and answers from the audience could not be included in the written proceedings herein. This was truly a disappointing development and the editors wish to apologize for their inability to provide a written record of this valuable informal dialog.

ACKNOWLEDGEMENTS

Appreciation is expressed to the speakers and authors of the papers for their many hours of labor and preparation, to the session chairman, and to the general registrants whose lively participation in the panel discussions contributed greatly to the success of the symposium. We also wish to thank the session moderators for the orderly progression of the sessions. Special thanks is expressed to the banquet speaker, Dr. Arthur Lane, Jet Propulsion Laboratory, whose banquet presentation entitled "The Voyager Odyssey to Jupiter and Saturn - The Legacy of a Master Storyteller" roused the fascination of all who attended.

The editors also acknowledge the perseverance and efforts of Ms. Sheri Marshall of the Dynamac Corporation and Mr. Denis Lussier of EPA's Center for Environmental Research Information for arranging for the hotel and banquet accommodations and coordinating registration and other administrative activities.

CONTENTS

1. DON'T CHLORINATE SEWAGE

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ABSTRACT

During the last decade, fisheries dependent on tributaries and freshwater reaches of Chesapeake Bay have declined significantly. The decline took place in waters that should have benefited most by an unprecedented investment in sewage treatment plant construction. In every case, inquiries into the possible reasons for the losses implicated chlorine. Investigation showed that the use of chlorine at sewage treatment plants discharging into vital fish spawning areas had increased by several fold. More thorough study shows that chlorine and its byproducts are toxic to aquatic life, repel and thus deny spawning grounds to anadromous fish, and at barely detectable concentrations, decimate fish larvae and other first emergent forms of life. Furthermore, it is found that chlorination of ordinary sewage treatment plant effluent provides no significant public health protection and to the contrary, could result in public health hazards that might go undetected.

INTRODUCTION

Chesapeake Bay is the most productive estuary in the world. Under the dual assault of increasing population and a rising standard of living, the Bay has remained surprisingly beautiful and productive after three centuries of civilization. Where the Bay is concerned, Maryland and Virginia have practiced strong conservation measures for more than a hundred years.

However, during the nineteen seventies, aquatic life dependent on the Bay's tributaries showed signs of unusual disturbance. It is in the tributary streams that anadromous fish come to spawn, other fish reside year round, and still others come to forage. For finfish, the struggle to preserve the chemical, physical and biological integrity of Chesapeake Bay will be won or lost in its tributaries and tidal freshwater reaches.

During the Seventies, shad runs almost ceased. The commercial catch from the Susquehanna River and its flats at the head of the Bay dwindled from 184,000 pounds in 1971 to 2,300 in 1979. The Maryland Department of Natural Resources banned further harvesting of shad to protect the last remaining brood stock. Striped bass, the famed rockfish of the East Coast,

went from a condition of plenty to one of relative scarcity. In 1970, the young-of-the-year averaged slightly more than thirty per seine haul during the annual survey conducted by the Maryland Department of Natural Resources. By 1981, the average was barely more than one per haul. Perch and other resident fish showed a marked decline in some tributaries.

It was puzzling that this deterioration took place during the Seventies, a decade of unprecedented expenditure for sewage treatment plants and other water pollution control measures. One possible solution to the puzzle began to emerge as the search for reasons for tributary crop failures progressed. In every case, chlorine was implicated. That led to a look at the use of chlorine. It was found in six spawning rivers that chlorine discharge increased 4.4 fold from 1974 to 1980.

An estimated 13,900 tons of residual chlorine per year are discharged by Maryland sewage treatment plants. Health Department records reveal that 115 sewage treatment plants annually discharge about 300 tons of residual chlorine into spawning rivers.

The practice of chlorinating sewage treatment plant effluent was examined to find if it is a significant factor causing damage to Chesapeake Bay's tributary dependent aquatic life. The public health aspects of the practice were examined also.

DAMAGE TO AQUATIC LIFE

Literature has proliferated in recent years as the damage to aquatic life caused by chlorinated sewage effluent has become more and more apparent. Space will not permit citation of all of the reports and publications reviewed. Instead, a small number have been selected to illustrate conclusions drawn from a far greater volume of literature.

Collins and Deaner (3) quoted literature (9) (10) to show that when wastewater is chlorinated, toxic compounds such as cyanogen chloride can be formed. Questions regarding the formation and nature of the various toxic compounds and their effect on aquatic life remain unanswered because of the complexity of sewage and chlorine reactions.

Work of Michigan's Department of Natural Resources was described which proved that chlorinated sewage is toxic to fish. Fathead minnows and rainbow trout were exposed to chlorinated and unchlorinated sewage effluents. Survival was high during the non-chlorinated phase but in every case, all trout were killed at chlorine residuals that were less than 0.1 mg/l and all minnows were killed at chlorine residuals less than 0.2 mg/l. The amperometric method was used to measure chlorine residuals. The extreme toxicity is demonstrated by the finding that amperometric chlorine of only 0.02 mg/l below two of the outfalls in Michigan killed 50 percent of the exposed rainbow trout within 96 hours.

Collins and Deaner reported also on chlorine-induced fish kills in California's Sacramento River. To test the thesis that chlorinated effluent was the culprit, king salmon fry were exposed to river water taken upstream, at the discharge point, 100 feet downstream, and 200 feet downstream. The upstream water caused no adverse effects. Water from the discharge point killed all of the fish in 12 minutes. In less than an hour, all of the fish in the water taken 100 feet downstream from the discharge point were dead and, in less than an hour and a half, all were dead in the 200 feet downstream water. In a companion test, salmon fry were suspended in the Sacramento River. All fish below the outfall were dead within 14 hours while all above survived. Downstream chlorine residuals ranged from 0.2 mg/l to 0.3 mg/l during the test period.

Osborne, et al, (17) studied the effects of chlorinated sewage effluents on fish in the Sheep River, Alberta, Canada. They found no mortality when caged fish were subjected to unchlorinated effluent but 100 percent mortality occurred when exposed to chlorinated effluent. They concluded that chlorination of effluent was the principal factor in fish death. Quantitative sampling of fish populations supported the contention that fish avoid chlorinated effluents.

Giattina, et al, (6) also investigated the avoidance of fish to chlorine at a power plant on the New River in southwestern Virginia. They reported that laboratory determined avoidance concentrations generally predicted the total residual chlorine concentrations that would elicit avoidance behavior under natural field conditions. In general, fish avoid chlorine residuals that are 50 percent or less of the median lethal concentration.

Tsai (21) studied fish life below 149 sewage treatment plants and concluded that turbidity and chlorine caused species diversity reduction below the outfalls. In the upper Patuxent River, (22) chlorinated sewage acts as a toxic material which seriously reduces fish abundance below outfalls, and chlorinated sewage will trigger fish to avoid the outfall water. Chronic physiological responses to chlorine include delayed mortality, depressed activity, decreased growth, and decreased spawning success.

Freshwater reaches of upper Chesapeake Bay are important spawning grounds for many fish species including striped bass. Annual surveys showed that by the end of the Seventies, egg-laden female rockfish still returned to their spawning areas each Spring in great numbers. Eggs were released and found fertilized in the water but few survived to become small fish. It has been shown (12) that chlorine in concentration as low as 0.01 mg/l greatly reduces the percentage of rockfish eggs that are hatched. To compound the problem it has been found (12) that the larvae once hatched continue to be decimated by chlorine. A total residual of only 0.04 mg/l is lethal in one hour to 50 percent of two day old larvae. Chlorine is equally toxic to 30 day old juvenile fish.

Chlorine in the saltwater portion of Chesapeake Bay produces toxic oxidants, chlorine-produced oxidants, from naturally occurring bromine. Eggs and larvae of oysters and clams are very sensitive to chlorine-produced oxidants. Roberts and Gleeson (18) demonstrated that 50 percent of four hour old oyster larvae are killed by only 0.026 mg/l of such oxidants. Rosenburg and co-workers (19) found that chlorine-produced oxidants were lethal to 50 percent of 96 hour old oyster larvae at concentrations of 0.06 mg/l and 16 hour old clams at 0.27 mg/l.

PUBLIC HEALTH JUSTIFICATION

Attention turned to alternatives as evidence began to demonstrate that sewage treatment plants chlorinating their effluent are a major source of toxic pollutants. Alternatives under consideration include: better control of chlorine; detoxification of the effluent; substitution of biocides that produce less toxic residuals; and use of a chemical or radiation that will produce a residual-free effluent. Unfortunately, each alternative has its own set of costly difficulties, and may damage aquatic life. Each may pose some danger to sewage treatment plant operators and perhaps to the surrounding community.

For instance, better control of chlorine application may seem to be a simple inexpensive matter, but it isn't. Much improvement can be obtained by eliminating wasteful, almost promiscuous, misuse of chlorine, but that is not enough. There are very few sewage treatment plants that have been built so that precise control of effluent residual in the part per billion range is possible. To meet an effluent standard that low, drastic changes have to be made in the capability of the sewage treatment plant and in its operation. The orthotolidine color comparitor is useless. Instead, the most precise method of analytical measurement must be used. Automatic chlorine residual monitoring and feedback control units are necessary. Only four percent of the sewage treatment plants that were surveyed (7) have feedback control. In contrast, 60 percent use a manual method to feed chlorine.

Before blindly accepting the proposition that there is a need to find a substitute for chlorine, the possibility that disinfection of sewage effluent is not necessary in most cases should be examined. The public health necessity of disinfecting sewage effluent under ordinary circumstances must be justified for the practice to continue in any form.

Some disagree (11) claiming that: "The cornerstone of public health is preventive medicine and to require the justification for wastewater disinfection is a giant step backward." The fault in that assertion is that the alleged "public health" and "preventive medicine" benefits of effluent chlorination are what need to be justified. As for requiring justification, the health of the human race was improved dramatically as soon as public health practitioners were required to justify their strongly held beliefs.

There is an assumption that the act of chlorinating sewage will decrease the danger of disease, but for all practical purposes, that assumption is not valid. Food or water contaminated with sewage will cause disease and remains dangerous whether it is chlorinated or not. After a decade of nationwide chlorination of sewage, there is no evidence to demonstrate that the incidence of any illness has decreased as a result of that practice. The United States chlorinates its sewage - England doesn't. There is no credible evidence to show that any related illness occurs more frequently in England than it does in the United States.

The U.S. Public Health Service with its Center for Disease Control in Atlanta, Georgia, is the world's outstanding authority on the causes of disease and how to prevent them. The Comptroller General reported to Congress (4) that "The Center for Disease Control has taken the official position that disinfection of sewage provides little public health benefits". In correspondence, G. F. Mallison of the Bacterial Diseases Division of the Center for Disease Control, wrote "I see, with rare exceptions, absolutely no need with respect to health in attempting to control microbial contamination after secondary sewage treatment".

Health Hazard to Workers

An examination of the health effects of chlorinating sewage might start with its effect on sewerage workers. In the debate over the public health benefit or lack thereof that comes from chlorinating effluent, the health of the sewage treatment plant operator is largely ignored. That is a mistake because chlorine creates an occupational hazard and there have been a significant number of incapacitating accidents. Chlorine in the air is almost as toxic to humans as chlorine in the water is to aquatic life. A concentration of 0.1 percent of chlorine in the air is likely to be fatal after a few breaths and almost certain to cause death within ten minutes. A safe allowable concentration of one part per million has been established by the Occupational Safety and Health Administration.

In a survey (7) conducted and reported by the Water Pollution Control Federation in 1980, it was found that over 11 percent of the sewage treatment plants surveyed reported chlorine accidents in which people required medical treatment.

Debate Over Recreation Water

Protection of the health of people using water for recreation is a frequently used justification for sewage chlorination even though epidemiological evidence of its value in that regard is nonexistent. In fact, no study has examined the proposition that recreation waters shown to cause disease can be made safe by chlorinating sewage effluent. Instead, the effort to date has been to demonstrate, if indeed it is possible to demonstrate, that swimming in polluted water causes a higher incidence of disease

and, if so. to find an indicator bacterium that correlates with risk. For thirty years the aim has been to establish a number for a particular indicator organism that will give assurance against disease contracted from swimming in sewage polluted water.

That there is a safe threshold of pollution for swimming, and that such a threshold can be identified through an allowable number of easily measured indicator bacteria, is a strongly held belief, but it is not shared by all. Stevenson (2) pioneered studies in Lake Michigan and the Ohio River. Though the studies were far from conclusive, he arrived at a concentration of total coliform bacteria as the best practical standard. Geldreich (5) related Salmonella detection to fecal coliform densities and recommended a standard based on fecal coliform detection. Cabelli (2) found an increase in gastrointestinal disturbances among those swimmers who immersed their heads in water. Based on a correlation with fecal enterococci , a mathematical expression of the risk of increased incidence of disease was developed.

A higher incidence of disease caused by swimming in polluted waters is not a universal finding. The National Technical Advisory Committee found Public Health Service studies on which the coliform standards are based to be far from definitive. They expressed an urgent need to find if there is a correlation between the various indicator organisms and disease attributable to water recreation. In Sydney, Australia, many years of epidemiological study in connection with Sydney's world famous bathing beaches produced no evidence of water-borne diseases caused by unchlorinated sewage effluent.

In the United Kingdom, a committee which Moore (14) headed did research for six years in the 1950's and failed to establish any significant bacterial hazard from sea bathing. Later work by the Water Pollution Research Laboratory also failed to find a satisfactory method for establishing bacterial standards for bathing waters. It is Moore's contention that no shred of evidence has been produced in Europe during the past 20-30 years that indicates that human health has been endangered in the absence of bathing water standards.

From a realistic public health perspective, the incidence of sewage pollution related diseases contracted through recreational use of water is trivial. Competent persons have searched for such a relationship. Some claim that it does exist and others find that it does not. Even if it does exist, the effort required to ferret out the relationship is strong testimony that swimming in polluted waters accounts for a miniscule fraction of the total incidence of serious disease. Most of the minor irritations that do occur are of the eye, ear, nose, and skin variety, making it likely that transmission is person to person and not sewage to person. It is highly unlikely that an enteric disease indicator bacterium will ever be found that correlates with those ailments.

Even if a sewage treatment plant discharge to swimming water disease relationship does exist, effluent chlorination would be the wrong thing to do. In fact, health receives better protection if sewage effluent is not chlorinated. Chlorination of ordinary sewage treatment plant effluent kills more of any of the various indicator bacteria than it does of the virus in sewage effluent, and virus as well as other chlorine resistant organisms are the main cause of concern. That being the case, chlorination of sewage effluent diminishes the indicators of pollution in relation to the prevalence of the real danger, thus, creating a false sense of security. A safer course of action is to provide better sewage treatment and greater separation between outfalls and bathing beaches.

Shellfish

Like bathing beaches, chlorinating effluent gives the illusion of public health protection, but the real protection of shellfish growing waters is provided by good sewage treatment and safe separation between outfalls and shellfish beds. Consumption of raw oysters harvested from sewage polluted waters caused a high incidence of disease prior to the shellfish sanitation program initiated by the U.S. Public Health Service in the late 20's. Since the time that the program became effective, not one case of illness has been traced to oysters harvested from approved waters in Maryland.

The principal elements of this effective program are separation between pollution discharge and shellfish harvesting beds coupled with a bacteriological standard applied at the place of harvest. The bacterial standard for shellfish harvest water was derived from empirical observations at a time when the discharge of untreated sewage was commonplace and many people became ill from eating oysters taken from polluted water. Unlike recreational waters, it was clearly demonstrated that when people ate oysters taken from polluted water with an indicator bacterial density higher than the standard, they got sick. When they ate oysters from waters cleaner than that indicated by the standard, they did not get sick.

The shellfish harvesting bacterial standard works because of the general relationship that exists between the density of indicator bacteria and the density of disease agents. Chlorination of ordinary sewage treatment plant effluent alters the indicator/disease producing organism ratio in a dangerous fashion. It is disconcerting that virus can persist even after indicator bacterial organisms have been killed, because shellfish contamination by virus has replaced bacteria as the disease agent of major concern.

Olivieri, et al, produced data that strongly supports the hypothesis that free chlorine is required for significant viral reductions (16). Free chlorine for the required contact time calls for break-point chlorination, rapid mixing, and precise hydraulic control, things that are rarely achieved in conventional sewage treatment plant operation.

Recognizing that chlorine can disrupt the traditional indicator-pathogen ratio, Bisson and Cabelli (1) have looked for alternatives. They have examined the feasibility of using a spore former, Clostridium perfringens, as an indicator for the potential for infectious disease from fecal pollution because the spores of C. perfringens are much more resistant to chlorination that E.coli. For specific applications against the potential for infectious disease arising from fecal pollution of the aquatic environment, they suggest that there is no universal microbial indicator.

Destruction of the Natural Barrier

The argument is sometimes advanced that chlorination of ordinary sewage treatment plant effluent provides another barrier in a multiple barrier concept of public health protection. The strategy is to provide as many barriers between a source of disease organisms and the public as opportunity and cost will permit. The idea is sound but chlorination of sewage treatment plant effluent does not impose a dependable barrier. Instead, it destroys one of the most effective barriers in existence. That barrier is nature's relentless antagonism to the disease producing bacteria and virus found in sewage.

Mitchell (13) studied the destruction of sewage bacteria and virus that were discharged into seawater. He found that enteric bacteria are destroyed by a specific antagonistic microflora that develops. Mitchell was able to classify three groups of native seawater organisms associated with the accomplishment of this destruction: native bacteria that destroy by enzymatically lysing enteric bacteria cell walls; obligatory parasitic bacteria; and, amebae which attack and consume bacterial cells. Of these, the amebae are the most active. With respect to virus, native marine microflora are involved in a manner similar to that observed with enteric bacteria but a chemical component of seawater was also shown to be involved in the virus destruction.

The specialized culture that develops in biological sewage treatment processes exhibits similar antagonism to disease producing organisms. Unfortunately, chlorination of sewage effluent kills the predators as well as the prey. The culture of specialized organisms that started their attack on sewage-borne pathogens within the sewage treatment plant are disrupted and the disruption carries over to the organisms of natural purification in the receiving waters. Walsh and Mitchell (23) found that chlorination of effluent produced hydrocarbons which can cause damage to the natural predators responsible for self purification in the vicinty of sewage outfalls.

In most situations the barrier imposed by nature's system is far more important to the protection of shellfish beds than the superficial protection gained by the mere reduction of indicator bacteria that occurs when chlorine is added to ordinary sewage treatment plant effluent.

Disinfection

Contrary to repetitive misuse of the word in water pollution control literature, the conventional practice of chlorination at sewage treatment plants does not produce a disinfected effluent. The term "disinfection" is used to describe a process that removes all organisms capable of producing a disease. In every other field of endeavor, including milk, food, drinking water, and hospital care, "disinfection" has that meaning. It does not imply sterilization where all forms of life are destroyed, but it does mean that a disinfected material will no longer produce infectious diseases.

Water pollution control workers are quick to point out that in the general case, they don't mean that kind of disinfection when they use the word. No matter what the professional means, it is what administrators, the press, and the informed public believe that counts. The public wrongly perceives that chlorinated sewage is disinfected because water pollution control workers continually tell them that it is.

No knowledgeable person would contend that chlorination of ordinary sewage treatment plant effluent would render it disinfected, incapable of producing disease. The reverse is true; chlorinated sewage treatment plant effluents are highly infectious and should be treated with appropriate caution. The use of the word, disinfection, is in itself dangerous in this situation because it promotes a false sense of security and that could lead to relaxation of the basic principles of sanitation that are, after all, the main bulwark of public health protection.

It is well established that stringent conditions must be met before chlorine or any chemical that acts in a related fashion can disinfect. Those conditions include the removal of essentially all suspended solids, turbidity, and interfering substances including BOD. Sewage effluent requires filtering and break-point chlorination to produce on the order of 1.0 mg/l of hypochlorous acid (HOCL) for 30 minutes to achieve disinfection. Chlorine must be completely and uniformly mixed as rapidly as possible. Careful engineering of a holding and contact chamber is a necessity. Morris (15) has pointed out that any measurable degree of short circuiting is ruinous. Only 0.01 percent of raw fluid may cause the water to fall below hygienic standards.

Obviously disinfection is not accomplished when chlorine is added to the solids laden, organic rich effluent from an ordinary secondary sewage treatment plant. Only in a very few instances where sewage is being conditioned for direct reuse in specifically designed and operated purification works is true disinfection practiced.

Chlorinated Hydrocarbons

While some persons within the U.S. Environmental Protection Agency

continue to support the chlorination of effluent as the best practicable measure, others in the agency are calling attention to the possible public health problem that chlorination of sewage effluent is creating. In a statement on the effects of chlorine on Chesapeake Bay organisms, the EPA pointed out that recently an unforeseen chlorine problem surfaced. Chlorine introduced into sewage effluent can form a large variety of daughter compounds of concern to drinking water supplies. Hunter and Sabatino (8) searched out the sources of halogenated hydrocarbons in an urban water supply from the Passaic River in New Jersey. The project which covered only the usual identifiable chlorinated compounds indicated that during the summer, chlorination practices account for the predominant volatile halogenated hydrocarbons observed.

DISCUSSION

It should come as no surprise that the chlorine in sewage effluent is killing valuable aquatic life. Chlorine has been used for seventy years to kill a wide variety of unwanted aquatic organisms. Pollution control experts use chlorine to kill bacteria in wastewater, to kill fouling organisms in cooling water, in fact, to kill many things for many reasons.

When sewage treatment plant effluent is chlorinated, the killing effect continues to be exerted on a host of organisms in the aquatic environment. The effects fall into three categories: toxicity to fish and other mature forms of life; fish avoidance of chlorinated effluent; and, destruction of larvae and other first emergent forms of aquatic life.

Fish kills are likely to occur where there is an excessive use of chlorine. Fish kills are spectacular and receive immediate attention in the form of field surveys and bioassays. But even though they go largely unnoticed, the deadly subtle effects on fish migration and reproduction are far more devastating to many forms of aquatic life. Unlike fish kills, the disruption of the reproductive process is unseen, but it is of fundamental importance because it strikes at the ability of a species to sustain itself through seasonal reproduction.

In the Maryland portion of Chesapeake Bay, there are more than a hundred sewage treatment plants that discharge into tributary streams where fish come to spawn. The discharge from a single sewage treatment plant is often a sizable fraction of the total stream flow and many tributaries have multiple points of discharge. Because spawning fish retreat from the slightest trace of chlorinated effluent, chlorination creates an impenetrable barrier that prevents the fish from reaching their spawning grounds. Should fish be able to find a place to spawn in a stream below a sewage treatment plant outfall, the killing effect of chlorine first on the eggs, then on the larvae, and then on the immature fish makes survival to adulthood very unlikely.

Oysters and clams have been shown to be susceptible to very low levels

of chlorine produced oxidants. As with fish, damage to oysters and clams is far greater to the first emergent forms of life during reproduction than it is to the adult.

To offset the damage being done to the aquatic environment, there would need to be an overriding public health benefit derived from the widespread chlorination of sewage treatment plant effluent. Instead of benefiting public health, chlorination of effluent produces unwanted chlorinated hydrocarbons, creates a hazard to sewerage workers, could create a hazard at bathing beaches, gives a false signal at shellfish harvesting grounds, destroys a natural barrier to transmission of disease, and fails completely to disinfect ordinary effluent.

Chlorination of ordinary sewage treatment effluent provides no appreciable public health benefit to offset the major damage that it causes. No other industry would be allowed to discharge a toxic pollutant capable of causing damage like that of chlorinated effluent. The practice should be stopped.

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2. PATHOGENS? IN SEWAGE?!

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ABSTRACT

Domestic sewage is an infectious material carrying human pathogens shed in the fecal discharges of infected individuals. Sewage effluents may affect shellfish growing areas, sources of domestic water supply, recreational waters, the ultimate users of reclaimed sewage and others. For most of these disinfection is necessary to prevent infectious disease transmission. Chlorine is almost universally used as a disinfectant for sewage. As to the suitability of two other disinfectants, ozone and ultraviolet light, questions must be answered concerning effectiveness, energy and dollar costs, and practicability. The need for disinfection to meet bathing water quality standards is more extensively discussed. Sewage discharged to recreational waters in significant concentrations will cause disease. How much disease, and what are significant concentrations, what are infectious doses, what is the best indicator organism, and what standards are to be used, are discussed. It is concluded that fecal coliform is the best available indicator organism, though not entirely satisfactory, but that sewage effluents must be disinfected to protect users of recreational waters.

INTRODUCTION

This will not be an exhaustive discussion of the need for sewage disinfection. A brief commentary should suffice. Domestic sewage is an infectious material - carrying human pathogens shed in the fecal discharges of infected individuals. The concentration of the pathogens depends largely upon the extent of infection in the tributary population. At the turn of the century domestic sewage may still have carried some cholera organisms and certainly carried significant numbers of typhoid organisms. Cholera is long gone today and typhoid organisms must be at an exceedingly low level. Does this mean that sewage is no longer hazardous? Not so. It is certain that sewage in this ninth decade of the century is infectious, containing bacterial pathogens, enteroviruses and parasites. Can there be any question, therefore, about the necessity for disinfecting sewage?

DISCUSSION

In order to assess the consequence of not disinfecting sewage let us consider the various modes of disposal of sewage effluents - to oceans and

estuarine waters, to streams and lakes, to land, and for planned sewage reuse projects. Discharged effluents may affect shellfish growing areas, sources of domestic water supply, waters used for food crop irrigation, recreational waters, salt or fresh, or may be reused in planned projects for an array of purposes. These include golf course - urban landscape, and crop irrigation, recreational lakes, process and cooling water for industry, groundwater recharge and others.

There is no question that undisinfected sewage effluents reaching shellfish growing areas in any substantial concentration will infect shellfish and that these shellfish will transmit disease. Through the first half of this century many outbreaks of typhoid fever and paratyphoid fever and cases of illness have been reported as shellfish associated diseases. Since the 1950's when raw shellfish was first shown to be a route of transmission for infectious hepatitis there have been about 17 outbreaks involving some 1339 persons in the U.S. (19). In a recent outbreak of some 268 cases, the oysters were traced to beds in Louisiana which earlier in the year had been closed due to pollution associated with sewage polluted waters (20).

There is no doubt that large doses of undisinfected sewage may overwhelm water treatment facilities, in extreme cases even those with filtration facilities. The hepatitis epidemic in New Delhi in 1955-1956 is evidence of that (3). Moreover, good practice dictates that for an adequate level of protection of domestic water supply, multiple factors of safety must be provided. Thus, effective disinfection of sewage discharges upstream from domestic water supply intakes is essential. There can be no doubt, either, that for most types of planned sewage reuse such as those cited above, a high degree of treatment including reliable and effective disinfection is essential to meet the appropriate water quality standards in order to prevent disease transmission (5).

Sewage chlorination also has been practiced for many years to delay bacterial action and to stretch out in time and distance the impact of BOD in receiving waters - to modulate the oxygen sag curve. The most recently recognized reason for sewage disinfection relates to identification of "R" factors in bacteria. These factors are nucleic acid elements in bacteria causing resistance to antimicrobial drugs. Coliforms may act as reservoirs of "R" factors and transfer them to pathogens. There is evidence that sewage polluted water may play a role in spread of coliform and other bacteria carrying "R" factors. This supports providing effective disinfection of sewage effluents (11).

There are of course, some situations where sewage disinfection may not be necessary. Land-disposal projects and planned groudwater recharge projects generally may be carried out without disinfection - though there may be some situation where disinfection is warranted. Long-outfall ocean discharge, particularly to deep-water sites may be accomplished without disinfection, if the effluent does not significantly degrade adjacent recreational waters. Determination of "significant" degradation depends upon guiding standards and will be discussed in some detail.

In a related situation, the recognition that chlorination results in creation of trihalomethanes (THMs) at some domestic water treatment facilities, lead to questions about continuing the use of chlorine for this treatment. Response of public health authorities to this question has been unequivical; in the balance the value of chlorination for public health protection far exceeds the possible adverse effects of THMs, and there is no comparable substitute. The same may be concluded for sewage treatment practice, particularly where discharge is to groundwater basins as discussed below. The central point of the forgoing is that for most applications sewage discharges must be disinfected for a satisfactory level of public health protection.

Now, a brief commentary on the question, should chlorine be used as the disinfectant? This question arises primarily because the addition of chlorine for sewage disinfection produces some biotoxicity in effluents (14,11). Biotoxicity has two different kinds of impact depending upon whether effluents are discharged to surface waters or to groundwaters. Regarding surface water the concern is about impact on fish ecology. With groundwater the potential impact is a health effect. This biotoxicity increases with increased levels of chlorine addition. Recent work by the Sanitary Engineering Section of the California Department of Health Services indicates that these biotoxic effects can be minimized by well engineered sewage chlorination facilities - rapid mix of the chlorine and plug flow contact basins. Also SO_2 dechlorination following chlorination will remove from effluents chlorine-induced biotoxicity to aquatic life (22). If the choice must be between no disinfection or chlorination, chlorination it must be.

As to the suitability of other means of disinfection, ozone or ultraviolet light, questions must be answered concerning effectiveness, energy and dollar costs, practicability, and perhaps others. Papers scheduled for later in this conference should provide some insights into these issues. My prediction is that chlorination of sewage effluents will be practiced as the primary method of disinfection for many more years.

Considering the scope of this conference the subject of recreational waters requires more extensive discussion. It is emphasized, however, that even for this category of affected environment there is no doubt that disinfection is necessary for protection of bathers. The issue instead is about the nature and extent of illness attributable to recreation in contaminated waters, which organism or organisms should be used for monitoring, the correlation of these two factors, what should be limiting values for an acceptable level of risk, and even whether or not any microbiological limits should be set.

Authoritative leadership concerning public health control of natural bathing places (as contrasted to artifical swimming pools) has been provided by two series of publications. The first of these were a set of ten reports from 1926 to 1957 of the "Joint Committee on Bathing Places" of the Conference of State Sanitary Engineers and the Engineering and Sanitation Section of the American Public Health Association. The tenth Edition - 1957 (12) reports

efforts in 1921, 1939 and 1955 to secure authentic information on reported cases of illness attributable to bathing places. Regarding the 1955 inquiry the committee states "It is striking that the returns from 45 states and one territory stated that they could report no authoritative cases of illness attributable to swimming pools and bathing places... Until new developments take place to warrant different conclusions, the summary of replies to earlier questionnaires and the recent survey of data obtained from state health departments, considered in the light of known epidemiological evidence, leaves this committee unconvinced that bathing places are a major public health problem even though bathing place sanitation because of the health considerations involved should be under careful surveillance of the public health authorities and proper sanitary control of bathing places should be exercised ... It is realized that new epidemiologic evidence may come forth in the future. It is agreed that common sense public health programs must recognize that bathing in polluted water is a potential danger, that unsanitary conditions surrounding public bathing places are a hazard, and that common decency as well as health considerations dictates that reasonable steps should be taken to secure bathing in clean environments..."

This report notes there is a wide divergence of opinion as to standards of acceptable bacteriologic quality for outdoor bathing places in streams, rivers, lakes and tidal waters. It emphasizes that final classification of bathing waters should depend largely upon sanitary survey information, and that bacteriologic analyses should be used as a guide. Further, pollution may be present in many waters where treatment of sewage removes visable evidence of sewage but does not eliminate dangerous concentrations of bacteria.

The second series of reports are three documents developed under Federal auspices. These three documents have served as a basis for water quality standards for the Federal regulatory water pollution control program: Water Quality Criteria - 1968 (17), Water Quality Criteria - 1972 (7), and Quality Criteria For Water - 1976 (24).

Selected excerpts from the 1968 Report are as follows: "The establishment of public health requirements for the protection of the primary contact recreation users has been a major problem for the sub-committee. Moreover, in recommending specific water quality criteria the sub-committee is faced with a sharp dilemma - that of balancing reasonable safeguards for the public health against possible undue restrictions on the availability of waters for contact recreation. The problem is further complicated by the inadequacy of studies correlating epidemiological data on waterborne diseases with degrees of pollution in recreational waters... There is an urgent need for research to refine correlations of various indicator organisms including fecal coliforms to waterborne disease. The sub-committee feels that the Public Health Service's three epidemiological studies on bathing water quality and health are the only base available for setting criteria. These studies were far from definitive and were conducted before the acceptance of the fecal coliform as a more realistic measure of health hazard... The sub-committee recognizes that localized bacterial standards may be justified, if based on sufficient experience, sanitary surveys, or other control in monitoring

systems..." In conclusion this report recommended that "...the fecal coliform content of primary contact waters shall not exceed a log mean of 200/100 ml, nor shall more than 10 percent of total samples during any 30-day period exceed 400/100 ml."

Selected excerpts from the 1972 Report are as follows: "... All recreational waters should be sufficiently free of pathogenic bacteria so as not to pose hazards to health through infection. This is a particularly important requirement for planned bathing in swimming areas. There have been several attempts to determine specific hazards to health from swimming in sewage contaminated water. Three related studies have been conducted in this country demonstrating that an appreciably higher overall illness incidence may be expected among swimmers than among nonswimmers (24). In evaluating microbiological indicators of recreational water quality it should be remembered that many of the diseases that seem to be causally related to swimming and bathing in polluted water are not enteric diseases or are not caused by enteric organisms. Hence, the presence of fecal coliform bacteria in recreational waters is less meaningful than in drinking water... When used to supplement other evaluative measurements the fecal coliform index may be of value in determining the sanitary quality of recreational water intended for bathing and swimming. The index is a measure of the sanitary cleanliness of the water and may denote the possible presence of untreated or inadequately treated human waste but it is an index that should be used only in conjunction with other evaluative parameters of water quality such as sanitary surveys..." In conclusion this Report states "No specific recommendation is made concerning the presence or concentrations of microorganisms in bathing water because of the paucity of valid epidemiological data."

The 1976 Report includes the following: "...Pollution of aquatic systems by the excreta of warm blooded animals creates public health problems for man and animals... The number of fecal coliforms present is indicative of the degree of health risks associated with using the water for drinking, swimming, or shellfish harvesting. Arguments against the use of fecal coliform bacteria to define swimming quality in water have noted the paucity of epidemiological evidence linking fecal coliform levels in bathing waters and the incidence of disease (15,16). The lack of epidemiological correlation between fecal coliform levels in coastal swimming waters and the incidence of disease may not have validity in fresh waters and it does not take into account non-reported diseases that may develop as an unrecognized result of swimming in polluted waters. Epidemiological evidence is but one consideration in setting microbiological criteria. The presence of fecal coliform bacteria indicates degradation of water quality and a relative risk of disease transmission." In conclusion, this Report states that evaluation of microbiological suitability of marine and fresh waters should be based on fecal coliform levels, and reiterates the 200/400 fecal coliform limit recommended in the 1968 Report.

In the aggregate these two series of documents express the need for protecting water quality in natural bathing places, support the use of a coliform-fecal coliform index of water quality, refer to four epidemiological

studies, note the paucity of evidence linking fecal coliform levels in bathing water and the incidence of bathing-associated disease, and indicate the need for more definitive epidemiological data. A careful review of the investigations of Moore (13,14) and those of the Public Health Service (23) lead to a conclusion that these epidemiological studies are flawed in procedural methodology and the resulting conclusions have limited significance. A much sounder but still limited epidemiological investigation has now been made by Cabelli and Associates (2,3). This work represents a three-year (1973-75) study of epidemiological-microbiological study conducted at New York City beaches as part of the U.S. Environmental Protection Agency (EPA) program to develop health effects-recreational water quality criteria. Symptomatology rates among swimmers relative to non-swimming but beach-going controls at a barely acceptable beach and a relatively unpolluted beach were examined. It was observed that the symptom rates categorized as gastrointestinal, respiratory, "other" and "disabling" were higher among swimmers than non-swimmers. The rate of G.I. symptoms was significantly higher among swimmers relative to non-swimmers at the barely acceptable but not the relatively unpolluted beach. I assume Cabelli will give us more details on this work later today.

The EPA has also sponsored two other investigations into recreational water quality and health. The first of these, a two-year study in 1976 and 1977, measured health effects of swimming in marine recreational waters in a subtropical climate at Lake Pontchartrain at New Orleans (13). In the preliminary first-year study, for children under age 10, a significant positive association was found between gastrointestinal symptom rates and exposure to E. coli levels above 200 per 100 ml. For 10 years and older persons, evidence of a relationship between symptoms and enterococcus density was strongly suggested. The second-year study partly substantiated the first-year results in that illness rates were again higher for swimmers than non-swimmers and highest for young swimmers. No association, however, was found between either microbial indicator and symptom rates for children. For the older group, there appeared to be a relationship between Escherichia coli and enterococcus levels with symptom rates. The second investigation (18) was a pilot study to develop and evaluate methods to determine the effect of recreational water quality on the health of persons bathing in fresh waters. This work has been carried on in Lake Erie at Cleveland, Ohio

The subject of the health aspects of recreational water quality was reviewed and summarized in detail at a workshop at Drexel University in 1978 (19). This workshop notes that the most commonly reported swimming-associated infections are those of the skin, eyes, ears, and upper respiratory tract. In contrast, reported outbreaks of recreational gastrointestinal disease have been rare - only a small group in this century. The largest and best documented outbreak of typhoid attributable to swimming occurred in Australia (9). Ten cases were associated with swimming at a beach contaminated by sewage from a broken outfall. Only one outbreak of hepatitis has been associated with recreational waters (1); however, drinking rather than swimming was the more likely route of transmission. The best documented outbreak associated with swimming was one in which 31 of 45 cases of bacillary dysentery were traced to swimming in a five-mile stretch of the

Mississippi River below Dubuque, Iowa (21). The workshop proceedings conclude with the following comments: "... Swimming per se carries with it an increased risk of infections and irritations of the skin, ears, nose, and upper respiratory tract. This risk appears to be infrequently associated with pollution of the bathing waters with human or animal fecal waste... Except under conditions of heavy contamination with human waste or during epidemic conditions among the population whose waste reach the bathing waters, the risk of contracting any of the severe, well recognized, well defined enteric diseases such as salmonellosis, infectious hepatitis, poliomyelitis, typhoid fever, etc. is minimal. Sporatic swimming-associated cases of these diseases possibly do occur. Even with moderately polluted waters, there is a significant risk of contracting a gastroenteritis which appears to be acute in its onset but benign in its course..."

The most recent report of significance is that of a National Research Council Committee (6) which deals solely with the subject of microbiological measures of recreational water quality. In summary, this Report states, "In essence, the current recreational water quality criterion is an indicator system for water that is contaminated by the feces of warm blooded animals. It is helpful only in the prediction of health hazards of recreational water where the fecal-oral route of transmission is involved. Fecal coliform tests detect mostly E. coli, which is not consistently pathogenic..." This Report notes also that fecal coliform tests detect organisms such as Klebsiella, Enterobacter, and Citrobacter, whose precise health significance remains to be resolved. Further, the fecal coliform test appears to be of little, if any, significance in the control of the many external ear, eye, and skin infections that can be traced to contact with contaminated water... The fecal coliform test is a reasonable indicator system for Shigella spp., Salmonella typhosa, S. typhimurium, E. coli, and other unidentified agents of the varied gastrointestinal symptomotology that appear to be associated with ingestion of swimming water. In a less direct manner, fecal coliforms may indicate the presence of viruses that could be transmitted by the fecal-oral route. There is evidence, however, that viruses may occur where fecal coliform counts are low or are not detectable.

Further this Report states, "One important epidemiological factor in fecal-oral transmission, which has not been adequately addressed is the volume of water unintentionally taken into the digestive tract by a swimmer or an individual that had been immersed in water at recreational sites. The volume of ingested water must be important in determining the numbers or dose of a pathogenic agent to which an individual has been exposed. The intake may vary with the individual's age, level of swimming proficiency, time of exposure, quality or salinity of water, etc. These variables have not yet been measured and it is unknown how they relate to the threshold dose for enteric infection ... The fecal coliform criterion remains a reasonable predictor for gastroenteric illness and possibly infections from non-coliform agents. Evidence indicates an increased risk of gastrointestinal illness when the fecal coliform criterion is exceeded."

"No criterion or guideline that is based on a single microbial indicator species will serve as a measure of the health risk from the wide variety of diseases that can be contracted from recreational water. For any indicator used, however, sampling and laboratory testing must proceed in conjunction with epidemiologic surveillance, public health engineering, sanitary surveys, and monitoring..." "Absolute protection of the public health by relying on a single water quality criterion is not feasible and, in fact, is not possible." This Committee concludes that "The fecal coliform test is acceptable for protecting the public health until additional epidemiologic data, improved laboratory procedures, and a better understanding of aquatic microbial ecology are obtained... Its use certainly is better than abandoning microbiological criteria altogether. The Committee recommends that the fecal coliform tests be replaced eventually by a test or series of tests that directly assess the health hazard posed by the presence of pathogens in recreational waters."

To recapitulate, sewage effluents discharged to recreational waters in significant concentrations will cause disease. How much disease, what are significant concentrations, and what are infectious doses, are not definitely established. A search continues for more suitable parameters. Until these are established, the fecal coliform test is considered the best available. Finally, on the basis of fecal coliforms, what should be limiting values? The answer to this last question depends upon a value judgment relating to acceptable risk. This value judgment cannot intelligently be made at this time because the risk cannot be measured with sufficient accuracy.

Epidemiology is the tool that is used, essentially the only one available. Unfortunately it is a blunt instrument.

Two things seem certain. One, very little epidemic disease has been associated with swimming in sewage polluted waters in this country, only acute disease has been detected by the retrospective epidemiology, and even then, only where the pollution has been gross. Two, disease unquestionably results from swimming in polluted water, not only gastrointestinal illness, but infections of the skin, eyes, ears, and upper respiratory tract; but apparently not at epidemic levels. Much of this disease is subacute, is not seen by physicians, and can best (perhaps only) be measured by prospective epidemiology. Cabelli (4), in commenting on this phenomenon states, "It would appear that data derived from published care and outbreak reports markedly understate the rates of recreational waterborne disease." It is also likely that some of the non-G.I. illness is more directly associated with the act of swimming than from pollution in the water.

Considering the fact that this subject has been discussed in public health circles for over 60 years, astonishingly little progress has been made in establishing a sound basis for standards. On the other hand, the crudely developed "standards" in use are probably not too far from the mark. They have a sort of common sense ring to them. Furthermore, when one considers the billions of construction grant dollars spent annually, some to meet

arbitrary requirements for secondary treatment, and the big-ticket cost of operating the facilities, it is tempting to wonder why so much issue is taken with the 200/400 coliform limits. The cost for meeting these limits is "peanuts" compared with the rest of the bill, and it buys a certain, though unquantifiable, amount of public health protection.

In conclusion, do disinfect sewage effluents to protect recreational water.

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3. INFECTIVE DOSE OF WATERBORNE PATHOGENS

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ABSTRACT

Infective dose studies with a variety of enteric organisms have been conducted over the past 30 years in human volunteers. The widest dose range required to produce a response was found with the bacterial agents. Salmonella spp. required the largest dose with the ingestion of 10^5 to 10^8 cells needed to produce a 50 percent attack rate. In contrast, three species of Shigella produced illness in a significant percent of dosed subjects with 10 to 100 cells. Protozoan infections have been produced with Entamoeba coli and Giardia lamblia dosed in gelatin capsules at the level of 1 to 10 cysts. Enteric viruses have produced infection at low dosage levels via oral ingestion, inhalation, and conjunctival exposure. These data produced with healthy human subjects show that members of all three categories of enteric pathogens can produce infection and/or illness at concentrations found in wastewater.

INTRODUCTION

The return to the soil of chemical nutrients and moisture existing in wastewater for more productive cultivation of desirable plant life is an ancient custom that still seems appropriate for our time. Wastewater that has received little or no treatment has the advantage of less cost and greater nutrient value, but it also has a greater number of pathogens and therein lies a health concern.

A wide variety of enteric pathogens including viruses, bacteria and parasites is known to occur in all community-derived wastewaters. The concentration of pathogens, especially viruses, is debated primarily due to limitations of currently available recovery techniques. However, the existence of any concentration of pathogens in the environment does not necessarily pose a health hazard. A mechanism of transmission back to man must exist and in order to warrant exposure-control expenditures, e.g., disinfection, the occurrence must be more likely than a rare event.

Three modes of transmission are perceived as the most likely potential routes for reintroducing wastewater pathogens to man: (a) direct exposure to wastewater aerosols, (b) ingestion of drinking water contaminated with wastewater seepage or runoff, and (c) ingestion of contaminated animal or plant foods produced on wastewater-amended soil. The hazard evaluation of these potential exposure routes may be approached basically in two ways: (a) by

epidemiological studies of exposed populations and (b) by a modeling approach which seeks to determine the likelihood of infection by considering a number of human and environmental variables. Considerable effort has been applied to the former approach and some of this work will be discussed in detail in other papers in these Proceedings.

A major variable important to utilization of the second approach is the infective dose of an agent. Information must be available on infection rates in populations exposed to diminishing numbers of a specific microorganism. The often used phrase "minimum infective dose" is a misnomer in that it does not acknowledge the concept of a changing probability of infection with exposure to varying dosage levels. For infective dose data to have meaning in this context, subjects must be exposed to multiple concentrations of an agent so that dose response curves may be developed. Several studies of this type have been conducted with animal models and human volunteers using a variety of enteric microorganisms. This review of the data will be limited to the human volunteer studies since they will pose fewer interpretative questions and thereby be of more practical significance to the subject of this symposium.

In considering host responses to microorganisms, a distinction is to be made between the two most common end points measured: infection and illness. Infection may be defined as multiplication of a microbial agent within a host with or without the production of disease. The occurrence of illness may be determined by the manifestation of a single pathogenic effect or a group of symptoms normally associated with an etiological agent. Both end points have been utilized to study host-parasite interactions in human populations. However, investigators, institutions, and the public in general are becoming increasingly reluctant to support studies designed to produce pathogenic effects in humans. Therefore, the more recent data, obtained with viruses, have determined the asymptomatic infectivity end point as indicated by fecal shedding of the test organism or by the detection of a specific antibody response. Of course, the asymptomatic infective dose of a pathogen for one individual may produce disease in another. However, it is generally assumed that severity of response is proportional to the degree of exposure and in feeding studies that administer relatively low doses, to determine the minimum infectivity level, adverse effects in healthy subjects are rare. Nonetheless, infective dose is an important parameter in hazard evaluations because infected persons may transmit viable organisms to others who may experience clinical disease. As added precautions, virus studies are normally conducted with vaccine or very mildly-pathogenic strains and in studies with bacteria or protozoa the termination of infection can usually be insured by the administration of antibiotics or other antimicrobial drugs.

DOSE RESPONSE TO ENTERIC BACTERIA

The organisms of greatest concern in exposure to wastewater-contaminated environments are the enteric bacteria and viruses and the intestinal parasites. In the United States, Salmonella and Shigella species are essentially the only enteric bacteria that have a recognized prevalence level sufficient to be of concern. Hornick et al. (9) have conducted dose-response studies in healthy adult male volunteers with Salmonella typhi, the etiological agent of typhoid fever. This study determined the number of organisms required to produce an illness end point. A positive response was determined when toxic symptoms of typhoid fever occurred, e.g., headache, malaise, anorexia, and temperature of 103°F for 24 to 36 hours. At this point, the infection was interrupted by the administration of an antibiotic. Table 1 shows the dose response obtained with the oral administration of various densities of organisms suspended in 30 ml of milk. No symptoms of typhoid fever were observed in 14 volunteers who received 10^3 organisms. Half of the volunteers who ingested 10^7 organisms became ill.

Table 1. Number of Salmonella typhi (Quailes Strain) Organisms Required to Produce Typhoid Fever in Healthy Adult Male Volunteers (9)

Number of Viable Organisms Administered*	Dose Response No. Ill/No. Challenged (%)
10^3	0/14 (0)
10^5	32/116 (28)
10^7	16/32 (50)
10^9	40/42 (95)

*Organisms suspended in 30 ml of milk

Similar studies were conducted by McCullough and Eisele (19,20) to determine a salmonellosis end point for several other Salmonella strains and species. Serial dilutions of the suspension of organisms were plated on trypticase soy agar in duplicate for final bacterial count. Table 2 shows the results of these studies and indicates that a wide variation in cell numbers was required to produce illness. Six adult male volunteers ingested various densities of the organisms suspended in a glass of eggnog. Subjects were selected who had no indication of Salmonella infections, i.e., no organisms in their stools and absence of high serum agglutination titers. The natural course of the illness was followed without specific treatment except when medically indicated. Symptoms generally included abdominal cramping, nausea, diarrhea, and low grade fever. Two strains of S. meleagridis did not produce illness in the 12 volunteers who received 5.5 million organisms. However, the ingestion of 1.7 million cells of S. bareilly produced illness in four of six (67 percent) subjects. The lowest pathogenic dose observed with eight organisms studied was 125,000 cells of S. bareilly. One of six subjects became ill after ingesting this number of organisms.

Table 2. Number of Salmonella Organisms Required to Produce Clinical Illness in Healthy Adult Male Volunteers (19,20)

Organism	Dose x 10^{-6}			
	0%*	15-20%	30-50%	65-85%
S. anatum, Strain I	.26		.58	
S. anatum, Strain II	24	45	67	
S. meleagridis, Strain I	5.5	24		50
S. meleagridis, Strain II	5.5	10	20	41
S. meleagridis, Strain III	1.5	7 7	10	
S. bareilly		.13	.70	1.7
S. newport		.15	1.4	
S. darby	6.4		15	

*Percentages are number of volunteers ill/total number dosed x 100; six volunteers normally exposed/dosage level

Shigella has been found to be a more virulent genus of enteric bacteria than Salmonella. Symtoms of shigellosis include diarrheal stools containing blood and mucus, abdominal cramps and high fever. A group comprised of federal and university investigators has conducted a series of Shigella feeding studies in inmates of a correction institution (7,16) Adult male volunteers were fed various types and strains of Shigella organisms suspended in milk. Inocula were prepared by diluting 24-hour agar plate cultures and the cell number confirmed by making pour plates of the inocula before and after each experiment was conducted. Illness was terminated in the volunteers by the administration of an antibiotic.

Results of studies with S. flexneri 2a and two strains of S. dysenteriae 1 are shown in Table 3. All three organisms produced illness in a significant number of subjects fed 200 cells. One of 10 subjects fed 10 cells of S. dysenteriae 1, strain M131 became ill. This strain had been responsible for a dysentery pandemic in Central America during 1968-1970. The virulence and multi-drug resistance of this organism actually led to work on a live immunizing agent for this disease. Although not reported in detail, these investigators mentioned in an additional report the production of disease in a significant percentage of adult volunteers fed 10 to 100 viable cells of S. flexneri 2a and S. sonnei as well as S. dysenteriae 1 (8)

A limited amount of dose-response data has been reported for three additional enteric bacteria that, on recent occasions, have been associated with waterborne disease in the United States and could be of health concern in wastewater exposure (4,5). Yersinia enterocolitica can produce in man an enteritis similar to salmonellosis. Szita et al. (25) have reported that illness was produced in a single volunteer fed 3.5×10^9 cells of the organism. Enteritis also has been recently associated with the ingestion of Campylobacter jejuni. One investigator experienced abdominal cramps and mild diarrhea after ingesting 500 organisms, in 180 ml of milk, of a strain that had

been isolated from a milk-borne outbreak (23). Pathogenic and toxigenic strains of the common intestinal bacterium, Escherichia coli, have been associated with waterborne enteritis. Koya et al. (14) studied the illness response of E. coli enterotoxigenic strain O-111 B4 in four male volunteers. No illness resulted in the subject fed 2.7 x 10^7 organisms. Mild diarrhea, abdominal pain, and fever resulted in three subjects fed 5 x 10^7 to 10^9 organisms. DuPont et al. (6) studied two nontoxigenic invasive strains of E. coli and observed illness in eight of 13 adult volunteers fed 10^8 cells. No clinical disease was apparent on ingestion of 10^6 cells unless an antacid was also administered. An additional enteric bacterium Vibrio cholerae, although no longer a significant pathogen in this country, remains as a major etiological agent in the developing countries. Studies with the Inaba 569B strain of V. cholerae have indicated that a dose of 10^8 cells is required to induce diarrhea in the absence of concomitant antacid administration (2). None of these studies except Koya et al. (14) and DuPont et al. (6) were designed to determine a minimal dose response. Of the enteric bacteria that have received considerable study, Shigella has been found to be the only genus that produces illness in healthy adults at relatively low exposure levels, i.e., ≤ 200 organisms. However, the very limited data from one exposure to 500 cells of C. jejuni indicate that this genus also may have a low infective dose.

Table 3. Response in Healthy Adult Male Volunteers to Various Doses of Virulent Strains of Shigella

Organism	Dose	No. Ill/No. Fed	% Ill	Ref.
S. flexneri 2a	180	8/36	22	7
	5,000	28/49	57	
	10,000	52/88	59	
S. dysenteriae 1 (Strain A 1)	200	1/4	25	16
	10,000	2/6	33	
S. dysenteriae 1 (Strain M 131)	10	1/10	10	16
	200	2/4	50	
	2,000	7/10	70	
	10,000	5/6	83	

DOSE RESPONSE TO ANIMAL PARASITES

Historically, amebiasis caused by the protozoan Entamoeba histolytica has been the most important environmentally transmitted disease of animal parasite origin in the United States. Most infections are asymptomatic; however, its pathogenicity is well documented and the occurrence of cysts in the stool is always of concern. Rendtorff was interested in determining the dose-response of this organism in man, but felt it improper to purposely expose prisoner volunteers to this pathogen. He chose instead to determine the infective dose of a non-pathogenic amoebae: Entamoeba coli (21). Adult male volunteers that

showed no amoebic infections on stool examinations were selected for this study. The volunteers were individually housed under specifically controlled environmental conditions during the 10-week study period to minimize extraneous infections. The cyst inoculum was obtained from volunteer donors and was separated from debris and other organisms by a flotation procedure and micromanipulation. Individual dosage levels were obtained by micromanipulator isolation of the cyst, a very accurate technique for obtaining low-dose numbers. Appropriate volumes of the cyst suspensions were placed in gelatin capsules and swallowed with 120 to 180 ml of tap water.

Table 4 shows the results of this study Infection, as determined by repeated fecal shedding of cysts, was achieved in one of eight volunteers who supposedly ingested only one cyst. The increasing infectivity rate with doses of 10 (30 percent) and 100 (50 percent) cysts lends support to the authenticity of the single positive response (12.5 percent) on ingestion of one cyst. In addition the rigorous experimental design that included isolation of the volunteers, negative controls, and a highly sensitive cyst counting procedure tends to lend credibility to the remarkable finding.

The reluctance of Rendtorff to conduct human feeding studies with E. histolytica was apparently not shared by Beaver et al. (1) They obtained cysts from an asymptomatic donor and fed adult volunteers a single dose ranging from 2,000 to 1 million cysts. Unfortunately, the lowest dose used in this investigation produced infection in all volunteers (42 of 42) which precluded the determination of a minimal infective dose (Table 4). It should be noted that the authors concluded that no disease symptoms could be attributed to E. histolytica infections in these subjects.

Table 4. Response in Adult Males to Various Doses of Amoebic Cysts

Cyst Type	Number of Cysts Given	No. Infected/ No. Fed	% Infected	Method of Adm.	Ref.
E. coli	1	1/8	12.5	gelatin capsule	21
	10	3/10	30		
	100	2/4	50		
E. histolytica	2000 to 4000	42/42	100	beverage suspension	1
G. lamblia	1	0/5	0	gelatin capsule	22
	10	2/2	100		
	100	2/2	100		

Although Giardia lamblia was not the important pathogen in the 1950s that it became in the 1970s, Rendtorff included this protozoan in his study of the E. coli model of amoebic infection (22). This early work represents the only human dose-response study conducted to date with this organism. Volunteers were fed either Giardia cysts alone or Giardia plus E. coli cysts in gelatin

capsules. The inocula counts were determined by micromanipulator isolation as stated above. A single volunteer received one of three dosage levels of Giardia alone or the same dosage levels of Giardia and E. coli cysts. Table 4 shows the results of this study and indicates that a dose of 10 cysts or less is sufficient to produce infection in a high percentage of exposed susceptible individuals. Even though none of the five volunteers was shown to be infected on ingestion of a single cyst, it is reasonable to assume, based on the infection of two of two with ten cysts, that with a larger number of exposed subjects, infection of a significant percentage would have occurred on ingestion of one viable cyst. Perhaps the apparently more virulent strain occurring in the United States today would be more infectious. However, the direct correlation of parasite virulence (as indicated by disease frequency or severity) with the exposure level required for a host response has not been adequately demonstrated. Interestingly, these data indicate that these animal parasites have about the same degree of infectivity as Shigella bacteria.

DOSE RESPONSE TO ENTEROVIRUSES BY ORAL INGESTION

The last group of enteric pathogens to be considered, viruses, represents a somewhat more difficult agent to be safely studied in humans. Since viruses are obligate intracellular parasites, even the infectivity end-point requires the occurrence of the somewhat uncontrollable process of cell destruction, i.e., virucidal drugs are not available to interrupt the infectious process. The development of avirulent strains of polioviruses in the 1950s provided opportunity to conduct dose response studies with live vaccine strains with minimal risk of adverse responses. It also allowed such studies to be conducted with a subset of the population that appears to be more susceptible to natural infections, i.e., infants and children.

Koprowski and his colleagues at Lederle Laboratories conducted the first reported dose response studies with attenuated strains of polioviruses. Strain SM of poliovirus type 1 had been attenuated by rodent adaptation followed by successive passages of the virus in chick embryo and monkey kidney tissue culture. The virus was non-pathogenic for monkeys on intracerebral injection. As a component of the field trials of this potential live vaccine virus, Koprowski et al. (13) conducted a dose response study in children at a state institution. A total of nine children, who showed no antibodies to type 1 poliovirus, were given various doses of the virus suspended in polyethylene glycol 400 (0.5 ml) within a hard gelatin capsule. Each subject swallowed two capsules consuming at the same time 8 ml of milk. The dosage was determined by making 10-fold dilutions of a virus preparation titrated by the plaque technique using monkey kidney cell monolayers. Infection was determined by fecal shedding of the virus or by a specific antibody response.

The results of the study are shown in Table 5. Of course, the dose of 0.2 plaque-forming particle (PFP) is a dilution-determined average value and indicated that a single PFP would not occur in most doses. The calculated dose of 2 PFP produced virus shedding and an antibody response in two of three subjects. The investigators, aware of the implications of this remarkable

finding, suggested that the following factors should be clarified before assuming that these data represented a practical occurrence (12): (a) interference in virus titration by the diluent thereby giving an inaccurately low value, (b) insensitive assay system that did not detect all the PFP that would be infectious for the subjects, and (c) artifically high sensitivity due to delivery of the encapsulated virus directly to susceptible cells in the intestinal tract. Concurrent studies with an attenuated type 2 poliovirus indicated that 300 units of this virus were required for infection (13). However, titration of the rodent-adapted virus was performed in intact mice yielding 50 percent mouse paralytic doses, a less sensitive quantitative procedure.

Poliovirus vaccine feeding studies were subsequently performed by others over the next few years (17). However, significant infection levels (>50 percent) were not reported with doses $<10^{3.5}$ tissue culture infective dose 50 percent ($TCID_{50}$) until the studies of Katz and Plotkin (11) in the mid-sixties. These investigators studied the dose response of poliovirus type 3, Fox strain in 22 premature infants in the nurseries of a general hospital. Within the first 48 hours of life, each infant was given a low dosage of the virus, suspended in 5 ml of Hanks' solution, directly into the stomach by gavage using a rubber oro-gastric tube. The tube was flushed with 10 ml of saline solution before removal. Infectivity was determined by fecal shedding of the specific virus type administered.

Table 5. Response of Infants and Children to Low Doses of Poliovirus Live Vaccine

Dose	No. Infected / No. Fed	% Infected	Method of Adm.	Virus Type (Strain)	Ref.
PFP*					
0.2	0/2	0	Gelatin	1 (SM)	13
2	2/3	67	Capsule		
20	4/4	100			
$TCID_{50}$†					
1	3/10	30	Gavage	3 (Fox)	11
2.5	3/9	33	Tube		
10	2/3	67			
16	0/2	0	Aqueous	1 (Sabin)	18
50	3/6	50	Susp.		
90	3/4	75			
160	3/3	100			

*plaque-forming particles

†50% tissue culture infective dose

Results of this study are also shown in Table 5. At the lowest dose administered, 1 $TCID_{50}$, three of 10 subjects were infected. The $TCID_{50}$ quantitation procedure is thought to be less precise than the plaque procedure.

However, the investigators demonstrated a high degree of accuracy in dose titration. The lower dosage levels (2.5 and 1 $TCID_{50}$) were titered before administration to the infants by adding 0.1 ml aliquots of each inoculum to 50 cell culture tubes. The titration results yielded positive findings within 0.5 tubes of the statistically predicted number for each titer. A line fitted to a log probability plot of the dose-response data indicated that the 50 percent infective dose for the subjects was 4 $TCID_{50}$. These findings and the authors' conclusion that "a dose of any pathogenic virus sufficient to infect tissue culture would also be infectious for man" have been extensively cited.

Recent Virus Dose Response Studies

In an effort to obtain virus infectivity data by more normal exposure routes, the U.S. Environmental Protection Agency (EPA) has funded an additional infant poliovirus feeding study at the University of Wisconsin (18). Infant patients of a private pediatric practice were recruited to receive a reduced dose of the commercial poliovaccine type 1 (Sabin) two weeks prior to the scheduled receipt of the full dosage. The commercial vaccine was diluted in sterile distilled water to the desired dosage and administered in 0.5 ml volumes to the oral cavity with a 1-ml syringe. Each infant was observed to detect expectoration so as to insure that the entire dose had been swallowed. Infection was determined by the shedding of virus in the stool within 10 days post inoculation. Viruses isolated in the stool were identified as polio 1 by specific neutralization test.

Thirty-two 2-month-old infants were fed doses of 7 to 280 $TCID_{50}$ of virus. Few infants were given the identical dose since experiments conducted at different periods with freshly diluted virus gave slightly different titers. Results obtained with multiple feedings of the same dose are shown in Table 5. In addition to the two infants fed 16 $TCID_{50}$, three more yielded negative findings at doses of 42, 27, and 7 $TCID_{50}$ (data not shown). Statistical analysis of all the data yielded a 50 percent infective dose of 72 $TCID_{50}$, considerably higher than the findings of Koprowski and Katz and Plotkin.

An additional study has been supported by EPA to obtain data on the infective dose in adults of a "wild" enteric virus. The virus, echovirus 12, used in the study had been isolated from an 8-year-old girl with erythema infectiosum (fifth disease). Previous volunteer studies had shown the virus to be a very mild pathogen normally producing asymptomatic infections with common-cold type symptoms occurring infrequently.

Healthy male students having no evidence of echovirus 12 infection were recruited from local colleges. Selected subjects were isolated, 2 to a room, from outside contact for 8 days, one day prior and 7 days after ingestion of the virus. Appropriate dilutions of the purified virus were suspended in 100 ml of distilled water. Volunteers ingested 100 ml of the virus suspension or 100 ml of sterile water (negative control) under a double blind experimental design. Neither the volunteers nor the investigative staff knew the contents of the inoculum. Health status of dosed subjects was monitored twice a day by a nurse and physician. Throat and rectal swabs were collected daily during the

isolation period for virus assay. Blood specimens were collected on the day prior to inoculation and on days 6 and 26 for echovirus 12 antibody tests.

Preliminary results from this study have recently been reported (24). None of the subjects became ill. Table 6 shows the infection response. At the lowest dose administered, 10 plaque-forming units (PFU), an infection rate of 19 percent was observed. Subsequent to this report, additional data have been obtained for a total of 108 subjects fed one of four concentrations of virus: 10, 30, 100, and 300 PFU. Fecal shedding of the virus was found to be a more sensitive indicator of infection than humoral antibody response (Schiff, personal communication).

Table 6. Infection Response in Healthy Young Adult Male Volunteers to Various Oral Doses of Echovirus 12 (24)

Dose (PFU)*	No. Infected/No. Fed	% Infected
10	6/32	19
30	2/7	29
100	14/21	67

*Plaque-forming units

Statistical analyses of these data and the data of Minor (18) are shown in Table 7 From these analyses, an estimate of the infective dose can be made at exposure levels lower than can be practically obtained by experimentation. It should be noted that viruses assume a Poisson distribution in very dilute suspensions. At a mean virus concentration of one PFU/unit volume (dose), the probability that a dosage volume will contain zero PFU is 37 percent. Therefore, it becomes impossible to distinguish between a non-response in a large percentage of the subjects due to administration of no virus versus administration of sub-infective numbers, a major objective of a dose-response study Under this condition, the experimental error may be greater than the extrapolation error from a statistical analysis. Nonetheless, it seemed appropriate to estimate the dose required to infect 1 percent of exposed subjects. Table 7 shows that 20 $TCID_{50}$ (7-52) of polio type 1 and 0.4 PFU (<1-2) of echovirus 12 would be required. The echovirus 12 data appear to be the strongest data yet available indicating that the oral ingestion of a single detectable unit of virus may be infectious for a certain portion of a susceptible population.

Table 7. Estimates of the Number of Cell-Culture-Infective Doses of Two Enteric Viruses Required to Produce Infection in Humans When Ingested Orally

Virus	% Infected	Virus Dose	95% Confidence Limits	Ref.
		$TCID_{50}$		
polio 1	50	72	55-93	18
	10	39	24-63	
	1	20	7-52	
		PFU		
echo 12	50	35	21-64	24
	10	3	<1-7	
	1	.4	<1-2	

DOSE RESPONSE TO ENTERIC VIRUSES BY INHALATION AND EYE EXPOSURE

The portal of entry into the body by enteric viruses is generally the alimentary tract through the mouth. Initial infection of susceptible cells in the pharynx and gut may then spread to other parts of the body. However, some enteric viruses apparently may produce primary infections in the respiratory tract and conjunctiva. Members of the coxsackie A and adenovirus groups have been associated with respiratory disease. Adenoviruses and enterovirus type 70 have been isolated from the eye in cases of keratoconjunctivitis.

Infective dose studies in antibody-free volunteers exposed to enteric viruses by the respiratory route have been conducted jointly by the U.S. Army Biological Laboratories and the National Institute of Allergy and Infectious Disease. Subjects received aerosol inoculations by means of a molded rubber face mask to insure inhalation of a measured dose. Particles were generated by a Collison atomizer at a size ranging in diameter from 0.2 to 3.0 micrometers (m). Most particles were in the 1.0 to 2.0 μm range and these particles also contained the majority of the recoverable viral units.

Dose-response studies conducted with coxsackievirus A21 and adenovirus 4 by Couch, et al. (3) are summarized in Table 8. Nineteen volunteers received a dose of coxsackievirus ranging from six to 71 $TCID_{50}$. The lowest dose that produced a response was 18 $TCID_{50}$. One of four subjects was infected at this dose as indicated by virus shedding and/or antibody response. The calculated 50 percent human infective dose (HID_{50}) for this study, as determined by the Spearman-Karber method, was 28 $TCID_{50}$. Since it is generally recognized that with respiratory viruses local multiplication produces local symptoms, it is interesting to note that seven of the nine infected subjects experienced respiratory illness.

In the study of adenovirus 4, nine antibiotic-free volunteers received low concentrations of virus contained in small-diameter aerosols. The result

of this study is also shown in Table 8. At the two lowest dosage levels administered, three of three subjects inoculated with five $TCID_{50}$ and one of three inoculated with one $TCID_{50}$ were infected which indicated that the HID^{50} approximated the $TCID_{50}$. All infected subjects experienced lower respiratory tract illness. These studies showed that aerosols in the 0.2 to 3.0 μm range penetrate into the lower respiratory tract and that cells here are highly susceptible to at least these two enteric viruses.

Table 8. Response of Antibody-Free Volunteers to Small-Particle Aerosol Inoculation of Enteric Viruses (3)

Virus	Inhaled Dose ($TCID_{50}$*)	Dose Response No. Ill/No. Exposed (%)	50% Human Infective Dose ($TCID_{50}$)
Coxsackie A21	71	5/5 (100)	28
	47	3/4 (75)	
	18	1/4 (25)	
	6	0/6 (0)	
Adenovirus 4	11	3/3 (100)	~1
	5	3/3 (100)	
	1	1/3 (33)	

*Tissue culture infective dose 50%

The same investigators also showed that larger diameter aerosols (approximately 15 μm) would transport these viruses to the upper respiratory tract and produce infection there (3). A similar infective dose, i.e., approximately 30 $TCID_{50}$, was obtained with coxsackievirus A21 suspended in particles of either size range. The lowest infective dose was obtained by administering the virus via nasal drops. The instillation of 0.25 ml of diluted virus suspensions into the nostrils of 14 volunteers produced an HID_{50} of six $TCID_{50}$. These data suggest that the nasal mucosa is the preferred respiratory site for infection. The HID_{50} for adenovirus suspended in large diameter aerosols was not given. However, data were presented that showed 1000 $TCID_{50}$ produced infection in six of six exposed volunteers.

Extensive minimal-infective-dose studies of enteric viruses via conjunctival inoculation have not been reported. However, in a broader study of adenovirus infections, Kasel, et al. (10) produced conjunctivitis in two volunteers who were swabbed in the lower conjunctival sac with approximately 0.1 ml of media containing 30 and 300 $TCID_{50}$ of adenovirus 26. Rectal shedding of the virus was detected a few days after eye exposure.

DISCUSSION AND CONCLUSION

Man's relationship with microorganisms is dynamic and generally unpredictable in healthy hosts. Organisms are classified as human pathogens with the

understanding that a disease outcome of a host-microorganism interaction is dependent upon many variables. This understanding has been given greater expression recently with the increasing use of the term "opportunistic pathogens". Host susceptibility is influenced by age, sex, nutritional and health status. Pathogenicity and virulence of the organism is altered by mutation, selection and gene transfer. Natural or man-made environmental factors may determine the route of exposure, persistence of the organism, and the level of exposure. In the study of dose response, an infectious end point as indicated by *in vivo* multiplication of the agent and/or antibody response is perhaps the more reproducible outcome of exposure to enteric organisms since many of these variables affect primarily the occurrence of a pathogenic result.

Enteric pathogens are obviously able to infect the gastrointestinal system. Therefore, they must possess characteristics that allow them to compete and multiply in this environment. Theoretically, one particle should be capable of initiating infection. However, for this to occur, a viable particle must be transported to the site of multiplication. For the bacterial and parasitic pathogens, this is normally the lumen of the small intestine. Stomach passage actually activates some of the parasitic agents. However, survival in this acidic and enzymatic environment may be the major limitation of bacterial colonization of the intestine. It has been shown that an altered gastric function produced by buffering agents or disease process can increase susceptibility to enteric bacterial infection (6,15). Infection by enteric viruses is a completely different process from that of other enteric pathogens. Before a virus can enter a cell and establish infection, specific attachment to the cell surface must take place. The existence of complementary receptor sites on the host cell and the virion surface is the major factor in host specificity to these agents. Apparently a limited number of cell types exist in the intact host that are receptive to virus attachment. Therefore, infection appears to be mainly controlled by the probability of the contact-adsorption process occurring.

From the data of Minor, *et al.* (18) one may conclude that there is a probability of 1:100 that on the ingestion of 20 units of poliovirus 1, at least one particle will make specific attachment to and replicate within a susceptible cell of a human infant. The data of Stefanovic, *et al.* (24) are more remarkable in that they indicate that the ingestion of a single detectable unit of echovirus 12 could produce the same probability of infection in young adults. The observation of an even more likely infectious outcome (1:2) with the direct nasal application of one unit of coxsackievirus A21 (3) is consistent with the view that cell contact is the limiting factor. The shorter distance from virus entry to susceptible cells would favor a lower infective dose for infection of the nasopharynx by nose drops versus intestinal infection via oral ingestion.

The studies reviewed here have clearly shown that specific enteric organisms of all three classifications, i.e., bacteria, animal parasites, and viruses, can produce infections at relatively low exposure levels. It should be mentioned that these data, with the possible exception of Rendtorff's *Giardia* work (21), do not provide data on the number of particles producing

infection. Quantitation of the agents is limited by the sensitivity of the *in vitro* assay used. With viruses and bacteria, stable clumps of particles may represent single counts in an assay system. It is also possible that some particles infectious for an intact host may not multiply in a specific *in vitro* assay system. The quantitation error produced by these opposing limitations of detection techniques is unknown. It is only mentioned here to indicate that each infective unit detected by these assays does not necessarily represent a single particle of a given microbial agent.

The high infectivity of echovirus 12 via oral ingestion suggested a high transmission rate for this virus. This assumption was not supported by the occurrence of specific antibody in young adult men. Of 385 volunteers screened for the study, only 46 (12 percent) demonstrated antibody as determined by hemagglutination-inhibition test conducted with 1:5 dilutions of sera. However, the importance of this observation is obscured by the finding that antibody response is an insensitive indicator of echovirus 12 infection when produced by low virus exposure levels (Schiff, personal communication)

The data summarized in this report indicate that enteric pathogens can cause infections at exposure concentrations that typically occur in raw wastewater. However, available data are insufficeint to evaluate the actual health hazards that exist for individuals exposed to wastewater subjected to varying degrees of treatment and dilution. Obviously, important factors in the environmental transmission of infectious agents are not adequately understood. The identification and quantitation of these factors must be accomplished before the modeling approach can provide a realistic assessment of potential environmental health hazards and the importance of wastewater disinfection.

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4. VIRAL GASTROENTERITIS CAUSED BY THE SNOW MOUNTAIN AGENT, A NEWLY RECOGNIZED NORWALK-LIKE VIRUS

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ABSTRACT

The Norwalk-like agents are common causes of acute viral gastroenteritis which undergo waterborne and person-to-person spread. Outbreaks are often explosive in nature, with high attack rates and short incubation periods. Disease manifestations last 24 to 72 hours, and generally remit spontaneously. Recent studies of an extensive waterborne outbreak at a mountain resort, indicate that the etiologic agent (the Snow Mountain agent or "SMA") was a 26-32 nm virus which is morphologically similar to, but antigenically distinct from previously described Norwalk-like agents. Challenge of normal volunteers with orally administered SMA resulted in induction of acute gastrointestinal illness in 9 of 12 volunteers. Illness was similar to that seen in the naturally-occurring outbreak. Virus particles were detected in the stools of 2 of 5 naturally-occurring and in 3 of 9 experimentally-induced cases by immune electron microscopy (IEM). Serum antibody rises were detected by IEM in 8/9 volunteers who became ill, in 0/3 volunteers who did not become ill, and in 3 of the naturally-occurring cases. A highly sensitive and specific solid phase radioimmunoassay (RIA) was developed which detects SMA antigen and anti-SMA antibody. This RIA should enable assessment of the epidemiologic significance of SMA and may be useful in the consideration of control measures for waterborne spread.

INTRODUCTION

The Norwalk-like agents are common causes of acute viral gastroenteritis which undergo waterborne and person-to-person spread (3). Outbreaks are often explosive in nature, with high attack rates and short incubation periods. Disease manifestations last 24 to 72 hours, and generally remit spontaneously. The Norwalk-like agents are found in diarrheal stools, are approximately 27 nm in diameter, and thus far have not been successfully cultivated in vitro (6). At least four antigenically distinct agents have been described: the Norwalk agent (5); the Hawaii agent (15); the W-Ditchling agent (1); and the Marin agent (13). Because of the lack of suitable methods of detection, little is known about the prevalence, mode of transmission, and spread of most of the Norwalk-like agents. An exception to this is the Norwalk agent itself, for which a sensitive radioimmunoassay

has been developed (7). Employing this assay, infection with the Norwalk agent appears to be world-wide in distribution, and in one study, accounted for nearly one third of the outbreaks of gastroenteritis which were examined (8). A waterborne source of infection is suspected for the original outbreak in Norwalk, Ohio, from which the agent was derived, although no common source was detected (2). The Norwalk agent, however, has subsequently been associated with multiple waterborne outbreaks of gastroenteritis in the United States and in Austrialia (4,12,14).

The current studies were carried out with material obtained from an outbreak of acute gastroenteritis at a resort, Snow Mountain, near Granby, Colorado, in December of 1976, which has been reported previously (11). In brief, the characteristics of illness were typical of those of acute viral gastroenteritis: namely, fever, vomiting, and diarrhea, which lasted 24 to 48 hours. The attack rate was high, involving 418 of 762 individuals at risk, and person-to-person spread was noted. Illness was associated with consumption of water or ice containing beverages in a dose response manner ($p < 0.0001$). Stool specimens were negative for conventional bacterial pathogens, and virus cultures employing standard tissue culture systems were similarly negative. Because a limited amount of material (stools and sera) was available from the naturally-occurring outbreak, we performed a series of studies in normal volunteers at the National Intitutes of Health to determine the infectivity of the preparations and to generate additional reagents for in vitro studies. These studies were performed prior to the laboratory studies which were supported by the Environmental Protection Agency and are described below. Gastrointestinal illness, similar to that observed in the natural outbreak, was transmitted to 9 of 12 normal volunteers who were challenged with bacteria-free stool filtrates from one of the naturally-occurring cases (Dolin R, Reichman RC, Roessner KD, Tralka TS, Schooley RT, Gary W, and Morens D, Detection by immune electron microscopy of Snow Mountain agent of acute viral gastroenteritis, submitted for publication, 1982). The current report describes the detection of the viral agent of this outbreak (the Snow Mountain agent or SMA) employing immune electron microscopy, and reports the development of a sensitive solid phase radioimmunoassay for the detection of both antigen and antibody to the Snow Mountain agent.

MATERIAL AND METHODS

Preparation of stool filtrates. The stool filtrates are prepared as 2% suspensions of diarrheal stool in veal infusion broth supplemented with 0.5% bovine serum albumin. After low speed centrifugation, the suspensions are filtered through nitrocellulose filters of decreasing pore size to a final filtration through filters of 0.45 u in size. The filtrates are free of detectable bacterial, viral, and mycoplasmal agents by conventional techniques (10).

Immune Electron Microscopy (IEM). The techniques employed for IEM are those previously described for visualization of the Norwalk agent (9). 0.8 ml of the stool filtrate is reacted with 0.2 ml of convalescent serum overnight at 4°C. The reaction mixture is centrifuged at 17,000 rpm for 90 min. in an RC5B Sorvall centrifuge with a fixed angle rotor (SS34). The supernatant is discarded, and the pellet is resuspended in 1 drop of distilled water. The suspension is then placed on a 400 mesh Formvar grid, stained with 2% phosphotungstic acid, and examined under a Philips 300 electron microscope. The agent is detected as individual particles or aggregates coated with antibody. Conversely, employing a filtrate which contains a known concentration of antigen, antibody content in serum specimens can be assayed in a semi-quantitative manner, on a scale of 1 to 4+.

Radioimmunoassay (RIA) for SMA Antigen. The RIA for SMA antigen is performed as previously described (7). Purified anti-SMA IgG is prepared by $(NH_4)_2SO_4$ precipitation of convalescent (post-challenge) serum and dialysis with 0.005 M phosphate buffered saline (pH 8) at 4°C for 5 days. The globulins are purified by passage through a DEAE cellulose ion exchange column (Whatman DE52) at pH 8.0 with 0.005 M PBS. The purified IgG is labeled with ^{125}I by chloramine-T reaction. Wells in polyvinyl microtiter plates are coated with a 1:10,000 dilution of either pre- or post-challenge serum overnight at room temperature. After washing out the wells, varying dilutions of stool filtrates are incubated in each well overnight at room temperature. After additional washing, 50 ul of purified IgG containing 200,000 cpm of ^{125}I are added to each well, and incubated for 4 hours at 37°C. The plates are washed again, and individual wells are counted in a gamma counting system. Differences in binding of greater than 2 ($P/N \geq 2$) when wells coated with pre- and post-serum are compared, indicate the presence of antigen.

RIA for antibody to SMA. The methods are as follows: wells in polyvinyl microtiter plates are coated with a 1:10,000 dilution of a post-challenge serum specimen derived from a case of SMA-induced illness. 25 ul of a standard stool preparation containing SMA is added to the wells, incubated overnight at room temperature, and subsequently washed. This stool preparation results in a $P/N > 3$ in the above assay. Ten-fold dilutions of the serum to be tested, in 40 ul aliquots, are added to each well and again incubated over night at room temperature. 10 ul of ^{125}I labeled anti-SMA IgG is added to each well and incubated for 4 hours at 37°C. The plates are washed and counted as above. The titer of serum obtained is the reciprocal of the highest dilution which results in 50% or greater reduction in counts when compared to a PBS control.

RESULTS

Detection of Snow Mountain agent by immune electron microscopy. Examination of stool filtrates from five of the naturally-occurring cases from the Snow Mountain outbreak revealed 27 nm virus-like particles in two of the specimens. Examination of the stools from nine cases of experimentally-induced illness revealed similar virus-like particles in stools from three of these cases (Fig. 1). Particles appeared to have cubic symmetry, were non-enveloped, and were morphologically indistinguishable from the previously described Norwalk and Hawaii agents. The vast majority of particles were 27 nm in diameter, but occasionally particles as large as 32 nm were also seen. The particles were seen most frequently 24 to 48 hours after the onset of illness, but were not detected more than 72 hours after illness had begun. The 27 nm particles were not observed in stools obtained prior to challenge nor in volunteers who were challenged and did not become ill. Virus particles were observed either as single particles or aggregates heavily coated with antibody.

Detection of antibody to Snow Mountain agent by immune electron microscopy. The results of antibody determination in serum specimens from subjects challenged with the Snow Mountain agent as determined by immune electron microscopy are presented in Table 1. Eight of nine volunteers with experimentally-induced illness demonstrated serum antibody rises to the 27 nm particle, along with three of the three naturally-occurring cases (Nos 11, 12, and 13) which were tested. None of the three volunteers who were challenged with the Snow Mountain agent but did not become ill manifested serum antibody rises.

Antigenic relatedness of Snow Mountain, Norwalk and Hawaii agents. Analysis of serum antibody responses to the Norwalk, Hawaii, and Snow Mountain agents as determined by immune electron microscopy is presented in Table 2. Serum specimens were available from previous volunteer studies which had been carried out. Significant antibody rises to the homologous antigens were demonstrated with all three agents. No heterologous rises were seen among any of the agents, suggesting that the three agents are antigenically distinct as determined by this technique.

Radioimmunoassay for the Snow Mountain agent (SMA). Employing serum specimens which had marked differences in antibody ratings by IEM, a radioimmunoassay for the Snow Mountain agent was developed. The results of the radioimmunoassay for SMA antigen in stool filtrates from the three volunteers challenged with SMA who shed virus in stools are presented in Tables 3, 4, and 5. The pattern of shedding of SMA antigen was similar to that previously observed for the Norwalk antigen. SMA antigen was generally detected 24 to 52 hours after challenge and was no longer detectable beyond five days after challenge. Antigen was not detected in stools prior to

challenge, nor in stools from volunteers who did not become ill. SMA antigen was detected in 11 separate stool specimens from the previously described volunteer studies. RIA antigen was always detected in stools in which virus particles were seen by immune electron microscopy, as well as in a number of IEM-negative stools. The latter observation suggests that the RIA is more sensitive than IEM, or that alternatively, the RIA may detect soluble as well as virion-associated antigens. The radioimmunoassay did not cross-react with stools which contained Norwalk or Hawaii agents.

Radioimmunoassay for antibody to the Snow Mountain agent. Employing the above stool filtrates as a source of antigen, a solid phase radioimmunoassay for antibody to SMA was developed. The results of RIA antibody determinations are presented in Table 6. Serum antibody rises were observed in 8 of 9 volunteers with experimentally-induced illness after challenge with SMA, and in 3 of the 3 naturally-occurring cases. No antibody rises were detected in the three volunteers who were challenged and did not become ill. Serum antibody rises determined by RIA correlated well with those observed by IEM.

DISCUSSION

The current studies have clearly identified a new waterborne viral agent of gastroenteritis, the Snow Mountain agent. This agent is present in the stools of naturally-occurring cases in the Snow Mountain outbreak and has induced gastroenteritis after oral administration to normal volunteers in the form of 2% stool filtrates. Additional lines of evidence which support the etiologic significance of the virus particle are serum antibody rises detected in subjects with naturally-occurring and experimentally-induced cases of gastroenteritis, and the absence of such rises in individuals who were challenged and did not become ill. In addition, the particle is shed during acute illness but is not detected prior to challenge or when illness is not present. The particle is approximately 27 nm in diameter, has cubic symmetry, and morphologically resembles the Norwalk and Hawaii agents. However, SMA appears to be antigenically distinct from the Norwalk and Hawaii agents by immune electron microscopy as well as by radioimmunoassay.

In vitro detection of SMA was accomplised despite the inability to cultivate the agent in conventional tissue culture systems. Initially, the virus particles were detected in stool specimens from the naturally-occurring outbreak, employing immune electron microscopy. This cumbersome although powerful technique, relies on the ability of virus particles to be aggregated by specific antibody which can be readily recognized under the electron microscope. The etiologic significance of such particles, however, remains uncertain until specific antibody rises in acute and convalescent

serum specimens have been demonstrated. Immune electron microscopy was then utilized to identify stools with high concentrations of particles and serum specimens with high titers of anti-SMA antibody.

Because of the laborious nature of immune electron microscopy, only a small number of specimens can be examined by this technique. Therefore, progress in the field depends on the development of an efficient, yet sensitive method for the detection of SMA, such as a radioimmunoassay. Employing reagents known to contain antigen and antibody to SMA as identified by IEM, a solid-phase radioimmunoassay was established. This assay detected SMA antigen in the stools of volunteers following challenge with the infectious inoculum, and demonstrated a shedding pattern similar to that previously shown for the Norwalk agent. Serum antibody responses were also demonstrated in both experimentally-induced and naturally- occurring illness, and correlated well with antibody responses as determined by immune electron microscopy. While the RIA appeared to be equally sensitive to IEM in the detection of serum antibody rises, RIA was significantly more sensitive in the detection of antigen in stools than IEM. The latter phenomenon may reflect either an increased sensitivity of the radioimmunoassay for virus particles, or alternatively, the detection of soluble (non-virion associated) antigen which is not detected by IEM. Additional studies are required to resolve this question.

These studies again document the development of waterborne illness caused by viral agents. Data concerning the overall impact of waterborne disease in the United States are fragmentary, but the most recent CDC summary indicates that the total number of cases associated with reported outbreaks in 1978 numbered 11,435, which is a three fold increase from those reported in 1977 (4). Both of these figures likely represent gross underreporting of the problem. Particularly interesting is the fact that no etiology has been established in more than 50% of outbreaks of gastroenteritis reported to the CDC during 1978, despite analyses of samples for a variety of bacterial, parasitic and viral pathogens, including the Norwalk agent by radioimmunoassay. Of outbreaks in which an etiologic agent was determined, the Norwalk agent accounted for approximately 20% (4). Since the radioimmunoassay for the Norwalk agent does not detect antigenically distinct agents such as SMA or the Hawaii agent, it is conceivable that the Norwalk-like agents as a group may account for a much greater proportion of water related illness. It should also be noted that in contrast to several other waterborne pathogens, the Norwalk-like agents undergo rapid person-to-person spread once infection has occurred, so that the impact of waterborne transmission may be multiplied many-fold.

The establishment of the radioimmunoassay for SMA now provides a powerful tool with which to pursue studies of this new agent. Clearly a major requirement for advances in this field would be the establishment of *in vitro* culture systems with which to detect and study SMA, as well as

other Norwalk-like agents. Since a large number of samples can now be analyzed efficiently for the presence of SMA, intensive investigation of promising *in vitro* culture systems can be carried out. Similarly, the epidemiologic impact of disease produced by this agent can now be evaluated in studies of both acute outbreaks and seroprevalence. Enviromental sampling for this agent can also now take place, along with evaluation of procedures for decontamination of drinking water. Because of the documented waterborne spread of these agents, effective methods of decontamination of water sources may represent an important control measure for diseases caused by Norwalk-like viruses.

Acknowledgement

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TABLE 1. ANTIBODY RATINGS IN SERUM SPECIMENS FROM SUBJECTS CHALLENGED WITH SNOW MOUNTAIN AGENT (SMA) AS DETERMINED BY IMMUNE ELECTRON MICROSCOPY+

Subject	Illness Following Challenge	Pre-challenge Serum	Post-challenge Serum#
1	yes	<1	4+
2	yes	2+	3+
3	yes	<1	2+
4	no	3+	3+
5	yes	<1	3+
6	yes	<1	2+
7	yes	1+	1+
8	yes	2+	4+
9	yes	<1	2+
10	yes	2+	4+
11	yes*	1+	2+
12	yes*	<1	2+
13	yes*	<1	3+
14	no	<1	<1
15	no	2+	2+

*Naturally-occurring challenge during outbreak
+Ratings determined on a scale of 0 to 4+ employing a stool filtrate as a source of antigen
#3 to 6 weeks after illness

Table 2. ANALYSIS OF SERUM ANTIBODY RESPONSES TO NORWALK, HAWAII, AND SNOW MOUNTAIN AGENTS*(SMA) BY IMMUNE ELECTRON MICROSCOPY (IEM)

	Rating of serum specimens to homologous and heterlogous antigens					
Agent which induced illness in challenge study	Norwalk Antigen		Hawaii Antigen		SMA Antigen	
	pre	post	pre	post	pre	post
Norwalk (1)	1+	4+	2+	2+	<1	<1
Norwalk (2)	1+	3+	2+	1-2+	1+	1+
Hawaii (1)	2+	2+	1+	3+	1+	1+
Hawaii (2)	1+	1+	1+	3+	2+	2+
SMA (1)	2+	2+	1+	1+	<1	3+
SMA (2)	1+	1+	2+	2+	2+	4+

*Challenge studies performed previously

TABLE 3. RADIOIMMUNOASSAY FOR SMA ANTIGEN IN STOOL FILTRATES FROM VOLUNTEER #5 CHALLENGED WITH SMA

Time at which stool was passed	P/N*
24 hrs pre-challenge	0.80
6 hrs post-challenge	0.82
13 hrs post-challenge	1.01
24 hrs post-challenge	2.01+
120 hrs post-challenge	0.83

*P/N ratio determined by solid phase radioimmunoassay
+P/N $\geq$ 2 indicates the presence of SMA Antigen

TABLE 4. RADIOIMMUNOASSAY FOR SMA ANTIGEN IN STOOL FILTRATE FROM VOLUNTEER #9 CHALLENGE WITH SMA

Time at which stool was passed	P/N*
24 hrs pre-challenge	0.90
4 hrs post-challenge	0.93
30 hrs post-challenge	1.47
50 hrs post-challenge	1.51
52 hrs post-challenge	3.17+
70 hrs post-challenge	3.79
72 hrs post-challenge	2.53
77 hrs post-challenge	3.61
96 hrs post-challenge	1.10
122 hrs post-challenge	1.19

*P/N ratio determined by solid phase radioimmunoassay

P/N $\geq$ 2 indicates the presence of SMA antigen

TABLE 5. RADIOIMMUNOASSAY FOR SMA ANTIGEN IN STOOL FILTRATES FROM VOLUNTEER #10 CHALLENGED WITH SMA

Time at which stool was passed	P/N*
48 hrs pre-challenge	1.05
24 hrs pre-challenge	0.90
6 hrs post-challenge	0.96
24 hrs post-challenge	0.71
31 hrs post-challenge	2.29+
34 hrs post-challenge	5.26
52 hrs post-challenge	12.69
72 hrs post-challenge	10.57
96 hrs post-challenge	6.66
120 hrs post-challenge	4.05
144 hrs post-challenge	1.32
164 hrs post-challenge	1.20

*P/N ratio determined by solid phase radioimmunoassay
+P/N $\geq$ 2 indicates the presence of SMA antigen

TABLE 6. RADIOIMMUNOASSAY FOR ANTIBODY TO SNOW MOUNTAIN AGENT (SMA)

Subject	Illness following challenge with SMA	Dilution of SMA inoculum	Pre Challenge Serum antibody titer	Post Challenge Serum antibody titer
1	yes	10^{0}	100	6400
2	yes	10^{0}	200	800
3	yes	10^{0}	100	800
4	no	10^{0}	3200	1600
5	yes	10^{0}	100	1600
6	yes	10^{0}	<100	200
7	yes	10^{-1}	200	200
8	yes	10^{-1}	<100	1600
9	yes	10^{-2}	<100	800
10	yes	10^{-2}	100	3200
11*	yes	-	400	1600
12*	yes	-	100	800
13*	yes	-	100	1600
14	no	10^{-3}	100	200
15	no	10^{-3}	200	200

*Naturally-occurring illness during the original Snow Mountain outbreak - acute and convalescent serum specimens are compared

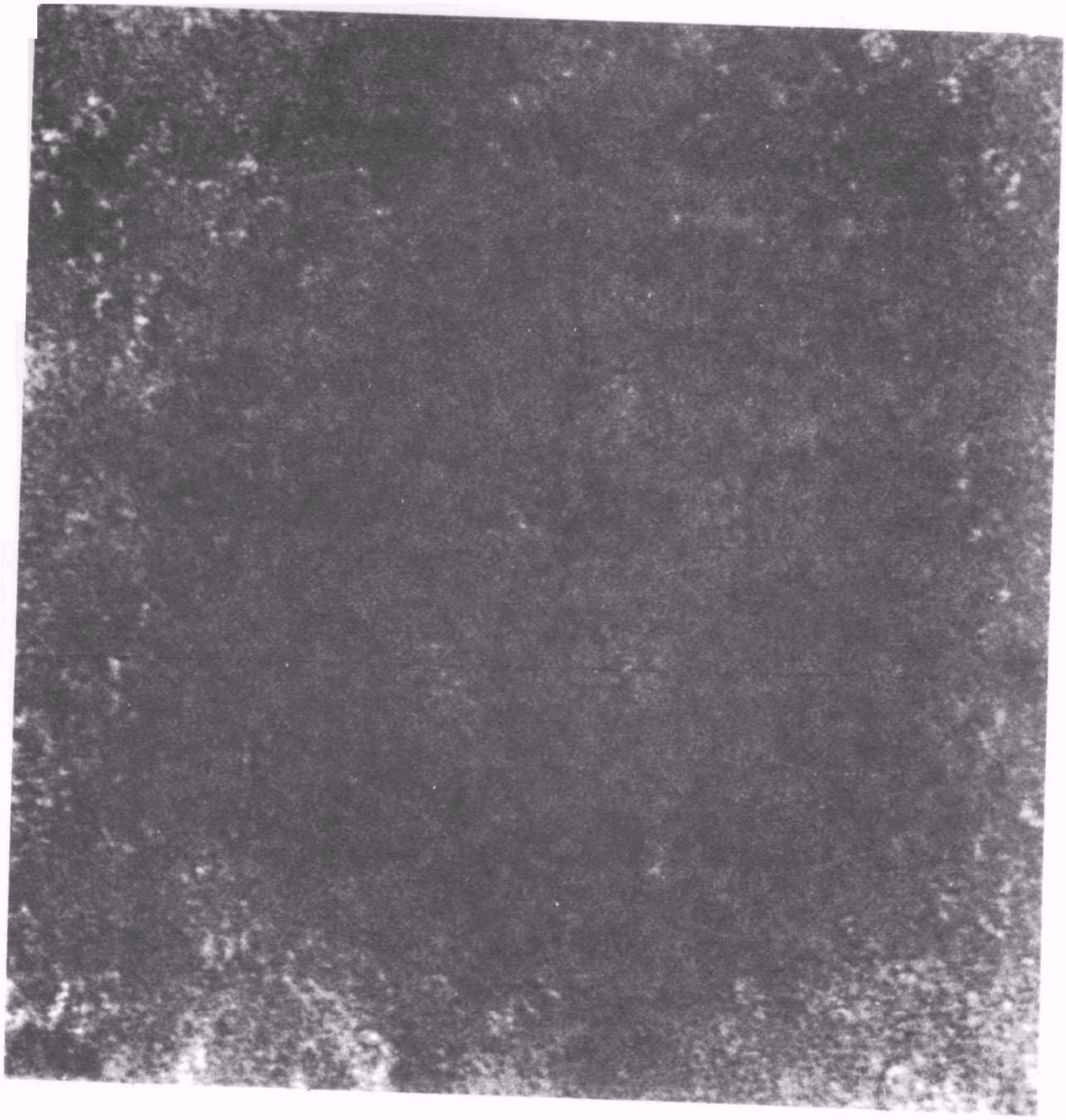

Figure 1. The Snow Mountain Agent as detected by immune electron microscopy. Virus particles are 27 nm in diameter and are heavily coated with antibody.

5. RISK ASSESSMENT OF WASTEWATER DISINFECTION

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ABSTRACT

An interdisciplinary team of University of Colorado at Denver faculty have performed a limited risk assessment of wastewater disinfection alternatives. The objective of the assessment was to provide policy makers with another tool to use in choosing among the alternatives of chlorination, chlorination/dechlorination, ozonation, ultraviolet radiation, and no disinfection. This paper summarizes some of the results of that study.

INTRODUCTION

What is risk assessment? I wish I knew. When we began our study months ago I thought I knew, but the experience of developing our risk assessment has convinced me that each risk assessment is a unique creation. Some have dubbed risk assessment an emerging science; others have called it an art. I have also heard it characterized as "jumping to conclusions from skimpy data". A veteran risk assessor, who advised us during our work, described the risk assessment process as gathering all the data you can find and then trying to make something out of it. The moral of this preamble is that you must approach any risk assessment with flexible objectives. Trying to fit a risk assessment into a preconceived structure will most likely lead to disappointment. On the other hand, viewing the results of a risk assessment within the constraints of a data base and of the assessment resources may produce a sense of accomplishment.

However, you cannot approach a risk assessment without objectives. In an earlier paper (1), I described our overall objective as the development of tools that decision makers, without expertise in the disinfection field, could use in the adoption of public policy relating to wastewater disinfection practices. I then subdivided this large objective into seven more specific objectives. The study results that I am presenting today approach the overall objective; however, not all of the seven more specific objectives proved to be reasonable goals.

Early in the project proposal stage we decided to narrow the focus of this project to only a few of the potential disinfection alternatives because the available funds would not support a study of all disinfectants. The criteria used to select the disinfection alternatives studied were: (1) the

alternative was not subject to constraints that would make its use unlikely, and (2) there was a good possibility that sufficient data existed to permit a risk assessment of the alternative. The second criterion was used because the project sponsors had asked us to confine our work to the available data base. The disinfection alternatives selected for study were: (1) chlorination, (2) chlorination followed by dechlorination, (3) ozonation, (4) radiation with ultraviolet light, and (5) no disinfection at all.

METHODS

Risk assessments often consist of four parts. The first part is the identification of the hazards. Each hazard is then investigated to identify all of the consequences that can arise if the hazard occurs. These consequences can usually be ranked or grouped according to severity. The third part is the development of a probability of occurrence (or frequency) statement for each consequence. Finally, the assessment interrelates the severity and frequency elements for all the hazards/consequences.

The hazards associated with each disinfection alternative were divided into three groups: (1) on-site use, (2) transportation, and (3) reaction product hazards. Hazards indirectly associated with the use of the disinfectants were not included in the study scope. For instance, hazards associated with manufacturing a disinfectant or constructing disinfection facilities were not studied.

The identification of some hazards was simple. For example, gaseous chlorine and ozone releases were obvious on-site and/or transportation hazards. On the other hand, the identification of the reaction product hazards required lengthy and sometimes fruitless literature searches.

The consequences of some hazards were also easy to identify and describe. For example, the effect of chlorine gas on humans is well known; and the ultimate consequences of death, physical impairment, and lost productive time were easily identified. Some consequences, such as the effects of chlorine gas on vegetation and inanimate objects required a little more literature searching effort for adequate identification. At the other end of the spectrum, the consequences associated with residual disinfectants and reaction products required massive literature searches. This latter set of tasks received a major portion of the study's resources.

Estimating the probability of occurrence or frequency of the consequences is usually accomplished using one of three methods. The simplest and most straight forward method is to collect the available data regarding the occurrence of the consequence in situations where the potential hazard exists. This method results in a quantitative statement of the consequence's probability of occurrence in terms of a common measuring unit; for example, deaths per man-year of exposure to the potential hazard.

A second method using available data can be used if a consequence can be quantitatively linked to a specific event, and the available data can be used

to estimate the probability of occurrence of the specified event. For example, there are no data relating exposure to chlorine gas to the occurrence of the expected health effects. However, there are data on lost time and total manhours worked in treatment plants which can be used to indirectly estimate the probability of the health effects occurring.

The third method of estimating the probability of a consequence occurring is used whenever the available data will not permit using either of the first two methods described above, i.e., there are no available data regarding the occurrence of the consequence or a directly related event. In this situation, the sequence of events leading to the occurrence of the consequence is defined in ever increasing detail until a level of detail is reached that will permit the estimation of the probability of occurrence of each subsequent event given that the previous event has occurred. For example, suppose that consequence C can result from event B, and event B is a result of event A. Also assume that we can estimate the probability of C occurring given that event B has occurred and the probability of event B occurring given that event A has occurred. Finally, assume that we can estimate the probability of event A occurring using available data. The probability of C occurring is then estimated using the following probability equation.

$$<C> = (<C/B>)(<B/A>)(<A>) \qquad (1)$$

where (< >) denotes probability of occurrence.
It is important to note that this simple example requires the estimation of three probabilities. Usually situations this simple are not found in risk assessments.

This third method becomes even more complicated and cumbersome when the consequence can result from two or more parallel sequences of events. When the event sequences and the consequence are described pictorially (see Figure 1) they resemble an upside down tree, which has led to this analytical method being labelled "fault tree analysis".

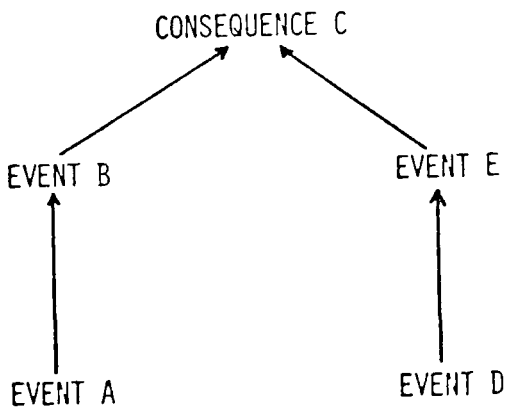


Figure 1. Simple Fault Tree

This simple fault tree results in the following probability equation for the estimation of the frequency of consequence C.

$$<C> = (<C/B>)(<B/A>)(<A>) + (<C/E>)(<E/D>)(<D>) \quad (2)$$

This fault tree method becomes even more complicated and cumbersome when Boolean logic and/or feed forward or feed back relationships are added to the fault tree.

Our original study proposal included extensive use of this fault tree method which accounted for over half the proposed budget. The use of this fault tree method was eliminated from the study to reconcile the needed resources with the available resources. A combination of the first two methods was, therefore, used in this study.

RESULTS

Chlorination

On-Site Use

The on-site use hazards of chlorination are human and vegetation exposure to liquid or gaseous chlorine. Liquid chlorine vaporizes so rapidly that exposure to liquid chlorine under normal working conditions is highly improbable. The consequences of exposure to gaseous chlorine are a function of exposure dose and length of exposure, and are summarized in Table 1.

Table 1. Consequences of Exposure to Gaseous Chlorine

Exposure Dose	Exposure Time	Consequences
Human Exposure		
<1 ppm	chronic	No consequences.
5 ppm	chronic	Respiratory problems, nausea, susceptibility to tuberculosis, corrosion of teeth.
7 ppm	1 hour	Mucous membrane irritation.
>7 ppm	1 hour	Cough, conjunctivitis, pulmonary edema, death.
100 ppm	seconds	Death.
Vegetation Exposure		
.5-1 ppm	1 hour	Spotting.
>1 ppm	1 hour	Death.

Note - ppm = part per million by volume

No data relating exposure dose or exposure time to illness, lost work time, or death could be found; however, large data bases relating death and/or lost time to manhours worked were found. The sources of these data banks

are shown in Table 2. The annual accident statistics publications were not useful because the accident data were not broken down sufficiently. The OSHA data were also not useful because municipalities are not required to submit reports to OSHA which eliminates too large a portion of the data sources. The OSHA data did report four deaths at wastewater treatment plants, but the reports did not identify the hazards causing the deaths. The SDS data provide adequate detail; however, two basic deficiencies of this data bank depreciate its usefulness. First, the data are collected at the state level, and not all states collect SDS data, which means the data are not based on a national sample. Second, the states do not require the reporting of lost time accidents when the lost time is less than a given minimum ranging from one to seven days. Thus, the minimal consequence accidents are not included in the data banks, and the amount of such data lost varies from state to state. We did, however, examine SDS data banks from several states and found no death reports due to chlorine exposure.

Table 2. On-Site Accident Data Sources

United States Department of Labor
- Bureau of Labor Statistics
  - Annual Accident Statistics
  - Supplementary Data System (SDS)
- Occupational Safety and Health Administration (OSHA)
  - Safety Programs Office
  - Office of Management Data Systems

National Safety Council (NSC)
Water Pollution Control Federation (WPCF)
American Water Works Association (AWWA)

Most of the NSC data do not provide sufficient detail; however, the NSC has reported one study of 156 treatment plants showing an accident rate of 40 lost workday cases per million manhours. Total lost time was 575 man-days per million manhours worked; however, these data include both collection systems and treatment plants.

The WPCF data are drawn from larger bases (7 to 10 percent of the plants in North America) and are separated into collection system and treatment plant groups. The most recent WPCF data for treatment plants are summarized in Table 3.

These data are in agreement with the NSC data shown above which indicate the accident rates for collection system employees and treatment plant employees are about the same. Comparing these rates with rates reported for other industries indicates that wastewater treatment plant work is about as hazardous as mineral mining. However, these data are still not sufficient for our risk assessment because the chlorine accident rate cannot be separated from our totals.

Data were collected for the broader based chemical and chlorine industries in an attempt to separate the chlorine accident rates from the totals by analogy. Ultimately, the AWWA data base was selected as the best analogy

because the treatment processes are similar. Furthermore, the AWWA injury frequency and severity rates were similar to the data shown above. And most important of all, the AWWA data base contains a grouping that is mostly chlorine accidents. According to the AWWA data chlorine related accidents represent about 4 percent of the total accidents reported. This is comparable to the accident rate reported for insect bites.

Table 3. 1979 WPCF Accident Data

Plant Size MGD	Man-hr per Employee	Injury Freq.	Severity Rate Lost Man-Days	Fatalities
<1.0	1861	22.16	252.9	0
1.0- 2.5	1985	38.99	210.9	0
2.5-10.0	1945	48.23	436.9	0
>10.0	1958	61.95	749.3	0
Average	1952	52.48	566.1	0

Note - MGD = million gallons per day
Injury frequency is cases per million man-hours
Severity rate is per million man-hours

Finally, we concluded that the only two consequences associated with the on-site use of chlorine that might be found in the data are death and lost work time. Furthermore, the data are not sufficient to permit the estimation of the probability of a death occurrence due to exposure to chlorine resulting from on-site use. However, the probability of lost work time can be estimated by applying the four percent figure found in the AWWA data to the WPCF data shown above. The resulting probabilities of occurrence are shown in Table 4.

Table 4. Probable Lost Work Time Resulting from On-Site Use of Chlorine

Plant Size MGD	Lost Work Time Man-hours Lost per Man-hour Worked
<1.0	0.00008
1.0- 2.5	0.00006
2.5-10.0	0.00014
>10.0	0.00024

Transportation

The consequences of the hazards associated with the transportation of chlorine are identical to the hazards associated with on-site use described above. The data on transportation accidents are also adequate for risk assessment purposes. They came from two sources, the United States Department of Transportation, and the Bureau of Census. These data banks permit the development of frequency estimates for deaths, injuries, and property damage; and those frequency estimates are summarized in Table 5. Regrettably, the injury

data do not include any measure of severity so the conversion of those data into lost work time is not possible. On the other hand, there is enough data to permit the disaggregation of the probability estimates for trucking into estimates for small cylinders, large cylinders, and tanker trucks.

Table 5. Probable Occurrence of Chlorine Transportation Consequences

Transportation Mode	Consequences Deaths	Injuries	Property Damage
Railroad	0.00063	0.02	$87.00
Railroad excluding Youngstown	0.0	0.0068	$1.80
Barge	0.0	0.068	$0.00
Truck			
Cylinders < 250#	0.0	4.0	$530.00
Cylinders 1 ton	0.0	0.02	$31.40
Tankers	0.0	0.047	$10.00

Note - Units are events or dollars per million-ton miles

The rail data included a catastrophic accident near Youngstown, Florida that included all of the recorded deaths and substantial amounts of injury and property damage. Therefore, two railroad entries are included in Table 5 to show the impact of that single accident.

The results shown in Table 5 can be used to estimate the probable consequences of using chlorine for wastewater disinfection for a given region if you can estimate the amount of chlorine being shipped into the region and the source of the chlorine shipments. Our final report will contain information allowing the development of these latter estimates.

Reaction Products

The identification of total residual chlorine as a hazard was obvious; however, the identification of chlorinated reaction product hazards was a problem. Numerous potentially hazardous chlorinated compounds have been identified, and it was not possible to determine the consequences resulting from the occurrence of each reaction product. Therefore, a subset of reaction products was selected for study. An interim report of an EPA study of priority pollutants in wastewater effluent provided an estimate of the chlorinated reaction products most likely to be found in a wastewater effluent. The availability of toxicity data and the use of some compounds as models for groups of compounds were also considered in the selection process. The reaction product hazards selected for study were: Total residual chlorine, chloroform, trichloroethylene, tetrachloroethylene, dichlorobenzene, chlorophenols, and 5-chlorouracil.

Exposure to the selected reaction products can produce consequences

primarily affecting humans, fish, and aquatic invertebrates. Human exposure to the reaction products requires ingestion via our water supply, water-based recreation, or consumption of aquatic flora or fauna. Available literature indicates the fraction of the chlorinated compounds in our water supplies that can be attributed to wastewater disinfection is negligible. Therefore, the probability of a consequence occurring via this route is almost nil. Similar analyses indicated that the probabilities of human consequences occurring via the other routes are also very small providing there is no bioaccumulation. Therefore, our study concentrated on the consequences occurring in the aquatic systems.

The consequences of exposing aquatic organisms, particularly fish, to the reaction product hazards cover a broad spectrum ranging from no effect to acute toxicity. Consequences falling between the two extremes include: avoidance, reduced spawning activity, reproductive dysfunction, and minor to severe physiological changes (e.g., decreased size, mutagenesis, carcinogenesis, etc.). In some cases, these consequences are further complicated by synergism among the reactants and by bioaccumulation. The literature relating these consequences to exposure doses and length of exposure is massive. Our biologists found over 400 pieces of data for residual chlorine alone. As a result, a major portion of our time was spent in this area.

Summarizing the results of this part of our study is not possible within the limits of this presentation. Therefore, I will show you only two of our more important results. The minimum reported acute toxic effects concentration and the minimum reported LC-50 divided by 100 are shown in Table 6 for each of the reaction products studied. The maximum reported effluent concentrations for each reaction product studied are also shown in Table 6.

Table 6. Reported Maximum Effluent, Minimum Acute Toxic Effects, and Minimum LC-50x0.01 Concentrations for Reaction Products Studied

Reaction Product	Maximum Reported Effluent Conc. mg/l	Minimum Reported Acute Toxic Effects Conc. mg/l	Minimum Reported LC-50x0.01 Conc. mg/l
Residual chlorine	8.0	0.001	0.00014
Chloroform	0.02	1.0	0.018
Trichloroethylene	0.04	1.0	0.36
Tetrachloroethylene	0.004	10.0	0.13
Chlorophenols	0.03	0.01	0.0003
Dichlorobenzenes	0.01	1.0	0.006
5-chlorouracil	0.004	0.01	No LC-50s reported

The results summarized in Table 6 indicate that residual chlorine will probably have acute toxic effects unless the effluent is well diluted in the receiving water body (not exactly a new finding). The summarized results

also indicate that all of the chlorinated reaction products, with the exception of the chlorophenols, will probably not cause any acute toxic consequences even with zero dilution in the receiving water body. And the worst case chlorophenol condition requires only a 3:1 dilution to reduce potential acute toxic effects to a negligible level assuming that the available data include the lower acute toxicity limit.

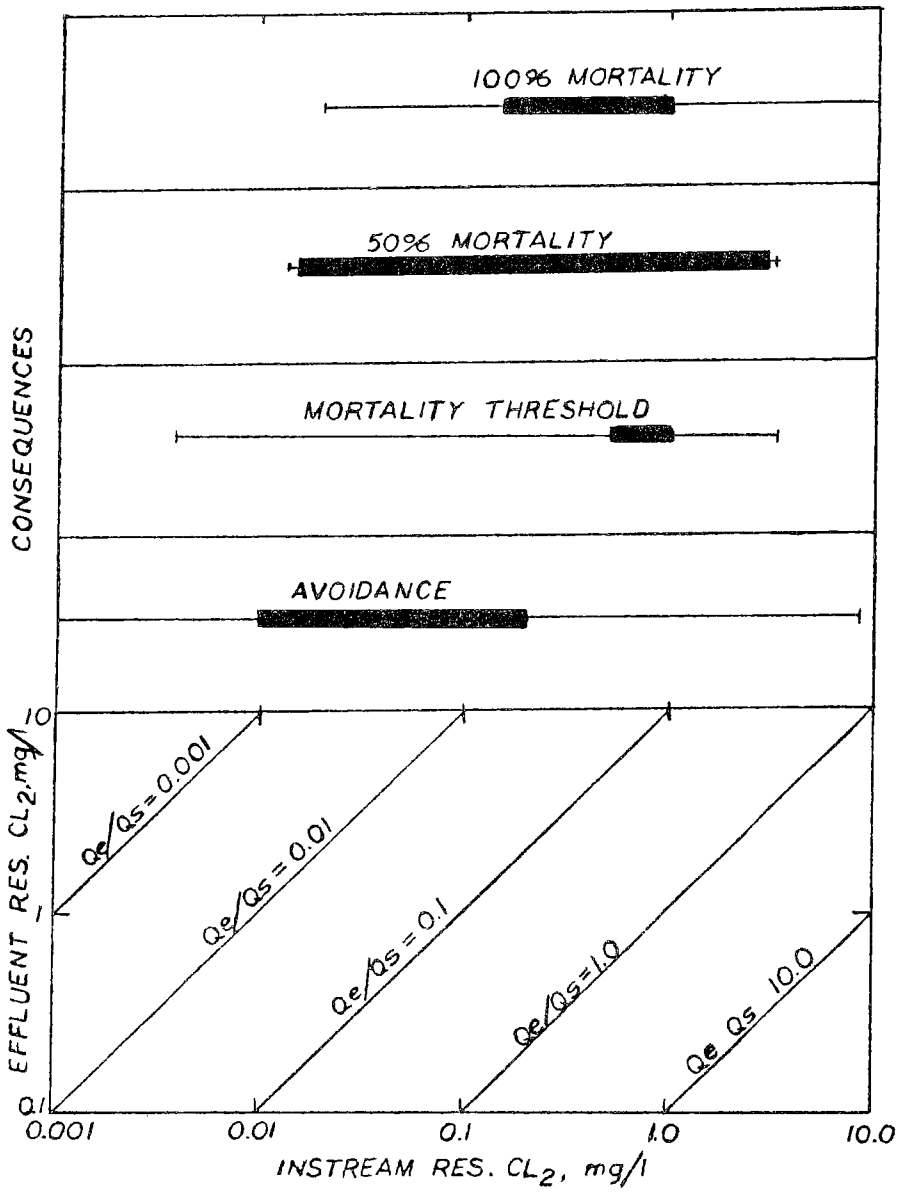
Assuming that the minimum LC-50x0.01 values are a reasonable estimate of the no effects threshold, the data summarized in Table 6 indicate reported effluent levels of chloroform, trichloroethylene, and tetrachloroethylene will probably have no effect on stream organisms even with zero dilution in the receiving water body. Chlorophenols may require dilutions up to 100:1, and dichlorobenzenes may require dilution up to 2:1 to reach the no effects concentration.

The residual chlorine data bank is so large that it permits a more detailed summary presentation. A simplified version of the method used in our study report to present the residual chlorine summary is shown in Figure 2. The thick bars shown in the upper half of Figure 2 enclose large concentrations of reported data for that consequence, and the thin lines reach to the outer limits of the reported data but do not enclose many data points.

Figure 2 illustrates some aberrations found in the chlorine data. For example, most of the mortality threshold data show the threshold occurs at concentrations much greater than many concentrations reported as LC-50's or LC-100's. Furthermore, many reported LC-50's are greater than the bulk of the reported LC-100's. These data help identify lower boundary conditions (i.e., worst case), but they are not much help in analyzing specific discharge and stream conditions.

Figure 2 also can serve as a fast method of estimating the impact of a specific discharge on a receiving stream. For example, a discharge of 1 mg/l residual chlorine with a Qe/Qs ratio of 0.01 results in a stream concentration near the left edge of the avoidance bar and just above the lowest values shown for mortality threshold.

Probability estimates can also be used with Figure 2. An example problem will illustrate this point. Assume the values shown in Table 7 can be developed from data available for a specific discharge. The first four columns can be used to calculate the probability of Qe/Qs exceeding 0.01 for each of the four flow combinations. The maximum probability is 0.338, and this is taken as the limiting condition. Suppose we want to estimate the probability of exceeding the 0.003 limit. At a Qe/Qs ratio of 0.01, the effluent residual should not exceed 0.3. Based on the data in Table 7, the probability of exceeding this value is 0.810 which means the overall probability of exceedance is 0.274 (0.338 x 0.810) or, in other words, the stream concentrations of chlorine will exceed 0.003 about once every 3.6 years on the average over all time. The same method can be used with Figure 2 to predict the probable onset of mortality, 50 percent mortality, and 100 percent mortality.



Note -- Qe = effluent flow rate
Qs = stream flow rate

Figure 2. Summary of Residual Chlorine Data

Chlorination/Dechlorination

The consequences resulting from discharging residual chlorine as presented above can be eliminated by adding an effective dechlorination process after the chlorination process. Usually the most cost-effective and, therefore, the chosen process is the addition of sulfur dioxide. Our literature search found that exposure to liquid or gaseous sulfur dioxide results in consequences essentially the same as those shown for chlorine in Table 1. Furthermore, the sulfonation process is very similar to the chlorination

Table 7. Assumed Data for Example Problem

Qs	<Qs>	Qe	<Qe>	ER	<ER>
1000	0.100	10	0.998	0.1	0.997
2000	0.450	20	0.750	0.3	0.810
3000	0.700	30	0.270	0.5	0.500
4000	0.845	40	0.065	1.0	0.110

<Qs> = Probability of non-exceedance of Qs shown
<Qe> = Probability of exceedance of Qe shown
ER = Effluent chlorine residual
<ER> = Probability of exceedance of the ER shown

process. Therefore, since no useful on-site use or transportation data were found for sulfur dioxide, we concluded that the frequency estimates developed above for on-site use and transportation of chlorine can also be used for the dechlorination process using sulfur dioxide.

Since the addition of sulfur dioxide removes the chlorine residual hazards and consequences, the reaction product hazards and consequences associated with the chlorination/dechlorination process will be the remaining chlorination reaction products plus any additional products or effects of the sulfur dioxide addition. The reaction products resulting from the addition of sulfur dioxide to wastewater are not discussed in detail in the literature, but available information indicates those products are mostly chlorides, sulfates, and sulfites. These products are not considered hazards in the aquatic ecosystem; however, the addition of sulfur dioxide may create low dissolved oxygen and pH conditions that can be hazardous. We could not identify the probability of these conditions occurring.

Ozonation

The on-site use hazards associated with the use of ozone for disinfection also create risks for both humans and nearby vegetation. The consequences of human exposure to ozone include: no effect; minor irritation to eyes, skin, and mucous membranes; headaches; respiratory distress; and death. Exposure of vegetation to ozone can include: stunting, defoliation, and death. The no effects threshold concentration for ozone is much lower than the chlorine threshold with some sensitive humans experiencing effects at concentrations as low as 0.02 ppm by volume. OSHA recommends a maximum exposure for an eight hour period of 0.1 ppm by volume (as opposed to a 5 ppm limit for gaseous chlorine). Since ozone is generated on-site using very high electrical voltages the ozonation process also includes electrocution as a potential on-site hazard.

No accident data were found that could be used to estimate the probability of realizing the consequences associated with the on-site use of ozone.

Therefore, we decided to survey the recently constructed ozonation plants to see if their limited experiences would provide a qualitative assessment of the risk. We hoped that we could then provide an intuitive assessment of the probability of realizing the consequences of on-site ozone use in terms of the on-site chlorine use estimates presented above. The probability of realizing the ozone consequences should be greater than the chlorine probabilities because (1) humans and vegetation are more sensitive to ozone, and (2) ozone is more likely to leak from the reactor because of its low solubility in water and its almost negligible vapor pressure in the atmosphere. Our telephone survey found several problems involving high ozone levels around the treatment plant. In our opinion, the probability of realizing the on-site use consequences associated with ozonation should be assumed to be several times greater than the same probabilities for chlorine.

Since we are not considering the hazards of transmitting power in this analysis there are no transportation hazards or consequences associated with ozonation.

The reaction product hazards include ozone and a massive group of low molecular weight alkanes, aldehydes, organic acids, and heterocyclics. The literature indicates consequences of ozone exposure for fish range from: locomotion and respiration impairment to death. These effects are also reported to occur at relatively low concentrations. The literature also includes data on ozone residuals; however, most of these data were observed in the ozone reactor effluent. Since ozone off-gases so readily, it is unlikely much of the reported ozone residuals would be found in the receiving water bodies. Therefore, we believe the potential consequences of aquatic exposure to ozone are unlikely to occur.

A subset of six low molecular weight organic compounds were selected from the mass of reported ozonation reaction products for toxicity analysis. The compounds studies were: Heptane, n-octane, n-hexanol, m-xylene, n-heptanal, and n-nonanal. No toxicity data were found for n-hexanal, n-heptanal, and n-nonanal. The literature did contain a few studies that show n-heptane and m-xylene are toxic in the mg/l range, and n-octane was found to be non-toxic at concentrations up to 100 mg/l. Even though no data were found regarding expected effluent concentrations it is unlikely any of these compounds will occur in hazardous amounts.

Ultraviolet Radiation

The hazards associated with the on-site use of UV radiation are: human exposure to radiation, electrocution, and human exposure to ozone. Human exposure can adversely affect both skin and eyes. The consequences of exposure include reddening, blistering, and peeling of skin and corneal damage, loss of visual acuity, and eye fatigue. Some literature was found linking skin cancer with UV radiation. UV disinfection processes operate at levels well above recommended human exposure limits so the probability of a consequence occurring is certainly greater than zero. However, the estimation of that probability was not possible with the available data.

UV radiation can produce ozone if oxygen is present in the exposure area. The ozone hazards and consequences have been discussed above; however, the available data indicate this is not a significant hazard.

Since UV radiation does not produce a residual the reaction product hazards are limited to changes in compounds existing in the wastewater. The limited amount of literature dealing with this phenomenon prevented any risk analysis of these hazards.

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WASTEWATER AEROSOL HEALTH EFFECTS STUDIES AND THE NEED FOR DISINFECTION

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ABSTRACT

A series of epidemiological studies of community exposure to aerosols from wastewater treatment plants, and a study of worker exposure to aerosols and sewage liquids/solids contact, were reviewed and evaluated. Gastrointestinal symptoms were reported in three of the studies and there was some serological evidence for viral infection, although a causal relationship to sewage exposure could not be established. The preponderance of data was negative and no definitive conclusions could be drawn. All of the studies were limited by having low numbers of exposed individuals and by being unable to adequately and quantitatively characterize exposure. Investigators have been unable to confirm the results of a 1976 report which indicated two to four times higher incidence of certain infectious diseases in agricultural communities using wastewater for irrigation. Two additional health effects studies on spray irrigation of wastewater are in progress.

INTRODUCTION

The principal potential routes for transmission of pathogens from sewage to the population are through contamination of drinking water and recreational water, or through aerosols from sewage treatment plants and spray irrigation practices. Contaminated food may also serve as a vehicle, but this review will focus on the potential for direct aerosol transmission of wastewater pathogens. The need for, and the efficacy of, disinfection of drinking water have been adequately demonstrated throughout this century. Typhoid and cholera have been dramatically reduced worldwide as a result of the combined effects of improved sanitation and drinking water disinfection. However, waterborne outbreaks of infectious disease still occur in the United States, and they can often be traced to absent or inadequate disinfection. Many of our surface water supplies and an increasing number of our groundwater supplies are subject to sewage contamination. Although the species of disinfectant and the point and manner of application may be subject to modification due to trihalomethane considerations, disinfection of drinking water appears to be a necessary practice that will continue to be widely used in this country.

The second significant route for exposure to pathogens from sewage is through recreational contact with contaminated fresh and marine waters. The evidence for health effects as a result of this contact, and consequently,

whether or not disinfection of wastewater discharged to receiving waters is indicated, is presented elsewhere in these proceedings.

The first two routes of exposure involve dilution of wastewater in an aqueous medium, and generally, oral ingestion of the contaminated water. However, it should be kept in mind that inhalation of aerosolized pathogens from contaminated drinking and recreational waters is also possible. The third route for potential exposure to pathogens from sewage is by dilution and transport in air at sewage treatment plants and spray irrigation sites. Over the last decade, the U.S. Environmental Protection Agency (USEPA) sewage construction grants program provided the impetus for the development of numerous activated sludge treatment plants. In the course of implementing this program, several communities questioned the health hazards associated with these plants, primarily from aerosols produced by the aeration basins. It soon became apparent that there was a lack of data on health effects of sewage aerosols.

A similar situation prevailed concerning aerosol hazards at land application and spray irrigation sites. Land application and agricultural reuse of municipal wastewater may have certain advantages over traditional treatment and disposal practices employed in the United States. Municipal wastewater can be considered as a resource rather than as an unwanted end product. It is possible to recover and utilize some of the nutrient value contained in the wastewater and to supplement water resources in water-poor areas. The acceptance and implementation of land application and reuse, however, depends upon resolving a variety of social, economic, and health effects issues.

Finding significant health effects associated with aerosols from activated sludge plants would most likely result in corrective actions other than disinfection, e.g., instituting procedures to minimize aerosol formation; the covering of aeration basins, or the erection of other physical barriers to aerosol transport. Even so, health effects data from sewage treatment plant studies could be useful in assessing the need for disinfection of wastewater effluents used at spray irrigation sites or discharged to recreational waters. Disinfection could be considered as an alternative or additional treatment process for effluents used at spray irrigation sites if significant health effects are demonstrated.

The purpose of this paper is to summarize the conclusions, limitations, and relevance to the question of wastewater disinfection, of several studies on the health effects of exposure to wastewater treatment plants and spray irrigation practices. All but one of the studies have been funded by the U.S. EPA. Two of the investigations are in progress and the rest were completed within the last four years. The results of most of these studies appear in the proceedings of a recent symposium (9).

STUDIES OF POPULATIONS NEAR SEWAGE TREATMENT PLANTS

Four epidemiological studies of populations exposed to activated sludge

treatment plants were completed in 1978 and 1979. The plant locations and type of exposed population are indicated in Table 1. Three of the studies involved heterogeneous community populations, and the fourth was of children at an elementary school near the surge and aeration basins of a treatment plant. Details and findings of each of these studies will be presented separately.

Tecumseh, MI

Tecumseh, Michigan was chosen as the site for this preliminary investigation because it was part of a comprehensive community health study conducted by the University of Michigan (3,9). Consequently, a considerable amount of retrospective health and demographic data were available. The study population was divided into five concentric rings radiating outward in multiples of about 600 m from an activated sludge treatment plant. In 1965, the plant was converted from a trickling filter facility to activated sludge treatment. The study period was from 1965 to 1971. Average monthly sewage flow rates during this time were from 0.64 to 1.18 MGD, although some of the data for this period are missing. Self-reported acute illnesses and symptoms from 4,889 participants during the 7-year period were grouped into total, respiratory, and gastrointestinal illness (GI) categories. Age-sex-distance-specific incidence and illness rates were analyzed using the minimum discrimination information statistic. Data on income and education were also used in the analysis.

The results indicated that there was a greater than expected occurrence of total respiratory and GI illnesses in those living within 600 m of the plant. However, this portion of the population also had lower income and education than the rest of the study group. The investigators suggested that high densities of lower socioeconomic families might be a more important factor in excess illness than would be proximity to a small wastewater treatment plant. Some excess illness (total, respiratory, and GI) was also reported for those living within the 2,400 m concentric ring. These people had both higher incomes and education than any of the other groups and there was no known source of exposure for acute illnesses in this area.

This retrospective study was obviously inconclusive and was undertaken because of the availability of the data and the relatively small expense involved. Limitations on interpretation of these results include: the presence of a confounding demographically heterogeneous population, the lack of exposure and meteorologic data, and the relatively low volume of the exposure source. Differences by distance were detected, but determining the causes of the differences was outside the scope of the project.

Schaumburg, IL

In this study, an opportunity was available to follow a community population before and after an activated sludge treatment plant went on line (5,9). The study period was from 1974 to 1976 and the John E. Egan plant became operational in December, 1975, with average daily flow rates of 10-15

MGD. The study design included a health survey of about 4,300 individuals (of a total population of about 100,000) living within 5 km of the plant. The residential area began 350 m from the plant. Clinical specimens from a subset of this population (226 individuals) living within 3.5 km of the plant were examined for bacteria, viruses, parasites, and viral antibodies in serum. Wind speed and direction, and relative humidity, were monitored on-site. Large-volume aerosol samples and wastewater samples were examined for indicators and pathogens.

The results of the health survey indicated no change in asthma-hay fever symptoms, decreases in worm infections, and decreases in sore throats after the plant went into operation. Statistically significant ($p \leq .01$) increases in the incidence of six diseases or symptoms were reported by those within 2 km of the plant (Table 2). Furthermore, all showed a relationship with direction from the plant, i.e , increases were at the close distances in the north and south directions, the predominant downwind quadrants. However, the results for at least five of the six show considerable variability with distance before the plant went into operation or show significant decreases in those living more than 2 km from the plant. Nevertheless, one symptom, diarrhea, showed remarkable uniformity in reporting throughout the study population and increased from 4.1 to 7.6 percent in those living 0-2 km from the plant. This finding is also interesting in that the post-operational survey included a lower proportion of young children who would be more likely to experience diarrhea.

These results must be considered in the context of the survey methodology employed. Participants were asked on two occasions to report all acute diseases occurring in the family in the previous year and to list all symptoms occurring in the previous three months. The accuracy of surveys requiring that much recall is open to serious question even if the respondent is reporting only on himself. In addition, survey participants knew that the study involved possible health effects of the sewage treatment plant.

Recognizing these limitations on survey information, the study included objective measures of infection as well, i.e., serology and isolation of pathogens from clinical specimens. Proteus, Pseudomonas, and Salmonella were the only pathogen isolates from fecal samples. There was a significant decrease in Proteus isolations during the operational period and no significant differences in Pseudomonas and Salmonella isolations. Streptococcus and Staphylococcus isolations from throat swabs increased during the operational period. However, the increases were not related to the treatment plant as shown by regression analysis of the incidence pattern with distance and direction from the plant. There were no significant differences in isolation of parasites from fecal specimens. No viruses were found in throat swabs but twenty viruses were found in fecal samples. There was a significant increase in virus isolations during the operational period but the increase could not be related to plant exposure. Antibody tests for 31 enteric viruses yielded no serologic evidence of an adverse wastewater treatment plant effect. The results of the aerosol monitoring indicated that levels of microorganisms in the air in residential areas were indistinguishable from background concentrations.

The investigators concluded that, at the exposure levels studied, sewage treatment aerosols from well-operated American plants do not appear to pose significant health hazards. They also indicated that there was insufficient evidence to determine if minor effects such as gastrointestinal symptoms and skin disease, were associated with aerosol exposure.

Tigard, OR

A retrospective study conducted in Oregon combined the rapidity and cost advantages of the Tecumseh study with the exposure categorization experience gained in the Schaumburg project. A new activated sludge treatment plant had been placed into operation in 1976 within 400 m of an elementary school and local public health officials expressed concern about possible health hazards. A preliminary study was performed to determine the types and numbers of microorganisms in air upwind and downwind from the treatment plant and to compare absenteeism rates, as a measure of possible health effects, at the affected school and control schools (6,9).

The study design involved collecting seven years of attendance data prior to initiation of plant operations and for two years afterwards. Data were collected for the exposed school (Durham) and for five control schools in the Tigard district. The plant had a design capacity of 20 MGD but averaged 9-13 MGD during the study period. There were two possible sources of exposure to the wastewater aerosols: from a surge basin located within 50 m of the school playground, and from an aeration basin about 400 m from the school building. The Durham Elementary School had six classrooms (one for each grade), open-window ventilation, and an enrollment in June, 1978, of 123 students. On-site measurements of wind speed and direction, temperature, humidity, solar radiation, and cloud cover were made. Composite wastewater samples and large volume air samples were collected from the aeration and surge basins and examined for indicator and pathogenic microorganisms.

The absenteeism results are shown in Table 3. As can be seen, the absenteeism at the exposed school actually decreased during plant operation. These negative data must also be considered in the context of the results obtained from the environmental monitoring and exposure calculations. The aeration basins were found to be a much stronger source of microorganisms than the surge basin. The geometric mean aerosol concentrations at 30 to 50 m downwind of the aeration basin were 12 colony forming units (cfu)/m^3 of total coliforms, 4.2 cfu/m^3 of fecal streptococci, 19 cfu/m^3 of mycobacteria, 1.5 plaque forming units (pfu)/m^3 of coliphage, and less than 0.0002 pfu/m^3 of enteroviruses. However, the exposure calculations based upon the meteorological observations indicated that the classroom area was steadily downwind of the aeration basin for only 10 days in the two operational years. In addition, because of rainfall, the playground may have been in use on only 40 percent of the days when it was steadily exposed to aerosols. The investigators concluded that the wastewater aerosols had no effect on infectious disease incidence as determined through absenteeism for this level of exposure.

Skokie, IL

Probably the most thorough and the last of the treatment plant health effects studies (1,9) was conducted by the University of Illinois near an activated sludge facility with an average daily flow of 290 MGD. A subset of the population living within a 1.6 km radius of the plant was studied for an 8-month period. A comprehensive health questionnaire survey was conducted of 2,378 persons at the beginning of the study to gather demographic and historical health information on chronic and acute diseases. A subset of this population (724 persons) was included in a health watch program where health diaries were collected on family members every two weeks throughout the study period. Although the keeping of health diaries is not without problems, this procedure was felt to be a significant improvement over study designs relying on recall over a 3-month or 1-year period. A subset of this population (161 persons) provided a total of 1,298 throat and stool specimens for bacterial and viral analyses. In addition, 318 persons provided paired blood specimens obtained at the beginning and end of the study period and these were used to determine prevalence and incidence of infections with enteroviruses. It should be emphasized that the participants knew only that this was a study of the possible health effects of air pollution--they were not told of plans to correlate effects with the sewage treatment plant.

The project also included microbial aerosol monitoring and meteorological data collection. These data were used to generate personal exposure indices for each household. The environmental data were then integrated with the health data to determine any associations with the treatment plant source. Regression analyses were performed between total viable particle exposure indices and self-reported illness rates, pathogenic bacteria isolation rates, prevalence rates of virus antibody, and virus antibody titers. An attempt was also made to determine if various subpopulations were at risk to infection. Regression analyses between illness rates and exposure indices were run with reference to length of residence, age, smoking, presence of young children in the family, chronic respiratory disease, and chronic gastrointestinal problems.

The results from all of these analyses were negative. No associations were found between any of the health factors and the treatment plant as an exposure source. However, the investigators cautioned that the overall conclusion that the plant had no obvious health effect on residents must be tempered by the small number of people who were exposed to the highest pollution levels.

Summary of Wastewater Treatment Plant Aerosol Studies

The primary health effect findings of the above four studies are summarized in Table 4. All of these studies had good designs and were performed by competent investigators. The positive findings in Tecumseh were indeterminate--they correlated with socioeconomic status as well as with distance from the sewage treatment plant. In Schaumburg, higher gastrointestinal symptom and skin disease rates occurred in those nearest the plant. However, significant decreases of some symptoms and diseases also occurred in all three zones. The interpretation of these results is clouded by obtaining the self-reported illness information through a long-recall survey instrument and the

inherent variability and inaccuracy of that technique. However, objective measurements, through pathogen isolation from clinical specimens and through serology, were negative. In the Tigard study, the exposure was low, the population at risk was small, and absenteeism is not necessarily indicative of symptoms or illness. The investigators in the Skokie study cautioned against overinterpretation of their negative results because of low exposure and small population.

HEALTH EFFECTS STUDY OF SEWER AND SEWAGE TREATMENT PLANT WORKERS

One limitation of the wastewater treatment plant studies of community populations has been low exposure. Residential areas in these studies were generally 400 m or more from the aerosol source and were not necessarily in a predominant downwind direction. Also, it is difficult to estimate the amount of exposure residents are subjected to in their homes near a treatment plant. Presumably, the population with greatest direct exposure to wastewater pathogens would be sewer maintenance and sewage treatment plant workers. With this idea in mind, the University of Cincinnati initiated a study (2,9) in 1974 of wastewater workers. The study subsequently continued for more than five years and additional analyses are still being done.

More than 500 workers in three cities (Cincinnati, Chicago, and Memphis) were recruited. The workers were divided into three broad categories: inexperienced and experienced wastewater exposed, and controls. A total of 336 workers remained with the study for the minimum 12-month requirement (Table 5). Inexperienced workers were those just beginning employment. To be placed in the experienced category, a worker had to have been on the job for a minimum of two years. The control groups consisted of highway maintenance workers in Cincinnati, water treatment plant workers in Chicago, and utility workers in Memphis.

Health monitoring included maintenance of an illness diary, examination of employer absentee and illness records, annual multiphasic physical examinations, and pathogen isolation attempts from stool specimens and throat swabs. Blood specimens were collected quarterly for subsequent serological analyses. A serologic survey was also conducted on the families of 82 wastewater and 41 control workers to determine possible associations with transmission of infectious agents to the home from the job. Limited aerosol and wastewater monitoring for indicators and pathogens was conducted in an attempt to refine exposure categorization.

The results of the illness analyses indicated no significant difference in illness rates by worker group or by city although gastrointestinal illness rates were two to four times higher in the inexperienced worker group. Combining the worker groups from the three cities did result in a statistically significant difference ($p = .004$) in gastrointestinal illness rates (Table 6). Rates were higher in the inexperienced group and there was no difference between experienced workers and controls. A seasonal peak during April-June was observed. The GI illness rates for the inexperienced group were analyzed

on the basis of time on the job and age of workers but no significant differences were detected.

There were no significant differences in virus or bacterial isolation rates among workers in the three cities although Salmonellae were isolated from sewage-exposed workers on six occasions (Table 7). One isolate was from an inexperienced worker at the time employment began; one was from another worker after one year on the job, and the remaining four were from experienced sewage treatment plant workers. One Shigella isolate was obtained from a control worker. There was a significant difference in parasite isolation rates--10 isolations were made, all from unexposed individuals.

The serologic analysis included a determination of immunoglobulin levels on the hypothesis that individuals exposed to low levels of microorganisms may develop higher levels of immunoglobulins. However, they were not found to be consistently higher in the sewage-exposed workers in any of the three cities. The virus serologic analysis involved comparing the geometric mean antibody titers, titer level changes (increases and decreases), and cumulative seroconversions among the worker groups in the three cities. A total of 594 comparisons were made, and based on chance alone, one might expect about 30 of these to be significant at the $p = .05$ level. Twenty-nine significant differences were found and they were distributed evenly among the exposed and control groups.

To improve the chances of detecting an effect, the inexperienced and control workers were further subdivided into low and high exposure categories on the basis of job observation and environmental monitoring. The virus serological results were then analyzed on a city-by-city and on a combined basis. A total of 510 comparisons were made and 23 of these were found significant. Nine out of 10 for the city-by-city comparison, and 10/13 for the combined analysis were in the direction indicating a sewage exposure effect and these were about equally divided between aerosol-exposed workers and sewage liquid/solids-exposed workers.

To summarize this study, inexperienced workers reported higher rates of gastrointestinal symptoms than did experienced workers or controls. These rates could not be related to a specific agent or exposure. The symptoms were mild and transitory and did not result in time lost from work. Pathogen isolations did not indicate any increased risk from sewage exposure.

STUDIES OF WASTEWATER SPRAY IRRIGATION HEALTH EFFECTS

The transmission of sewage pathogens through aerosols at spray irrigation sites is a potential route of exposure where effluent disinfection may be considered as a treatment. In the United States, the lack of suitable exposed populations at such sites has prevented conducting health effects studies of spray irrigation. However, studies have been and are being conducted in Israel where this practice has been in use for many years, and a study is now in progress in Texas (Table 8).

In the Israeli study reported in 1976 (7), the investigators compared Ministry of Health communicable disease data from 77 kibbutzim (agricultural communities) using partially treated nondisinfected oxidation pond effluent with that from 130 kibbutzim not practicing wastewater irrigation. For certain infectious diseases, they found incidence rates 2 to 4.3 times higher in the kibbutzim utilizing wastewater for irrigation (Table 9). The agents of these diseases are found in wastewater and transmission by this route is logical. No significant differences were found for diseases not considered to be transmitted by wastewater, such as streptococcal infections, tuberculosis, and laboratory-confirmed influenza. In addition, there were no significant differences in enteric disease rates among kibbutzim during the nonirrigation season. The investigators recommended disinfection of sewage effluent used for irrigation near residential areas because of the potential public health risks.

This study did not provide any evidence for an aerosol route of transmission. The irrigated fields were located 100 to 3,000 m from the residential areas. It was indicated that pathogens could reach the community on the bodies and clothes of the field workers when they returned at mealtime and at the end of the day. The quality of the drinking water was reportedly good and a food-borne route was discounted because regulations did not permit use of sewage to irrigate vegetables or other crops for raw consumption.

In an attempt to get more detailed information, another retrospective study of kibbutzim examined age-illness distribution, the quality of reporting, crop types and irrigation schedules, distance of fields to residences and dining halls, and length of irrigation season. One group of 13 kibbutzim was in a switch category, i. e., they used effluent irrigation for two consecutive years and then switched to non-effluent sources for another consecutive two years, or vice-versa. A second group of 68 kibbutzim was divided into effluent irrigating, effluent use in fish ponds, and non-effluent irrigating categories.

Two preliminary analyses of the results have been reported thus far (4,9). In the switch category kibbutzim, a significant increase was found in the relative risk of enteric disease during effluent-irrigation years only in the 0-4 age group. In the group of 68 kibbutzim, a slight excess of enteric diseases was found in kibbutzim using effluent in fish ponds. Although there was no difference in annual enteric disease rates between effluent and non-effluent irrigating kibbutzim, there were increased seasonal rates (May-July), coinciding with the irrigation period, in effluent irrigating kibbutzim. These rates fell below those in the non-effluent irrigating kibbutzim in the fall, thus accounting for the similar annual rates. Significant increases were noted for shigellosis and streptococcal sore throats in effluent irrigating kibbutzim. Streptococcal sore throats are not considered to be associated with a wastewater mode of transmission. There was no relationship of the enteric disease rate to source of effluent (own or others), size of the irrigated tract, or distance from residences, although an excess of enteric disease was noted for kibbutzim irrigating with effluent volumes >5600 m^3/year. These investigators also found that numerous kibbutzim

in the 1976 study (7) were incorrectly classified as to effluent utilization. They suggested that no firm conclusions on the degree of health risk should be based on either that study or the 1981 retrospective study because of the poor quality of the data (4).

A third study in progress in Israel is scheduled for completion in 1983. The quality of data in this prospective study is expected to be much improved over the previous two retrospective studies, especially for illness reporting. In addition, high risk sub-populations such as field workers and visiting volunteer groups, will be specifically followed serologically and through illness monitoring.

The only spray irrigation health effects study presently under way in the United States is being conducted near Lubbock, Texas. About 7.4 MGD of unchlorinated secondary effluent from Lubbock is being piped to 3,000 acres of farmland 18 miles southeast near the town of Wilson. Construction of the pipeline and installation of 22 center-pivot spray rigs was completed in 1981. About 450 people, including about 40 persons in the farm families living on-site, have been participating in a health watch. Baseline environmental and health data have been collected over a two-year period. Spray irrigation has started and one year of the same types of data will be collected. The project is scheduled for completion in 1984.

DISCUSSION

In the studies described above, conscientious efforts were made at site selection and study design. The projects were run by competent investigators from respectable institutions. In the wastewater treatment plant studies, some effects were noted, but they could not be conclusively associated with the treatment plant source. These studies all had two major limitations that make it difficult to attach any significance to either the positive or the negative findings: (a) low numbers of highly exposed persons, and (b) the inability to adequately and quantitatively determine that exposure. There is presently no suitable indicator for airborne pathogens from a sewage source and populations are subject to exposure to the same pathogens through other routes in the community.

The Israeli report in 1976 (7) appeared, at first glance, to produce some clear evidence of health effects associated with spray irrigation of wastewater. The results led the investigators to recommend disinfection of wastewater applied near residential areas. Subsequent investigations by one of the original authors have not been able to confirm those findings. In fact, many of the kibbutzim were incorrectly classified with regard to wastewater usage. Also, it has been discovered that a number of the kibbutzim may exceed even the liberal Israeli drinking water standard of 10 coliforms/100 ml. If there is a wastewater-related effect, it may be due to contamination of drinking water or to person-to-person transmission peculiar to the kibbutz communal way of life.

The two current studies of spray irrigation health effects should

provide useful additional information to impact the decision on whether or not to disinfect wastewater. However, these results will not be available for up to two and a half years. In a discussion of his paper on bacterial aerosols at a spray irrigation site, Sorber (8) indicated that terminal disinfection would be more effective and economical than buffer zones if such were considered necessary. He concluded that a safeguard of some type would be prudent until an adequate public health risk assessment can be made. Until such time, it may be necessary to consider each particular situation on a case-by-case basis.

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Table 1. Health Effects Studies of Populations Near Activated Sludge Treatment Plants

Plant Location	Exposed Population	Reference
Tecumseh, MI	Community	3,9
Schaumberg, IL	Community	5,9
Tigard, OR	Grade School	6,9
Skokie, IL	Community	1,9

Table 2. Partial Listing of Health Survey Results From the Schaumburg, IL Study

Disease or Symptom	Percentage Incidence	
	Baseline	Operational
Skin disease		
0-2 km[a]	0.5	1.7
2-3.5 km	1.6	1.3
3.5-5 km	1.4	1.4
Chest pain on deep breathing		
0-2 km	0.5	1.9
2-3.5 km	1.6	1.5
3.5-5 km	1.4	1.1
Diarrhea		
0-2 km	4.1	7.6
2-3.5 km	4.3	4.8
3.5-5 km	4.8	4.3
General Weakness		
0-2 km	0.7	1.9
3.5-5 km	1.5	0.6
Nausea		
0-2 km	1.2	3.0
3.5-5 km	3.0	1.7
Vomiting		
0-2 km	1.3	3.1
2-3.5 km	3.0	1.4

[a]Participants lived within the indicated distance from the plant

Table 3. Absenteeism at Durham Elementary and Control Schools

School	Absenteeism (%) Preoperational[a]	Operational[b]	Change
Durham	5.36	4.67	-0.69
Controls	4.96	4.64	-0.32

[a]Last 2 years prior to plant operation

[b]First 2 years of plant operation

Table 4 Summary of Health Effects From Four WWTP Epidemiological Studies

Study	Health Effect	Comment
Techumseh, MI	Higher respiratory, GI illness within 300 m of plant	Indeterminate cause; socioeconomic confounders
Schaumberg, IL	Higher GI & skin disease rates within 1 km of plant	Long-recall survey; objective measurements negative
Tigard, OR	Negative	Small population; low exposure; absenteeism, not illness
Skokie, IL	Negative	Small population; low exposure

Table 5. Number of Workers Remaining in the Study a Minimum of 12 Months

City	Worker Group Inexperienced WWE[a]	Experienced WWE	Controls
Cincinnati	35	94	61
Chicago	38	35	27
Memphis	27	0	19
Total	100	129	107

[a]WWE = wastewater exposed

Table 6. Seasonal Comparison of Gastrointestinal Illness Rates for Combined Three-City Groups

Season	Illness/100 Worker-Months Exposure		
	Inexperienced WWE[a]	Experienced WWE	Controls
Jan.-Mar.	3.6	1.8	1.3
Apr.-June	5.7	2.0	1.6
July-Sep.	2.9	2.0	1.9
Oct.-Dec.	2.6	1.7	1.4

[a]WWE = wastewater exposed

Table 7. *Salmonella* and *Shigella* Isolations From Workers

Worker Group	No. of Isolations	
	Salmonella	*Shigella*
Inexperienced WWE[a]	2[b]	0
Experienced WWE	4	0
Controls	0	1

[a]WWE = wastewater exposed

[b]One isolate from initial employment specimen; one isolate from another worker after 1 year on job

Table 8. Health Effects Studies of Populations Near Wastewater Spray Irrigation Sites

Location	Exposed Population	Reference
Israel (1976)[a]	Community	7
Israel (1981)	Community	4,9
Israel (1983)	Community; workers; volunteer groups	-
Lubbock, TX (1984)	Farmers; rural and town populations	-

[a]Date of completion or expected completion of study

Table 9. Summer Incidence of Infectious Diseases in Kibbutzim With and Without Spray Irrigation

Disease	Incidence/100,000		
	WW Irrigation (A)	No WW Irrigation (B)	A/B
Shigellosis	1002	455	2.2
Salmonellosis	234	63	3.7
Infectious hepatitis	88	44	2.0
Typhoid fever	11.6	2.7	4.3

REQUIREMENTS FOR WASTEWATER DISINFECTION AS SEEN FROM THE RESULTS OF EPIDEMIOLOGICAL-MICROBIOLOGICAL STUDIES

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ABSTRACT

The United States Environmental Protection Agency in 1976 abandoned its policy requiring universal microbial standards for municipal wastewater effluents discharged into fresh and marine waters and, hence, the requirement for universal disinfection of these effluents. It was replaced by a policy in which the microbial limits and the need for and level of disinfection are determined on a case by case basis. A flow diagram, with feed-back loops, of the informational needs in making such decisions is presented. It starts with a target area criterion and ends with the balance between treatment and disinfection and outfall location.

One of the needs (also a feed-back for risk acceptability), a site-specific, cumulative frequency distribution of swimming-associated illness rates, was obtained for beaches along the New York Bight. The inputs to the model used in making these predictions were the illness (gastroenteritis) - indicator (enterococcus) regression equations obtained from the bathing beach epidemiological program and the frequency distribution of enterococcus densities at sampling stations near the beaches. The rates for "posted" and "open" beaches were then compared to the predicted enterococcus densities and illness rates (calculated by the application of the regression equation to the enterococcus densities in primary and secondary sewage effluent) following various treatment, initial dilution and subsequent transport decay options. This preliminary analysis indicated that, with primary treated effluents, disinfection would probably be required in the absence of the option for long, deep ocean outfalls and that, in many, if not most, situations, this would also be true of secondary effluents.

INTRODUCTION

There would be no argument against universal disinfection of wastewater effluents to levels which virtually eliminate all pathogenic microorganisms therein if there were a relatively inexpensive, energy-efficient, facile, reliable and effective disinfection system which produces minimal or no adverse ecological or human health effects. In fact, there was a time in the early 1970's when universal disinfection (specifically chlorination) of wastewater effluents was considered a reasonable requirement by the United States Environmental Protection Agency (USEPA) as a means of attaining one of its objectives, to make all waters "swimmable and

fishable" (8). The corollary to this requirement was that the coliform and fecal coliform limits for the target (stream standards) would be applied at the source (effluent). This objective, as applied to the microbial target area standards or guidelines most commonly used by the various states or recommended by the Federal Government (16) could be achieved even in primary treated effluents by chlorination to "reasonable" levels (18). Moreover, the need for relatively restrictive microbial standards for shellfish growing waters and hence for sewage disinfection, at least in certain circumstances, was firmly supported by the history of shellfish-associated outbreaks of disease during the preceding several decades (22) although the epidemiological information in support of microbial standards for bathing waters was more limited and less compelling (12). Finally, it was generally accepted that coliforms were a reasonable surrogate for salmonellae and the other "important" pathogens as regards the effectiveness of disinfection, although there was some evidence that at least one of the agents in question was viral (22)and that viruses were generally more resistant to chlorination than the coliforms (13).

However, by 1975, it had become clear that, for a number of reasons, the requirement for universal disinfection was no longer realistic. First, it was shown that adverse ecological effects could be, and presumably were, produced from the chlorination of sewage effluents (19) although the quantitative relationship of the levels producing adverse ecological effects to those required for "adequate" disinfection were not defined. Second, the demonstration of the carcinogenicity of some chlorinated organics produced during the chlorination of municipal wastewaters raised the possibility of adverse human health effects from the movement of these compounds up through aquatic food chains. Third, the energy crisis of 1973 increased the awareness that both the quantity of energy required to produce the chlorine and its cost could not be ignored in decisions on how, when, and where to chlorinate. Fourth, additional data were obtained showing that coliforms were not a good surrogate for viruses with regard to chlorination (20). Finally, although the early findings from an epidemiological program conducted by the USEPA clearly showed health effects (a gastroenteritis) consequent to swimming in waters having relatively low indicator densities, they also suggested that the etiological agent was viral and indicated that total and fecal coliforms were defective as recreational water quality indicators (6). In effect, chlorination did not meet the requirements of a wastewater disinfection process which could be applied universally; and, at that time, there was no practical alternative.

Because of the considerations noted above, in 1976 the USEPA reversed its position and abandoned the requirement for universal effluent standards and, hence, universal disinfection. It was replaced by a policy whereby the requirement for and extent of disinfection would be made on a case by case basis with regard to all the factors involved. Some of us within the Agency who advocated the change also did so on the grounds that the existing policy was conceptually deficient. First, there was no provision for the effects of dilution, sedimentation or biological decay (die-off) in reducing the levels of pathogenic or indicator microorganisms between

the effluent source (outfall) and the potential targets (bathing beaches, shellfish growing areas, and raw drinking water inlets). Second, as noted above, the requirement for universal disinfection derives from uniform source and, hence, target indicator standards. The promulgation of a single standard applied on a nationwide basis does not provide for local input on risk acceptability.

INFORMATIONAL NEEDS FOR WASTEWATER DISINFECTION

The achievement of the balance between the need for wastewater disinfection and its undesirable consequences, along with the change in USEPA policy, made it even more important to obtain the data bases needed in determining the required level of wastewater disinfection on a case by case basis. The informational requirements for doing so are illustrated in Figure 1. The starting points are health effects criteria. They are mathematically expressible relationships between the predicted rates of infectious disease among the users of the sewage impacted aquatic resources and some measures of the qualities of the resources. The resources in question are bathing beaches (including areas used for water skiing, surfing and other direct contact activities), shellfish growing areas, and the raw sources of drinking water. Of necessity, the criteria are generalizations which should be reasonably applicable over extended periods of time to large geographic areas since the cost of their development is considerable. However, both temporal and spacial variability in the relationships can occur due to several factors (e.g. the incidence of illness in the "discharging" population, the immune status of the users)(3). Moreover, relationships based upon fecal indicator densities in waters impacted by small wastewater discharges are not reliable (3). Only one such criterion is currently available, that for saltwater bathing beaches, although a similar one for fresh water bathing beaches will be described in the next paper. The marine recreational water quality criterion was developed from a series of prospective epidemiological studies conducted over multiple years at several locations in the United States. It relates the incidence of swimming-associated gastroenteritis to the enterococcus density in the bathing water. The importance of these two specific inputs with regard to wastewater disinfection will be considered later in this paper (4).

Guidelines and standards can be derived from a criterion once a decision is made as to incidence of illness which is considered acceptable ("acceptable risk") as illustrated in Figure 2. This decision has economic, sociological and political inputs at both the national and local levels. The guidelines and standards for all the potentially impacted targets in the area can then be translated into effluent standards using as inputs estimates of the physical and biological decay of the pathogenic microorganisms or appropriate surrogates during transport between the source and the potential targets. The final decision concerns the trade-off between treatment and disinfection and outfall location needed to achieve the effluent standard for that specific pollution source.

There are three feedback loops in the system (indicated by broken lines in Figure 1). First, the choice of the outfall location will influence the physical and biological decay inputs needed in the translation of target to source standards. Second, the costs and consequences of wastewater treatment and disinfection and outfall location can be inputs towards determining the acceptable risk of disease among the users of the impacted resources. Third, once the wastewater treatment, disinfection and disposal system is in operation, the decisions on the acceptable risk can be reexamined and modified from information on the frequency distribution of indicator densities at the target, resource usage, and the illness-indicator relationship.

OUTPUT FROM USEPA EPIDEMIOLOGICAL STUDIES

Four necessary pieces of information were obtained from the USEPA epidemiological-microbiological program to develop recreational water quality criteria (4,7) -- the next paper will describe a fifth. The first is the illness (swimming-associated gastroenteritis) -- water quality (enterococcus density in the bathing water) regression line. It predicts the former (Y) from the latter (X). The formula for the regression line shown in Figure 3 is $Y = 12.25 \log_{10} X + 0.073$. The second was information on the "best" indicator of those examined. It was defined as the one whose mean densities in the bathing water correlated the best with the swimming-associated rates of gastroenteritis. Table 1 shows the correlation coefficients (r) for four of the most commonly considered indicator systems. By this criterion enterococci was the best indicator. The third was the criterion (regression line and its confidence limits) itself. It predicts the mean enterococcus density in the water (X) from the "acceptable" swimming-associated gastroenteritis rate (Y). The formula is $\log_{10} X = 0.0456\ Y + 0.677$. The fourth was a membrane filter method for enumerating the enterococci which does not require the picking of colonies for identification (14). It was subsequently simplified even further (9).

SITE-SPECIFIC ILLNESS RATE PREDICTIVE MODEL AND ITS USE AT NEW YORK BIGHT BEACHES

A model for predicting the swimming-associated gastroenteritis rates (the second feedback loop noted above) was developed and applied in a study of bathing beaches along the New York Bight sponsored by Marine Ecosystems Analysis, National Oceanic and Atmospheric Administration. The detailed findings are being prepared for publication. Two of the three inputs to the model as noted earlier were obtained as follows. The illness-indicator regression line was obtained from the USEPA epidemiological study. The distribution of enterococcus densities --- *E. coli* data also were obtained by the mTEC method (10) --- was obtained from assays performed in the author's laboratory from water samples collected by the USEPA. They were collected by helicopter during the summers of 1980 and 1981 from just beyond the surf zone at 78 sampling stations located from Cape May, New Jersey around to the Shinnicock Inlet, Long Island, N.Y.

Indicator Density Frequency Distributions

The cumulative frequency distributions of enterococcus densities at some New Jersey, New York City (Staten Island and Coney Island), and Long Island sampling stations are shown in Figure 4. The number of values for each station varied from 17-26. However, at most of the New Jersey and Long Island stations, many of the enterococcus densities were below the sensitivity of the assay method, 0.5 per 100 ml. This made it difficult to fit straight lines to the distributions as shown. The E. coli densities were generally higher than those for the enterococci, especially at the New York City stations (data not shown).

Illness Rate Frequency Distributions

Each of the enterococcus density estimates per 100 ml (X) was used to predict a swimming-associated gastroenteritis rate/1000 persons (Y) using the formula given earlier. The cumulative frequency distributions of Y corresponding to the indicator distributions for some selected stations are shown in Figure 5. Enterococcus densities $\leq$ 0.5/100 ml yielded negative gastroenteritis rates; these were recorded as 0s. There were 27 New Jersey and 8 Long Island stations where no more than one positive Y value was obtained. With rare exceptions, the distributions of illness rates predicted from the E. coli densities were higher than those predicted by the enterococci, although the slopes of the latter generally were greater than those of the former (data not shown).

Three percentile values for the predicted illness rates were selected as being especially informative and useful (75, 90, and 95) in that they could provide an individual some idea of the risk of gastroenteritis incurred while swimming at a particular beach. The rates are presented for the stations already considered and a few others in Table 2. For example, the prediction is that, at station J-93 near Wildwood, the gastroenteritis rate will not exceed 11.3/1000 swimmers more than 5 percent of the time, 6/1000 more than 10 percent of the time, and 0.0 more than 25 percent of the time. The comparison of the 75 and 95 percentile values provides some idea of the relative slopes of the distributions and hence the constancy of the risk from time to time. This can be seen from the comparison of the values for stations J-93 to J-97, LI-2 to LI-4 and SI-Sou to CI-MB. It is of interest that the 75 percentile rates exceeded 0.1/1000 at only seven stations J-97, SI-Sou, CI-35, CI-29, CI-20 LI-4, and LI-16. South Beach on Staten Island and W. 35th Street on Coney Island are posted as unsafe for swimming. The data presented would suggest no greater justification for closing South Beach than that for those beaches at 29th and 20th Streets on Coney Island. However, as noted earlier the acceptability of risk has other than illness inputs. LI-16 is at an inlet to Great South Bay, and there apparently are some marginally treated discharges near Cape May, N.J. (J-97).

The disparity in the illness rates predicted from the enterococcus

and E. coli densities can be seen from Table 3, which compares the predicted rates at the 35 (27 + 8) stations noted earlier.

Beach usage data were obtained from the project and are still being analyzed. Once the output, the seasonal number of swimmers at each beach, is obtained, the more useful information for the managers of the bathing resources will become available, i.e. the predicted annual number of cases of swimming-associated cases of gastroenteritis at the three percentile levels for each beach area.

PREDICTED DISCHARGE OPTION ILLNESS RATES

Not all the necessary inputs for making the decisions on the required level of wastewater disinfection are available and some must be determined from site specific data (e.g. decay coefficients). However, some insight can be obtained by an examination of the information presented against one further input, the enterococcus densities in primary and secondary treated sewage. The mean $\log_{10}$ densities for the influents and the primary and secondary treated effluents as determined at a number of sewage treatment plants in Rhode Island were 5.45, 5.32, and 3.94, respectively. Table 4 shows the expected densities following initial dilutions at the "boil" of 1:10, 1:50, and 1:100 followed by reductions of 90, 99, and 99.9 percent (1,2, and 3 orders of magnitude, respectively) during transport between the boil and the target. The residual densities then were used to predict the mean swimming-associated gastroenteritis rates (Table 5) using the appropriate illness-indicator regression equation.

Comparison of Predicted Discharge Option and Bight Beach Illness Rates

These mean rates predicted from the treatment-discharge options can then be compared to the 75 percentile values (much less the 50 percentile values) for those stations associated with beaches which are and are not posted as being unsafe for swimming according to the local guidelines and standards. The beaches and "associated" 75 percentile values for those beaches which are posted are South Beach, Staten Island (1.69) and W. 35th Street, Coney Island (3.76). The beaches with high 75 percentile values which are not posted are Wildwood (3.38), W. 29th Street, Coney Island (1.69), W. 20th Street, Coney Island (4.27), 67th Street, Rockaways (2.23), and Cedar Island Beach, Long Island (2.23). If a 75 percentile value of 2.7 is used as the "break-point" (the average of the two posted and of the five unposted beaches/stations), then one could make the following inferences. With primary sewage effluent, it would appear that initial dilutions slightly in excess of 1:100 and/or subsequent reductions slightly in excess of 99.9 percent would be required as an alternative to disinfection. These can only be obtained from the discharge through long distance, deep outfalls such as those in place along the Pacific Coast. With rare, if any, exceptions, this alternative is not available along the Eastern Seaboard or the interior. The alternative would be the sacrifice of some nearby and not so near resources. With secondary treated effluents, subsequent reductions of

slightly less than three logs, more than two, and less than two would be required with initial dilutions of 1:10, 1:50, and 1:100 respectively.

DISCUSSION

The predicted, beach specific, swimming-associated illness rates presented herein were meant to demonstrate the use of the model as one means of evaluating the water quality at bathing beaches under existing conditions. As noted earlier, the output information can then be used in risk assessment and, as required, the modification of the specific treatment, disinfection, and disposal strategies for the wastewater discharges reaching the beaches. The accuracy of the specific predictions made herein was limited by the quality of the input data; and this was due to logistic constraints on the intensity of the sampling effort. First, the sampling stations should be chosen with regard to the spacial distribution of the swimmers at the beach; this was neither logistically feasible nor necessary in the present study. Second, depending on the length of the beach, two or more samples should be collected; in some instances, at least, the samples could be pooled prior to assay. Third, at least 25 samples should be collected from each sampling station. However, all the above deficiencies can be rather easily corrected in a local effort of more limited geographic scope, especially since the membrane filter assay method for enterococci is relatively facile. Moreover, because of the nature of the gastroenteritis enterococcus regression line, the assay sensitivity need not exceed 1/100 ml. There also are a number of conceptual limitations on the use of any fecal indicator in predicting water-related health effects; and these were considered in an earlier publication (3).

The adequacy of even the enterococci as a health effects water quality indicator also needs to be addressed. There can be little doubt that it was the best of those indicators examined in USEPA epidemiological-microbiological studies with regard to an illness (gastroenteritis) whose etiological agent(s), in all probability, was viral (5). Furthermore, there is increasing evidence that enterococci better simulate the survival characteristics of certain viruses than do the coliforms, at least in sludge (1) and during transport in marine waters (21); and coliforms are much more sensitive to chlorination than most animal and bacterial viruses (19). First, in the one epidemiological study in which the presumed source of the etiological agent was at the greatest distance away (in time), the swimming-associated rates of gastroenteritis were disproportionately high relative to the enterococcus densities (4,5). Second, one explanation for the relatively high illness rates associated with very low enterococcus densities (Figure 3) is differential survival of the etiological agent and the indicator. The third cause for concern is related to the second and will become apparent from the next paper. Fourth, coliphages (notably male-specific phages such as f-2 and Fd) were not examined as possible viral surrogates of the viral pathogens -- they are not called a fecal indicator because they are consistently found in sewage but not feces (15) -- in the epidemiological studies because of methodological problems. This was unfortunate since

field data collected in our laboratory (15) showed that the coliphages were much more resistant to chlorination than coliforms, chlorination kinetic studies with phage stocks conducted by Scarpino (20) showed coliphages, especially the RNA male-specific phages f-2 and MS-2, were more resistant than E. coli, and some additional studies conducted in our laboratory (17) with phage stocks and the viruses as found in sewage showed that the DNA male-specific phages were even more resistant than the RNA male-specific phages.

Five additional data inputs are needed in determining the level of wastewater disinfection required on a case by case basis. The first, a criterion for fresh recreational waters has been developed and will be described in the next paper. The second is a similar criterion for shellfish growing waters. The third is the development and evaluation of the technology needed for obtaining biological decay coefficients at specific locations (i.e. with consideration to so-called "backyard effects") and without recourse to open water, in situ studies. These would be used as inputs to physical transport models which have been or are being developed. Incidentally, Clostridium perfringens spores appear to be an excellent conservative tracer for the conduct of such studies both for the examination of the water column (2) and the underlying sediments (11). The fourth is more information on the killing kinetics for enterococci in sewage during chlorination and other disinfection procedures. The fifth is a cost-benefit or cost-effectiveness model to be used in determining the acceptable risk of the water-related diseases.

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Table 1. Correlation Coefficients (r) for Gastroenteritis Against Mean Indicator Density New York City Study 1973-1975

Indicator	r_1 Values by Summ.[1]	Density[2]
Enterococci	.75	.96
E. coli	.52	.56
Fecal Coliforms	-.01	.51
Total Coliforms	.19	.65

[1] Trial days grouped by summer.

[2] Trial days grouped by indicator density.

Table 2. Predicted Rates of Swimming-Associated Gastroenteritis at Some New York Bight Beaches

Station	Location	Rate/1000 Swim. at Perc.[1] 75	90	95
J-24	Ocean Grove	0.00	0.07	4.71
J-75	Atlantic City	0.00	4.95	5.92
J-81	Ocean City	0.00	0.07	1.03
J-93	Wildwood, N. Wildwood	0.06	5.98	11.23
J-97	Cape May City	3.38	9.39	11.19
SI-SB	South Beach	1.69	6.51	12.15
CI-35	Coney Isl, W. 35th	3.76	7.05	17.01
CI-29	Coney Isl, W. 29th	1.69	8.04	16.57
CI-20	Coney Isl, W. 20th	4.27	9.34	16.98
CI-MB	Coney Isl, Manh. B.	0.00	2.26	12.34
LI-2	Riis. Pk., Rockaways	0.00	4.84	11.11
LI-4	92nd St, Rockaways	2.23	6.33	7.27
LI-8	Long Beach	0.00	2.23	4.27
LI-12	Jones Beach	0.00	0.07	0.07
LI-16	Cedar Island Beach	2.23	5.63	5.92
LI-18	Great South Beach	0.00	0.05	4.22
LI-26	Tiana Beach	0.00	3.76	10.43

[1]Swimming-associated gastroenteritis rate (Y) for given percentiles; calculated from applying regression equation $Y= 12.25 \log_{10} X + 0.073$ where X is the observed distribution (N= 17-26) of enterococcus densities/100 ml at indicated sampling station.

Table 3. Comparison of Swimming-Associated Gastroenteritis Rates Predicted from the Distributions of Enterococcus and *E. coli* Densities

General Area	No.[1] Stns	Indicator Used	Rate/1000 Pers. at Perc.[2] 75	90	95
New Jersey	27	Entero.	0.0	0.01	0.05
		E. coli	6.6	9.3	10.5
Long Island	8	Entero.	0.0	0.01	0.04
		E. coli	5.8	8.3	9.0

[1]Only includes stations where 95th percentile, swimming-associated gastroenteritis rate predicted from enterococcus densities did not exceed 0.05/1000 persons.

[2]See Table 2 for calculation of rates: formula for calculating *E. coli* rates, $Y= 6.32 \log_{10} X + 5.71$; values given are the averages from all the stations.

Table 4. Calculated Residual Enterococcus Following Hypothetical Reductions Due to Initial Dilution and Decay During Transport

Treatment	Initial Dilution	Log_{10} Residual/100 ml after Transport Reduction of 90%	99%	99.9%
Primary (5.31)[1]	1:10	3.31	2.31	1.31
	1:50	2.61	1.61	0.61
	1:100	2.31	1.31	0.31
Secondary (3.94)[1]	1:10	1.94	0.94	-0.06
	1:50	1.24	0.24	-0.76
	1:100	0.94	-0.06	-1.06

[1]Mean log_{10} enterococcus density/100 ml in sewage (18); influent density 5.45.

Table 5. Predicted Mean Swimming-Associated Gastroenteritis Rates/1000 Persons for Residual Enterococcus Densities Given in Table 4

Treatment	Initial Dilution	Predicted Illness Rate (Y)[1] After Transport Reduction of 90%	99%	99.9%
Primary	1:10	40.6	28.3	16.1
	1:50	32.0	19.8	7.5
	1:100	28.3	16.1	3.9
Secondary	1:10	23.8	11.6	0.0
	1:50	15.3	3.0	0.0
	1:100	11.6	0.0	0.0

[1]Gastroenteritis predicted from the formula $Y= 12.25 X + 0.073$, where $X= log_{10}$ enterococcus density/100 ml and Y is in cases per 1000 swimmers.

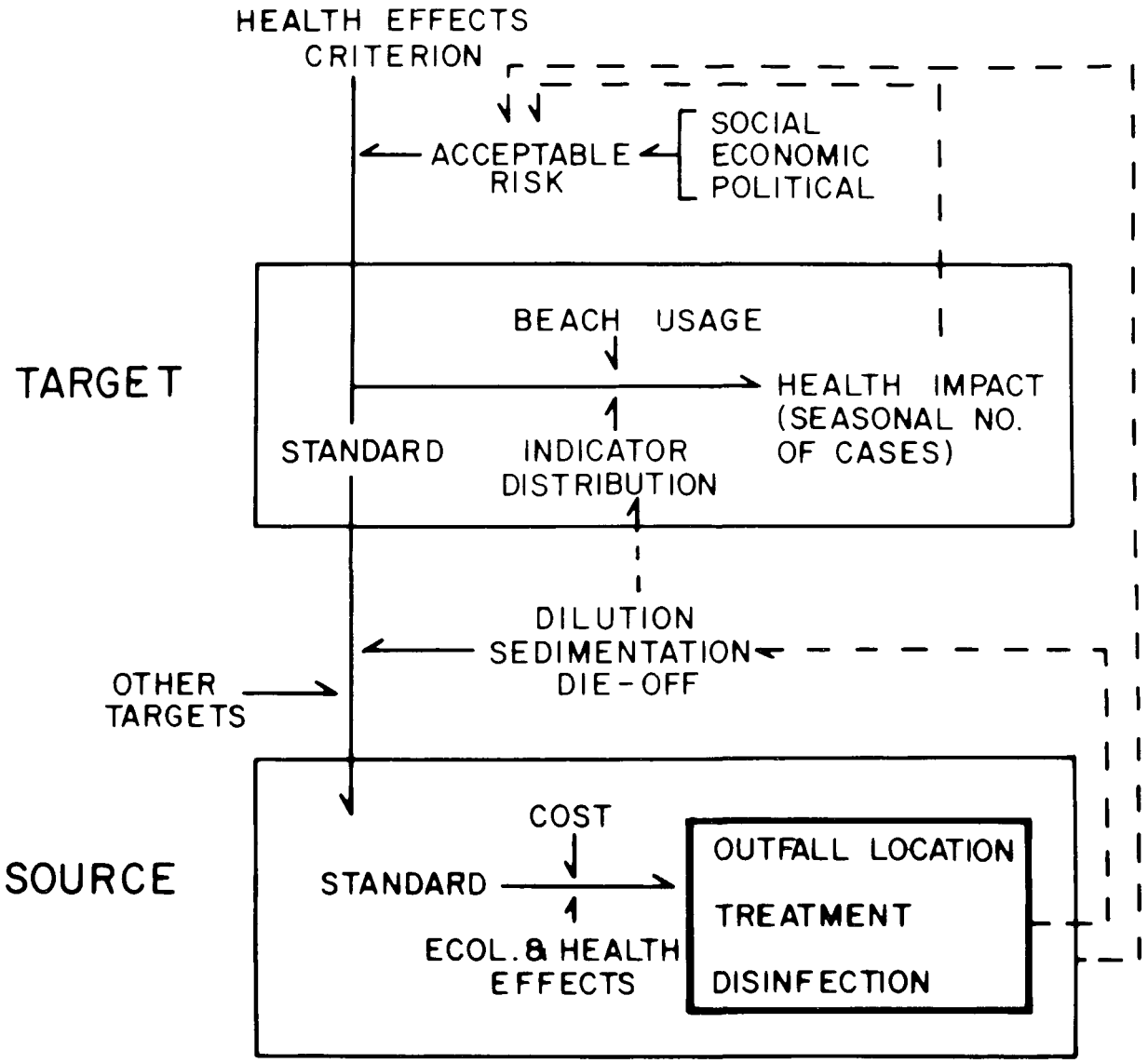

Figure 1. Information flow scheme for case-by-case decision making on the need for wastewater disinfection.

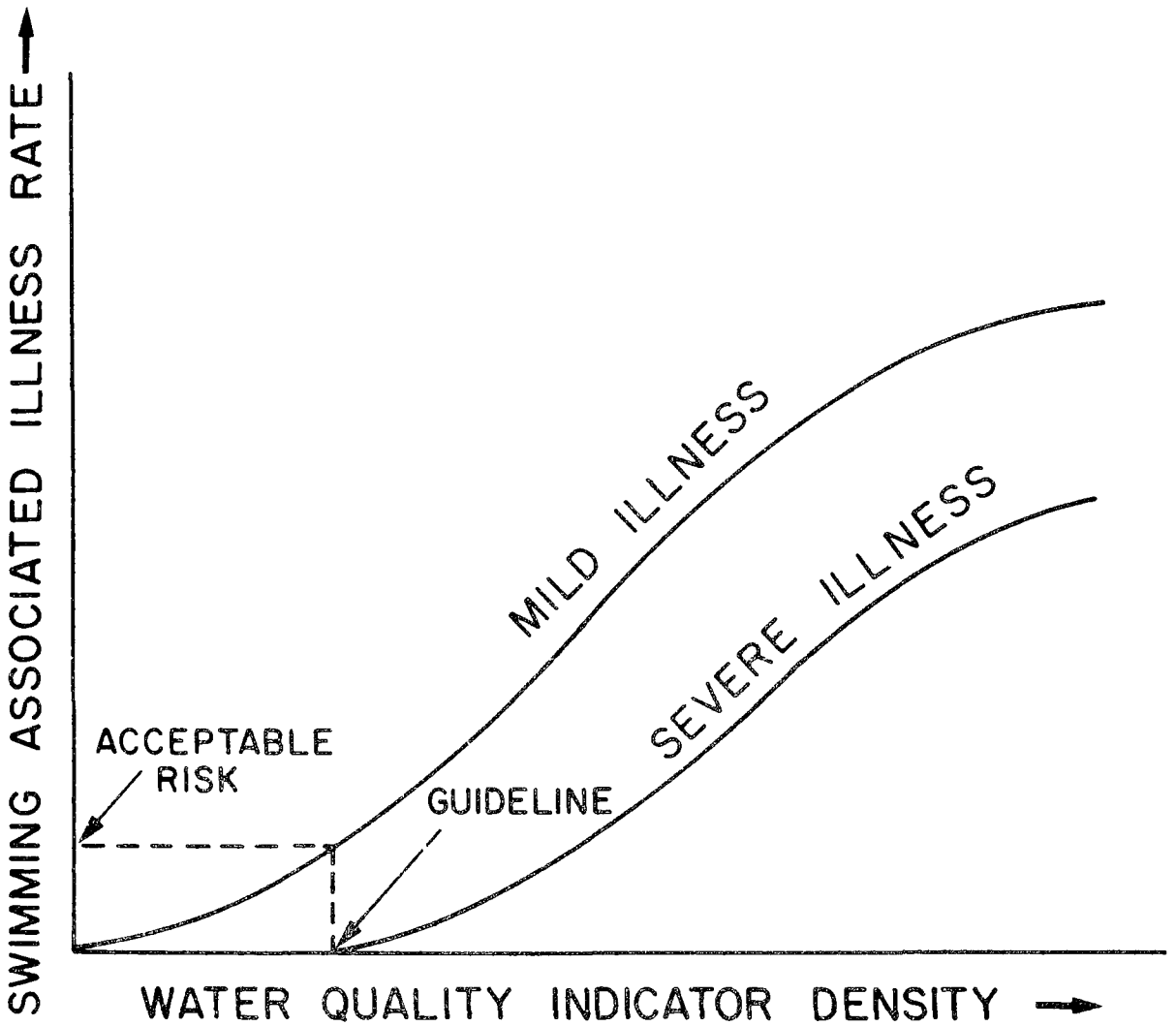

Figure 2. Relationship of criteria to guidelines and standards.

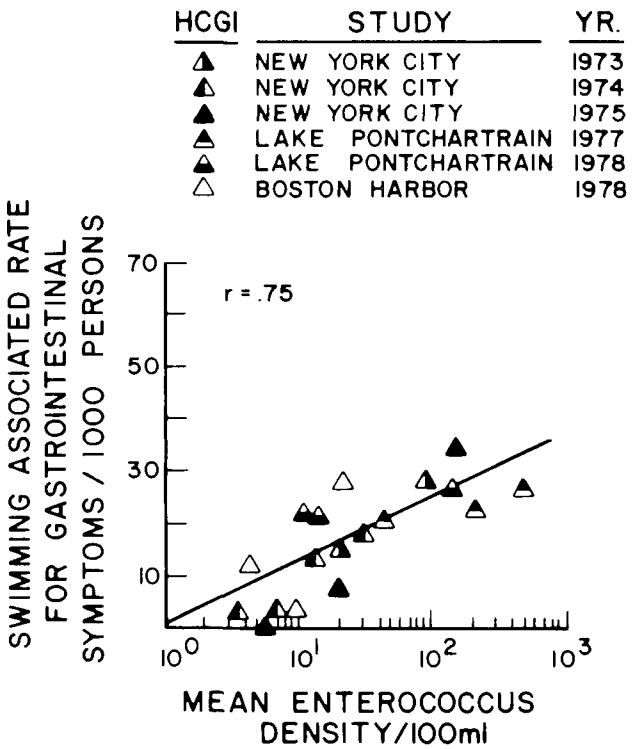

Figure 3. Relationship of the rates for swimming-associated, highly credible gastrointestinal symptoms (gastroenteritis) to the mean enterococcus densities in the water (see references 4 and 7 for more details).

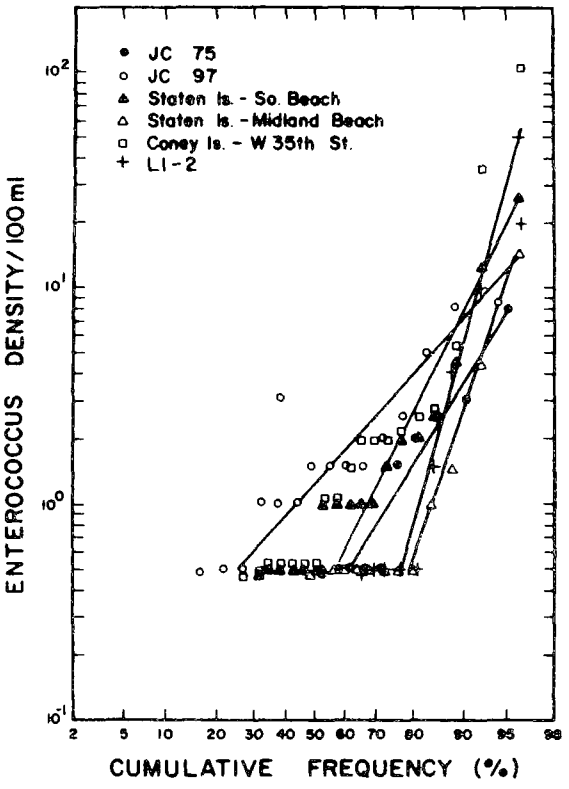

Figure 4. Cumulative frequency distributions for enterococcus densities at some New York Bight sampling stations.

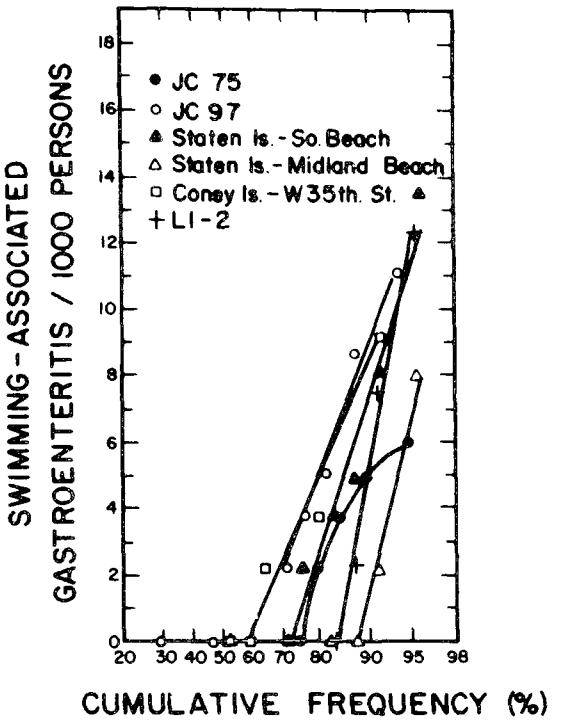

Figure 5. Cumulative frequency distribution of swimming-associated gastroenteritis rates predicted from enterococcus density distributions at some New York Bight sampling stations.

FRESH RECREATIONAL WATER QUALITY AND SWIMMING-ASSOCIATED ILLNESS

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ABSTRACT

Prospective epidemiological-microbiological studies were carried out at two freshwater bathing beaches, one at Erie, Pennsylvania and the other at Tulsa, Oklahoma. The purpose of these studies, which covered a two-year period, was to: 1) Examine the relationship between swimming-associated gastrointestinal illness and freshwater quality; 2) Determine if the water quality criteria established for marine bathing beaches are applicable to freshwater beaches; 3) Investigate the relationship between waterborne, microbe-bearing particulates and swimming-associated health effects.

Swimming-associated gastrointestinal illness was observed at freshwater beaches. In general, significantly greater illness rates occurred at barely acceptable beaches than at the relatively unpolluted beaches. The swimming-associated rate of gastrointestinal illness observed in freshwater swimmers was found to be appreciably lower than that observed in marine water swimmers. Finally, the preliminary evidence indicates that there may be a relationship between microbe bearing particulates and gastrointestinal illness.

Freshwater epidemiological-microbiological studies indicate that water quality criteria established for marine recreational waters may not be applicable to fresh recreational waters.

INTRODUCTION

Discussions about the need to regulate recreational water quality or chlorinated wastewater effluents that may ultimately reach recreational waters invariably lead to the question of whether or not wastewater-contaminated surface waters have the potential to cause illness in swimmers (23,28,34). This uncertainty about recreational waters being the vehicle of transmission for pathogens that cause enteric disease in swimmers has persisted because much of the evidence supporting the relationship was far from conclusive. The basis of this doubt can be found in the list of some of the most frequently referenced swimming-associated disease outbreaks shown in Table 1. Only two of the outbreaks present a reasonably strong case supporting the premise that the observed illnesses were due to pathogens from wastewater effluents. One was the Walmer outbreak in 1909 in England where young recruits swam in a pool filled with seawater contaminated with effluents from a nearby sewage treatment plant (29). The other was the Dubuque, Iowa outbreak which occurred

in 1979 (32). Thirty-one individuals were reported ill with shigellosis and the only common factor to all the illnesses was swimming in the Mississippi River. Water samples from the river, examined some days after the peak of the outbreak, were found to contain high densities of coliforms. The suspected pathogen was also isolated from the water. However, it was not unequivocably established that a sewage treatment plant 17 miles upstream was the source of the causative agent. In the other outbreaks the linkage between sewage-contaminated water and swimming-associated illness was quite tenuous. The United States reports of illness in swimmers were not very well documented, especially for the early outbreaks (18,19,29,30). Similarly, the evidence in the Australian (2,14) and French (12) outbreaks was not conclusive.

Since the study of outbreaks was found to be an unsatisfactory means to show that swimming in polluted water is a health hazard, the epidemiological approach was examined. Table 2 lists four epidemiological studies that have been conducted since 1950 The conclusion of the 1959 retrospective study reported by Moore (27) was that an association between poliomyelitis and swimming in poor quality water as a causal factor could not be found. This finding has frequently been used to justify the case against regulating recreational water quality and disinfecting wastewaters. However, the proponents of this position seldom take into account that negative findings in retrospective studies should not be interpreted to mean that the relationship does not exist, but rather that the case is not proven. The results of the 1981 retrospective study conducted by D'Alessio et al. (11) clearly show the correctness of this interpretation. They found an increased risk of enterovirus-caused illness in children who swam in lake water. The risk of illness due to swimming in wastewater-contaminated waters was further substantiated in the two prospective studies listed. The United States Public Health Service studies reported by Stevenson (37) in 1953 concluded there was a risk of enteric illness associated with swimming in polluted fresh waters. However, these studies have been criticized on a number of issues, such as the adequacy of study design and the way in which the data were analyzed. As a result of these criticisms the United States Environmental Protection Agency (EPA) initiated a series of epidemiological-microbiological studies at marine bathing beaches that were designed to correct the major deficiencies of the studies reported by Stevenson. The results of the EPA studies have been reported by Cabelli (8). These studies have unequivocably established that there is a risk of gastrointestinal illness associated with swimming in polluted recreational waters and that this risk increases as the water quality decreases. Furthermore, the studies showed that the enterococcus group was the most efficient indicator of marine recreational water quality from which a prediction of the rates of swimming-associated illness can be made.

Although the criterion or model established by the Cabelli studies offers conclusive evidence that marine recreational waters contaminated by sewage effluents are a vehicle for transmitting enteric illness to swimmers, some questions remain to be answered. For instance, it is not known whether the model established with marine bathing beach data is applicable to freshwater beaches. Another obvious question is why do statistically significant swimming-associated illness rates occur in apparent high quality water. An

example of this is the significant swimming-associated rates of highly credible gastrointestinal illness observed when the density of Escherichia coli in the water at New York City beaches was only 14 per 100 ml (8). The data to be presented here will attempt to shed some light on both of these questions.

Freshwater Epidemiological Studies

Freshwater studies similar to those conducted at marine beaches were initiated in 1978. Several freshwater sites were surveyed to determine their potential for full-scale epidemiological-microbiological studies. Two sites were found to be suitable. One at Keystone Lake, a man-made lake 15 miles from Tulsa, Oklahoma and the other on Lake Erie at Erie, Pennsylvania. Each site had two beaches whose beach-going populations were demographically similar, but whose water quality was significantly different. Two groups of investigators, one at the University of Oklahoma led by Dr. James Robertson and the other at Gannon University led by Mr. Stan Zagorski, were supported through grants from the Environmental Protection Agency in 1979 and 1980 to carry out the epidemiological-microbiological studies. The data presented here were supplied by the respective principal investigators who are preparing manuscripts for publication that will describe the studies in detail.

Experimental Procedures

Although the epidemiological protocol has been described elsewhere (8,9,10), a brief summary of the illness inquiry sequence of events is given in Table 3. The freshwater trials closely followed the procedures used during the marine beach studies to insure comparability of the data. However, the method of data analysis had to be modified because the swimming activity of freshwater swimmers differed from that of marine swimmers. Freshwater beach goers, unlike those at marine beaches, had a tendency to do a great deal of swimming and therefore only a limited number of non-swimming beach goers at any one beach were available to serve as control subjects. Since the non-swimming beach goers at each study site were demographically similar, the non-swimmers from both beaches at each study site were combined to form a single control group.

The follow-up telephone survey obtained information on a number of symptoms that might have occurred during the 9- to 10-day interval between the swimming activity and inquiry Gastrointestinal symptoms and disabling information were used to generate two variates reported in the marine water studies. Definitions of the variates are shown in Table 4. Since symptoms were self-diagnosed, multiple symptoms or unmistakably recognized symptoms were used to establish the credibility of the gastrointestinal illness. These were called "highly credible" symptoms. Swimming-associated illness rates were determined by subtracting either the total or "highly credible" symptom rates observed in the non-swimming control groups from the respective symptom rates observed in the swimming groups. These swimming-associated symptom rates were used to establish health effects/water quality relationships from the freshwater data.

Microbiological Methods

Multiple indicators were examined at both sites, but only three, E. coli, enterococci, and fecal coliforms, will be considered here. The methods for enumerating E. coli and enterococci have been described earlier (13,22). Fecal coliforms were monitored using a standard method (1).

Particulate Study Procedures

In the second year of the bathing beach studies at Lake Erie, the group at Gannon University was asked to conduct a small pilot study to determine if an association exists between particles larger than three microns and the incidence of gastrointestinal symptoms in individuals swimming on the day the measurements were made. The usual epidemiological and microbiological variates were measured during the course of each trial and two new microbiological measurements also were determined. The first of these was the density of particles three microns or larger that were associated with E. coli colonies. The second was the average number of E. coli per particle. Figure 1 is a flow-chart diagram of the procedure used to determine the two characteristics. Each water sample was divided into two parts. One part was treated in the usual manner. The other was filtered through a three micron pore size Nuclepore filter. The bacteria on the particles retained by the filter were desorbed and dispersed by blending in a buffered surfactant solution (24,33). The E. coli in the desorbed bacterial suspension and the filtrate were enumerated on MTEC Medium (13) after refiltering each through a 0.45 micron filter (Gelman, GN6). The number of particles associated with E. coli colonies was determined by subtracting the density of E. coli found in the filtrate from the density obtained using the customary technique. It was assumed that the E. coli in the filtrate were non-particle associated cells. The number of E. coli per particle was obtained by dividing the total number of E. coli desorbed from the particles by the total number of particles associated with E. coli colonies.

RESULTS

The number of participants in the Oklahoma and Pennsylvania recreational water quality studies and their "highly credible" G.I. symptom rates are shown in Table 5. The mean indicator densities per 100 ml for enterococci, E. coli and fecal coliforms for each swimming season are included in the table. The average swimming-associated illness rate in freshwater swimmers for all of the trials was 6.2 per 1000 individuals. In contrast, the average "highly credible" swimming-associated illness rate in marine water swimmers was 14.8 per 1000 individuals (data obtained from reference 8). The difference between these two swimming rates was shown to be statistically significant ($p<0.05$) using the Wilcoxon Rank Sum Test (36). Correlation coefficients and lines of best fit were calculated using freshwater indicator densities as the independent variable and swimming-associated illness as the dependent variable and these were compared to similar statistics from the marine studies (8). The relationship of enterococci density to "highly credible" G.I. illness for fresh and marine waters is shown in Figure 2. The slope of the line of best fit

for the marine bathing beach data is about twice that observed with the freshwater data (11.6 versus 6.1). The correlation coefficients (r), on the other hand, are similar in magnitude (0.71 versus 0.65). The regression lines describing the relationship of "highly credible" G.I. symptoms to E. coli densities are shown in Figure 3. The slope of the line for the marine data is again greater than that observed for the freshwater data; however, the difference is much less than that for enterococci (7.3 versus 4.7). The correlation coefficient for the freshwater points is approximately equal to that obtained for the marine water points (0.514 versus 0.513). The regression lines for highly credible G.I. illness on fecal coliform densities present an interesting contrast to the relationships observed with enterococci and E. coli. Figure 4 shows that the slopes of the lines calculated from marine (7) and freshwater data are very flat (3.2 and 2.0, respectively). Since the correlation coefficients are in part a function of the magnitude of the slope, they too have small values (marine = 0.15, fresh = 0.23).

It was shown in the marine recreational water quality studies that of all the bacteriological water quality indicators examined, enterococci had the best correlation to the health effects observed in swimmers (8). E. coli were ranked second and fecal coliforms ranked eighth among eleven indicators studied. A similar ranking was observed in the freshwater studies. The three freshwater health effects/water quality indicator regression lines shown in the previous figures are compared directly in Figure 5. If the three health effect indicator relationships are ordered according to the correlation coefficients of their regression lines, enterococci would clearly rank first, E. coli second, and fecal coliforms would rank third.

The results of the pilot study conducted to determine if particles were related to swimming-associated illness are shown in Table 6. The health effects data are given in terms of total gastrointestinal symptoms rather than "highly credible" G.I. symptoms because the frequency of occurrence of the latter was less than one on many of the trial days. The risk attributed to swimming was calculated as described by Rimm, et al. and these are shown in the first column of Table 7 (3). These values are ranked in increasing order, and the companion water quality indicator and particle data collected on the same trial day are shown in columns 2, 3, and 4. The relatedness of the attributable risk to the E. coli density per 100 ml, to the density of E. coli-associated particles at least three microns in size, and to the density of E. coli per particle was determined using Spearman's rank-difference correlation coefficient (31). A comparison of the correlation coefficients is shown in Table 8. The number of E. coli associated particles per 100 ml had the highest degree of relatedness to the swimming-associated risk. The E. coli per 100 ml also had a positive correlation to swimming-associated risk but are about one-third the magnitude of that found with particle density. The correlation coefficient for attributable risk relative to E. coli density per particle was a relatively large negative value, indicating an inverse relationship between these two variables.

DISCUSSION

The results of the freshwater bathing beach studies are significant in at least three respects, all of which may be important to those interested in wastewater disinfection. The first aspect is that the direct relationship between swimming-associated gastrointestinal illness and water quality observed at marine bathing beaches was also found at freshwater beaches. This finding was not unexpected since Stevenson (37) observed a detectable risk for gastrointestinal illness in freshwater swimmers. However, the new data confirm the fact that as the quality of bathing water deteriorates, the risk of gastrointestinal illness increases. This information will be very useful for establishing water quality criteria. The second notable aspect of the freshwater studies is that, as in the marine studies, the enterococci correlate best with gastrointestinal illness. The superiority of enterococci over *E. coli* and fecal coliforms as an indicator of recreational water quality is most likely a reflection of their ability to survive better in aquatic environments (4) and also because of their greater resistance to the effects of chlorination (25). Enterococci also have been shown to be less sensitive than *E. coli* to the effects of solar radiation (15,35). The attributes of this indicator frequently have been overlooked because of methodological considerations and its lower density in fecal wastes relative to coliforms or fecal coliforms. However, it has been proposed in the past as a water quality indicator for recreational waters (16,21) and perhaps the time has come that its use be given serious consideration.

The most conspicuous aspect of the freshwater bathing beach studies is the low swimming-associated gastrointestinal illness rates relative to those observed in marine water swimmers at equivalent indicator densities. This difference in illness rates is probably a function of dissimilar indicator die-off patterns in the two swimming environments. Mitchell and Chamberlain (26) have pointed out that the time interval for 90 percent die-off of coliforms was approximately 52 hours in freshwater and only about two hours in seawater. This appreciable die-off rate difference between coliforms in marine and freshwater environments may well account for the observed differences in swimming-associated illness rates. It is assumed that the similarity in the symptoms of marine and freshwater swimmers is due to the same or closely related enteric pathogens. The difference in illness rates is probably the most significant finding of the freshwater study, since it will preclude the use of a single criterion for marine and fresh recreational waters.

Although the swimming-associated illness data may prove useful for establishing effluent guidelines, it is the data dealing with microbial laden particles and swimming-associated risk that may hold the greatest interest relative to wastewater disinfection processes. The rationale for the particle experiments was based on the conceptual hypothesis that a particle of fecal material may contain thousands or millions of bacterial or viral pathogens and therefore the swallowing of a single particle by a swimmer would be sufficient to initiate an infection. This hypothesis is supported by data indicating that polioviruses encapsulated in fecal material are more resistant to chlorination than nonencapsulated viruses (20). It also is known that particle-associated bacteria and viruses survive longer than non-particle-associated bacteria and

viruses (5,6,17). Therefore, if it could be shown that a health effect was directly related to either the number of E. coli per particle or the density of particles associated with E. coli, then it would not be difficult to infer that pathogens behave similarly and are responsible for the effect. The results of the small pilot study conducted at the Lake Erie beach indicated that the density of particles containing E. coli appeared to be more closely related to the observed health effects than either the density of E. coli per 100 ml or the number of E. coli per particle. This result implies that a high probability of ingesting a single particle is more important than the average number of viable pathogens per particle. This factor may provide an answer to the question, "Why does significant swimming-associated illness occur in good quality water?" If E. coli are valid surrogates for pathogens in feces, then the ingestion of a single particle containing multiple infectious units could easily account for the observed effects. The findings also suggest that the swimming-associated illness rate could be lowered by some type of intervention at the treatment level. Further studies, to confirm these preliminary data, are being planned by the investigators at Gannon University.

CONCLUSIONS

The Environmental Protection Agency-supported studies at freshwater bathing beaches during 1979 and 1980 have produced a great deal of data. The small part of that data which has been presented here leads to the following conclusions.

1. There is a risk associated with swimming in freshwater and this risk is proportional to the quality of the water.

2. The bacterial water quality indicator that correlates best with swimming-associated gastrointestinal illness is the enterococcus group.

3. The swimming-associated gastrointestinal illness rate in freshwater swimmers is significantly lower than that observed in marine water swimmers at equivalent bacterial indicator densities. This difference rules out the establishment of a single water quality criterion for both fresh and marine bathing beach waters.

4. The appreciable swimming-associated gastrointestinal illness rate that occurs in good quality water may be due to the presence of particles which contain high densities of pathogens.

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Table 1. Swimming-Associated Enteric Disease Outbreaks

Year	Country	Type of Water	Disease or Agent	Water Quality
1909	England (29)*	Sea	Typhoid	Poor
1921	U.S.A. (29)	Sea	Typhoid	Poor
1932	U.S.A. (29)	Sea	Typhoid	Poor
1936	U.S.A. (18)	Fresh	Typhoid	Unknown
1958	Australia (1,14)	Sea	Typhoid	Poor
1973	U.S.A. (19)	Fresh	Coxsackie B	Unknown
1974	France (12)	Fresh	Coxsackie A	Poor
1978	U.S.A. (32)	Fresh	Shigellosis	Poor
1979	U.S.A. (30)	Fresh	Enteritis	Unknown

*Reference number in parenthesis

Table 2. Epidemiological Studies of Swimming-Associated Illness

Type of Study	Year	Etiologic Agent	Swimming Illness	Water Quality
Retrospective (27)*	1959	Poliovirus	No	Variable
Retrospective (11)	1981	Enterovirus	Yes	Good
Prospective (37)	1951	Unknown	Yes	Variable
Prospective (8)	1972	Unknown	Yes	Variable

*Reference number in parenthesis

Table 3. Sequence of Events for Epidemiological-Microbiological Trials

Day of Week	Day	Activity	Function
Saturday	1	Beach interview, sample water	a. Obtain Personal Data b. Reject Pre-Trial Midweek swimmers c. Query on beach activity d. Assay of water samples
Sunday	2	(same as above)	(same as above)
Monday	3	Reminder letter	a. Reminder to note illness
Monday	10	Phone interview	a. Obtain illness information b. Reject post-trial midweek swimmers c. Obtain remainder of demographic information

Table 4. Definition of Total and Highly Credible G.I. Health Effects

Health Effects Variates	Definition
Total G.I. Symptoms	Any one of the following: vomiting, nausea, diarrhea or stomachache
Highly Credible G.I. Symptoms	Any one of the following: 1. vomiting 2. diarrhea with fever or disabling condition* 3. stomachache or nausea accompanied by a fever

*indicates individual remained at home, remained in bed or sought medical advice

Table 5. Highly Credible Gastrointestinal Illness Rates Among Swimmers and Non-Swimmers at Freshwater Bathing Beaches

	Oklahoma		Pennsylvania	
	Beach A	Beach B	Beach A	Beach B
1979				
Swimmers				
Total No.	2491	1864	3248	2139
Illness Rate[1]	25.29	20.92	17.24	14.49
Non-Swimmers[2]				
Total No.	787		1854	
Illness Rate	18.53		9.17	
Enterococci	39[3]	7	11	16
E. coli	138	19	23	47
Fecal Coliform	436	51	-	-
1980				
Swimmers				
Total No.	4503	3085	2383	1995
Illness Rate	15.32	12.96	13.42	22.06
Non-Swimmers				
Total No.	1063		1532	
Illness Rate	9.11		9.30	
Enterococci	23	20	38	85
E. coli	52	71	139	246
Fecal Coliform	230	234	37	104

[1]Per 1000 participants

[2]Non-Swimmers from Beaches A and B combined to form single control group

[3]Density per 100 ml

Table 6. Total Gastrointestinal Symptom Rates in Swimmers and Non-Swimmers by Individual Day

	Swimmers		Non-Swimmers	
Trial Day	Number of Participants	% Ill	Number of Participants	% Ill
1	292	9.2	103	8.7
2	126	5.6	77	3.9
3	244	7.8	76	5.3
4	105	9.5	129	3.9
5	140	7.7	63	3.2
6	269	8.2	88	3.4
7	172	2.9	105	0.9

Table 7. Summary of G.I. Illness, Water Quality and Particle-Related Variates

Risk Attributable to Swimming Exposure	E. coli/ Particle	E. coli/ 100 ml	Particles With E. coli/100 ml
4.2*	1406	141	32
21 7	49	253	124
28.3	419	110	65
41.0	20	567	420
47 9	309	127	88
53.1	157	308	172
56.0	47	200	130

*Percent of illness in swimmers due to swimming exposure

Table 8. Correlation of Swimmer-Associated G.I. Illness With Water Quality Indicator and Particle-Related Illness

Comparison	Correlation Coefficient
G.I. Illness vs. E. coli density per particle	-0.5
G.I. Illness vs. E. coli density per 100 ml	.21
G.I. Illness vs. density of E. coli associated particles per 100 ml	.61

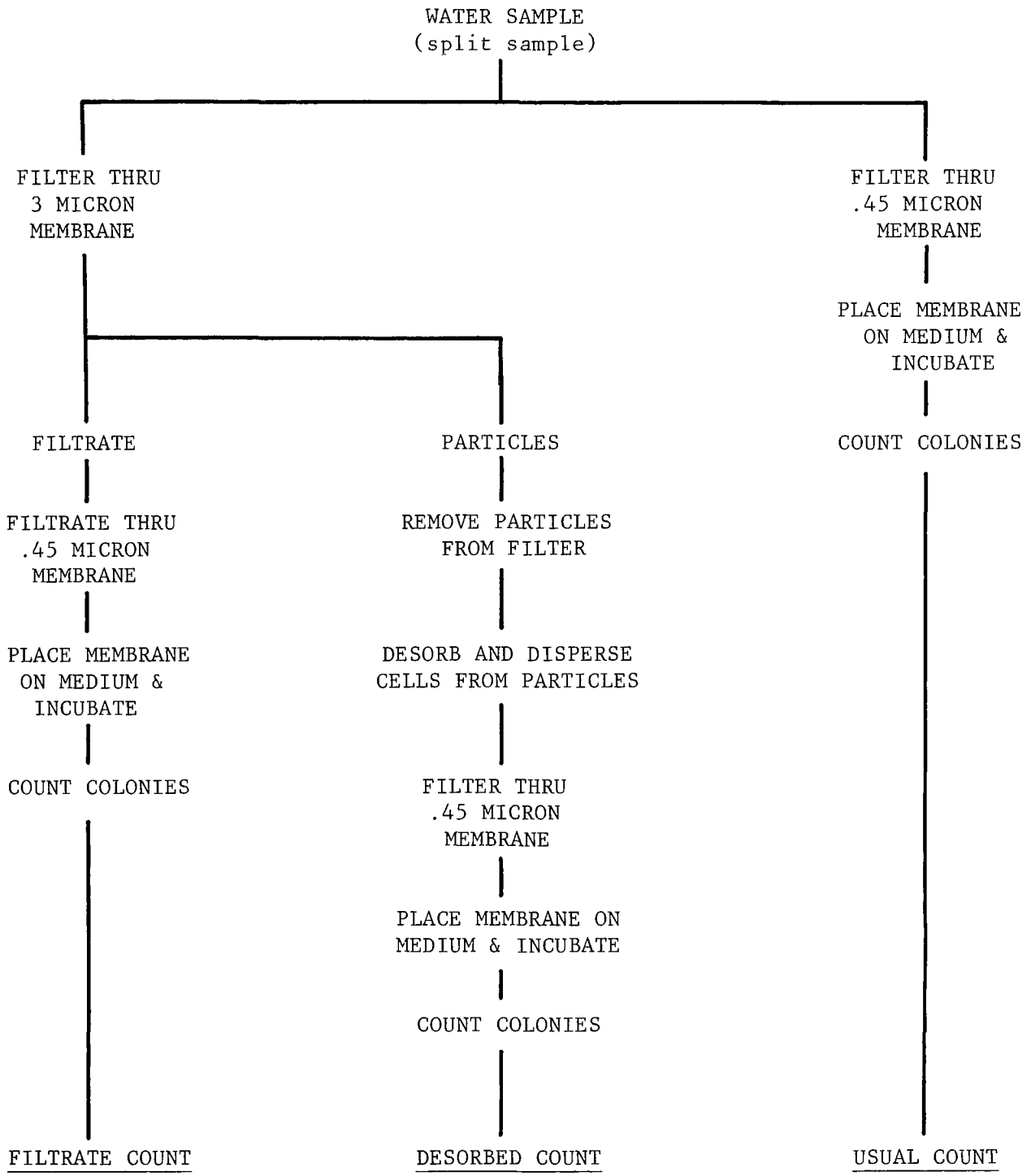

Figure 1. Sample Treatment Protocol

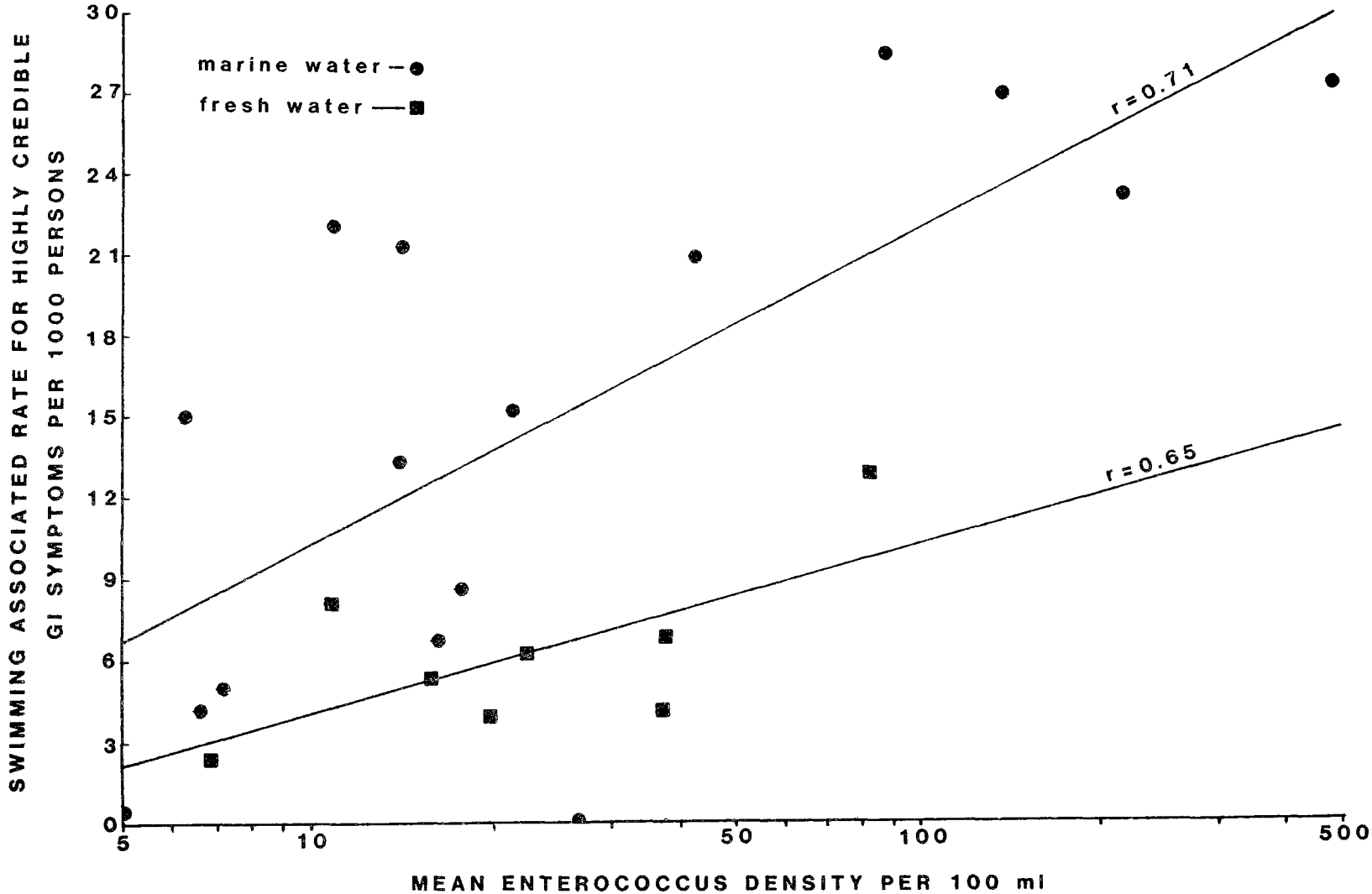

Figure 2. Comparison of Gastrointestinal Symptom Rates at Marine and Freshwater Bathing Breaches Using Mean Enterococcus Density as the Index of Water Quality

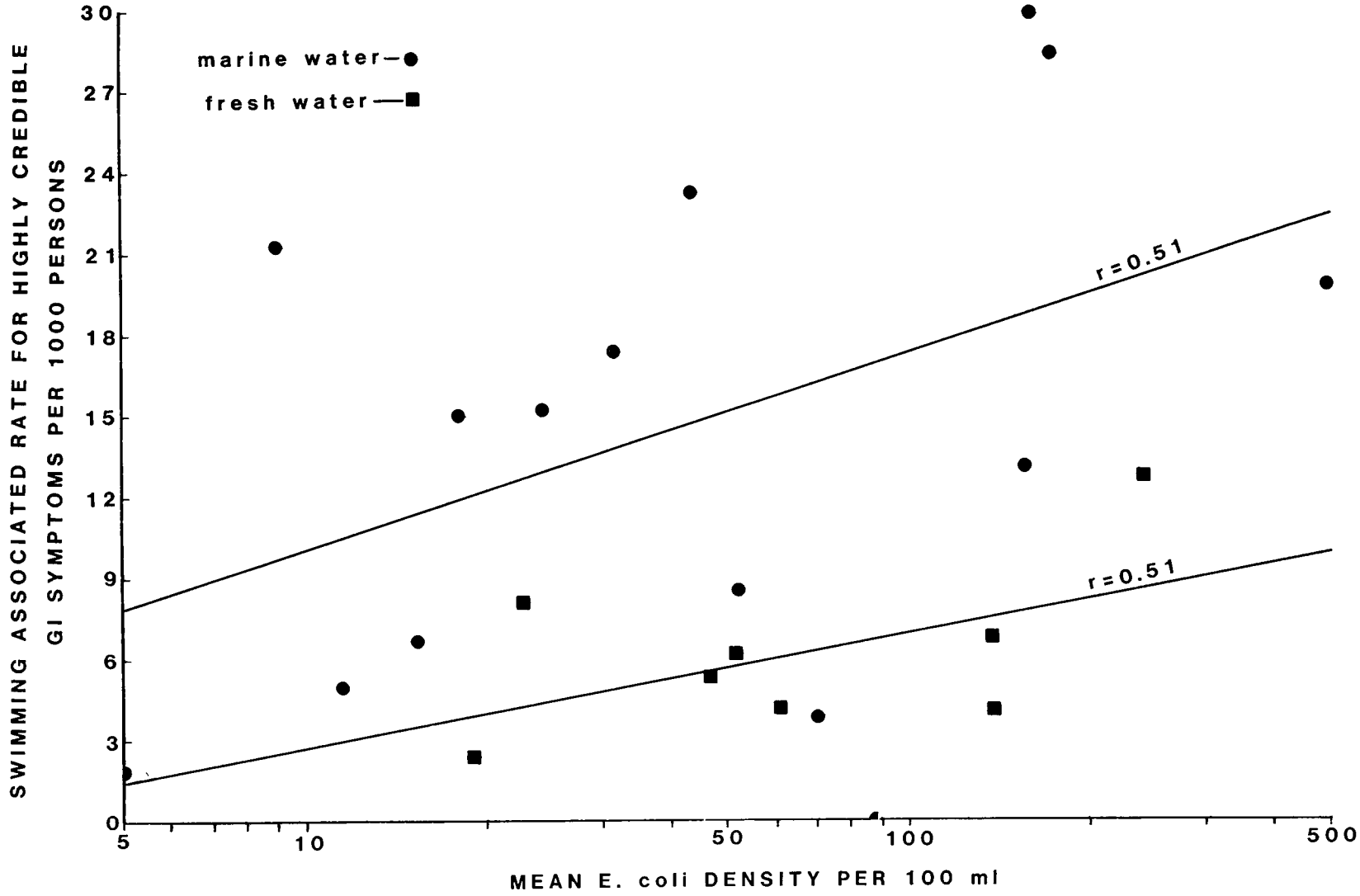

Figure 3. Comparison of Gastrointestinal Symptom Rates at Marine and Freshwater Beaches Using Mean E. coli Density as the Index of Water Quality

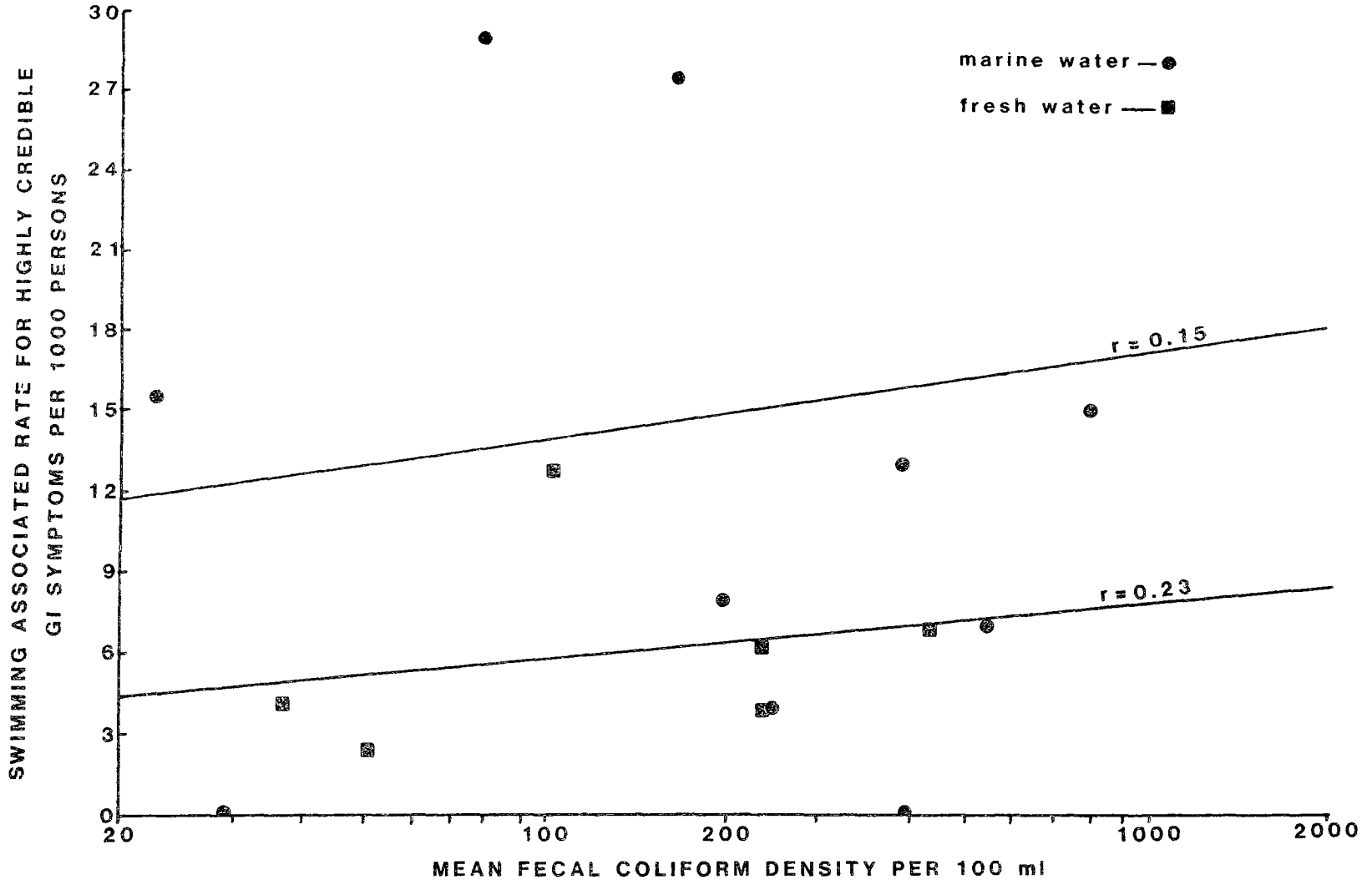

Figure 4. Comparison of Gastrointestinal Symptom Rates at Marine and and Freshwater Bathing Beaches Using Mean Fecal Coliform Density as the Index of Water Quality

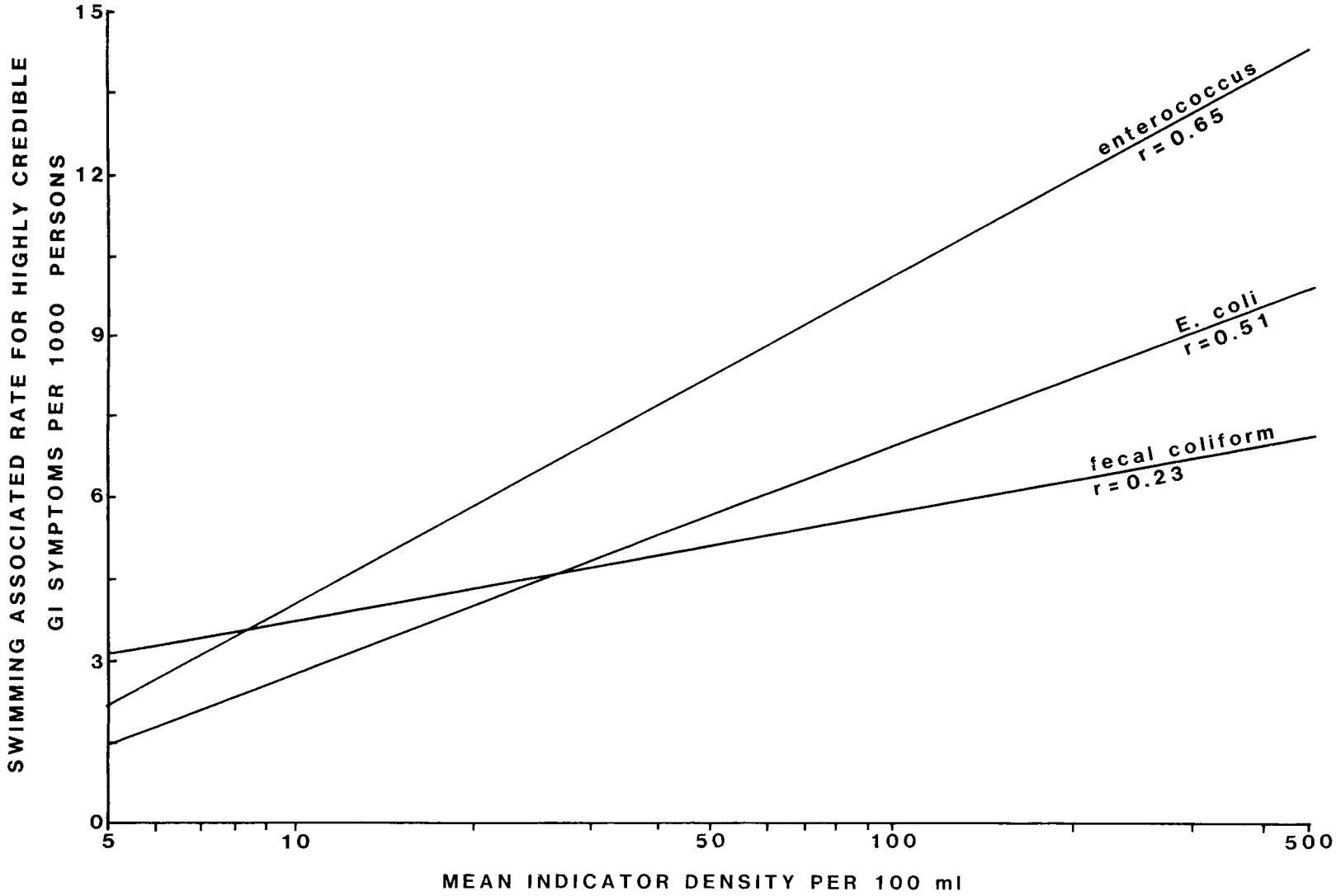

Figure 5. Relationship of Enterococcus, E. coli and Fecal Coliform Mean Densities to Gastrointestinal Symptom Rates at Freshwater Bathing Beaches

1. OPTIMIZATION OF MIXING FOR DISINFECTION

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ABSTRACT

Rapid bulk diffusion of chlorine solution introduced into a wastewater stream has been demonstrated to markedly improve disinfection efficiency and decrease chlorine dose requirement which, in turn, decreases the formation of deleterious chlorinated by-products. Rapid mixing of chlorine with a wastewater stream initially provides increased contact between the bacteria and virus with chlorine before the chlorine is dissipated in other reaction pathways. Controversy exists concerning the identification of the initial active disinfecting chlorine species in a well mixed system and further work is required to identify whether this chlorine species is free chlorine (hypochlorous acid), a chloroorganic, or some other chlorine species. The design of chlorine mixing is generally accomplished using empirical procedures and disinfection models which do not account for system geometry or energy input into the system. Rational optimization of the mixing of chlorine into a wastewater stream for disinfection purposes requires consideration of wastewater quality and flow rate, the flow rate of the chlorine stream and its chlorine concentration, and the geometry of the mixing process. The Prandtl eddy frequency and the mean velocity gradient have properties which make them useful for disinfection process design. Though the Reynolds number quantifies the amount of turbulence in a mixing system, it is not a universal descriptor for the rapid mixing, disinfection process. The Collins' model, though not containing mixing descriptors, was modified on an individual case basis to describe bacterial inactivation for a disinfection system having greatly improved mixing.

INTRODUCTION

Effective mixing of chlorine with a wastewater stream is recognized as an important factor for optimizing a chlorine disinfection process. However, most wastewater treatment plants employing chlorination add the chlorine as an aqueous solution through a diffuser at the head of a chlorine contact basin with little or no effective mixing. Under these transport and reaction conditions free chlorine is not mixed throughout the mass of the incoming wastewater. Since the formation of chloramines and other chlorinated by-

products at wastewater pH values of 6 to 9 is very rapid, essentially complete in a few seconds, normal methods of adding chlorine to a wastewater stream do not optimally mix the chlorine into the wastewater stream, thereby impairing the disinfection process. For the designer and operator to rationally optimize the chlorination process, a model must be available for their use which ideally incorporates parameters describing intensity of mixing relative to the system's physical geometry, chemical reactions, and the resultant inactivation of bacteria and other organisms of relevant health significance. This paper describes work by the author and others to develop the practice of mixing chlorine into a wastewater stream and to rationally describe the disinfection process for coliform bacteria.

Stenquist and Kaufman (17) mixed an aqueous chlorine stream at bench-scale into a wastewater stream by means of multiple source grid placed in a pipe. The purpose of the grid was to achieve rapid mixing of the chlorine solution with the wastewater stream. As a control for laboratory studies, chlorine was introduced through a single inlet in the direction of flow so that the primary source of turbulence generation for the control was wall friction. Other conditions were similar. After 0.32 minutes of contact time, coliform inactivation was 50 - 55 percent for the control (single inlet) and 97.4 percent for the grid mixer. However, for a given chlorine dose, detention time varied inversely with chlorine residual yielding similar amounts of coliform inactivation for both types of reactors. Subsequent field studies conducted on a plant of approximately 1.7 mgd (6,440 cu m/day) demonstrated no improvement of a grid chlorine diffuser relative to a diffuser placed in-line. White (18, 19) reported on a survey of the chlorination facilities of several wastewater treatment plants discharging into San Francisco Bay. Plants introducing chlorine at a point of turbulence demonstrated consistently higher coliform removals. Kruse et al. (12) studied the chlorine disinfection of the secondary effluent (trickling filters) from a 1.5 mgd (5,680 cu m/day) wastewater treatment plant. Under normal plant conditions an aqueous chlorine solution was introduced through a diffuser at the head of the contact basin. Improved mixing was achieved by introducing the aqueous chlorine feed stream at a point of turbulence in the wastewater line upstream from the contact basin. Coliform inactivation after 2 and 10 minutes contact time showed no significant increased coliform inactivation attributable to improved mixing.

Sepp and Bao (15), in a study of seven California wastewater treatment plants, passed unchlorinated wastewater effluent through an optimized pilot disinfection system employing turbulent mixing. In each instance they compared the results of bacterial inactivation achieved with the plant full-scale disinfection system to the bacterial inactivation achieved with the optimized pilot plant. Sepp and Bao (15) found that better bacterial inactivation resulted from better mixing.

Collins and Selleck (3) and Collins et al. (4) described the below model specific for coliform inactivation in a wastewater stream.

$$\frac{N}{N_o} = (1 + 0.23\ ct)^{-3} \qquad (1)$$

where N_o = coliform bacteria density at time zero

N = coliform bacteria density at time t

t = mean detention time (reactor volume divided by the flow rate), minutes

c = combined amperometric chlorine residual, mg/l.

Collins and Selleck (3) and Collins et al. (4) also reported that backmixing appreciably decreased the germicidal potential of the chlorine residual. They found the effect of initial turbulent mixing on the bactericidal effectiveness of chlorine introduced into a wastewater stream to be highly significant.

Haas (8) enumerated the overall sequence of events during chemical disinfection as follows:

"1) The disinfectant, either as a solution, or as a gas must be brought into intimate contact with the wastewater, and mass transfer into the bulk solution must be allowed to occur.

"2) The disinfectant, entering the bulk liquid, must be transported to the exterior of the microorganism which is to be inactivated.

"3) The active species, located at the microbial exterior, must be transported or bound to or at the lethal site.

"4) The microorganism is inactivated at a rate proportional to the concentration of the disinfectant species which is in an 'active' form at the lethal site.

"5) Simultaneously to the above events, liquid phase decomposition of disinfectant may occur via the exertion of demand or the formation of less active species (such as chloramines)."

Longley (13), reporting on disinfection studies carried out at a wastewater treatment plant located at Fort Meade, MD, found that the mean velocity gradient and the Prandtl eddy frequency are descriptors for disinfection employing rapid mixing.

The Reynolds number (R_e) is a dimensionless number relating inertial and viscous forces. While the intensity of turbulence for a particular system is directly related to R_e, severe limitations exist for using R_e as the criterion to classify in different mixing systems the degree of material homogeneity (completeness of mixing) which can be attained as a function of time. A prime limitation is that for a pipe flow system the temporal and mixing relationships are inversely related to the pipe diameter, whereas R_e is directly related to the pipe diameter.

Brodkey (1) observed that the statistical theory of turbulent mixing has been developed parallel to turbulent motion theory. The basic linear equation for turbulent mixing is that of mass (or heat) conservation, which is the counterpart of the nonlinear Navier-Stokes equation for turbulent motion. The problem of turbulent mixing presents all the difficulties that turbulent motion does because of the nonlinearity of the governing physical equations when expressed in terms of averages. Hinze (10) discusses a phenomenological theory describing the distribution of mean values of a quantity, such as momentum or mass, by the effect of turbulence. One of these, Prandtl's theory, has its analogy in the kinetic theory treating the molecular transport processes of gas which describes the mean free path of a gas as the average distance a gas molecule travels before striking another.

Davies (5) reports extensively on the development of the Prandtl mixing length theory and its application to experimental data. Over the core of a pipe and away from its wall it has been shown experimentally that an empirical approximation of the velocity profile for Reynolds numbers to 10^5 is

$$\frac{V_x}{V_x\ (\text{center})} = \left(\frac{y}{a}\right)^{1/7} \qquad (2)$$

where V_x = time-average axial velocity of the flowing fluid at any point away from the wall

V_x (center) = time-average axial velocity of the flowing fluid at the pipe center

y = distance from the pipe wall

a = pipe radius.

The above equation may be developed to define the Prandtl eddy frequency, f, as

$$f = 0.33\,\frac{V_m}{a}, \text{ where } V_m = \text{mean flow velocity.} \qquad (3)$$

The analogy between mass and momentum fluxes is sufficient that the effective mean eddy length may be approximated as being the same for both momentum transfer and mass transfer.

Camp and Stein (2) stated that the concepts concerning the mean velocity gradient, dv/dy, are applicable to all phenomena involving fluid friction loss. The mean velocity gradient may be determined from the expression

$$G = \sqrt{\frac{P}{\forall \mu}}$$

where G = mean velocity gradient (dv/dy)

P = power input

$\forall$ = volume of system through which power is dissipated

μ = dynamic (absolute) viscosity.

Glover (7) has related observations of coliform disinfection by Collins et al. (3) to the product of the velocity gradient and time of contact (GT). He credits the GT product as being a good parameter to describe mixing intensity in a chlorine contact system.

McKee et al. (14) found that data obtained from bacterial inactivation by chlorination, plotted as a function of time, best fit a line described by

$$\frac{N}{N_o} = \left(\frac{t}{t_o}\right)^m \tag{5}$$

where N = number of organisms surviving at time, t

N_o = number of organisms at initial time, t_o

m = exponent characteristic of disinfection system

After fitting the data of several sources, they found that the value of m had a range of -0.8 to -3.8 with an approximate mean of -2. When plotting data obtained from bacterial inactivation as a function of chlorine dosage they found two straight line relationships, one fitting data for chlorine dosages less than about 11 mg/l, and the other fitting data for chlorine dosages greater than about 11 mg/l. This disparity was explained as possibly relating to the most susceptible coliforms which are not protected by solids.

Hom (11) studied the inactivation of coliforms in stabilization pond effluent chlorinated at selected doses between 0.25 and 2.0 mg/l. He found that the reaction kinetic is a complex m and n order reaction which is dependent on chlorine dosage, contact time and the number of surviving organisms. He postulated and developed the general model shown below.

$$\frac{dN}{dt} = -KNt^m C^n \tag{6}$$

where K = first order rate constant

N = number of coliform organisms per unit volume

t = time

C = concentration of applied chlorine dosage

m = reaction rate constant

n = coefficient of dilution

Setting $m = n \neq 0$ results in the below equation which is a form of Chick's Law.

$$\frac{dN}{dt} = -KN \tag{6.1}$$

Setting $m = 0$ and $n = 0$, an n-order model results.

$$\frac{dN}{dt} = -KNC^n \tag{6.2}$$

An inspection of equation (6.2) shows that it is a variant of the C^nt relationship used by many investigators where the product of this relationship equals a constant for a given percentage of organism inactivation.

Setting C^nt = constant = k', then

$$C^n = k'/t \tag{6.3}$$

Substituting, setting limits, integrating, converting to $\log_{10}$, and developing the above model yields,

$$\log \left(\frac{N}{N_o}\right) = -K_n \log \left(\frac{t}{t_o}\right) \tag{6.4}$$

K_n is determined by plotting log (N/N_o) as a function of log t.

Letting $m = 0$ and $n = 0$, an m-order and n-order reaction results.

Substituting into equation (6) the expression $C^n = k'/t$,

$$\frac{dN}{dt} = \frac{-K\ Nt^m k'}{t} \tag{6.5}$$

Further development of Hom's model yields,

$$\log \left(\frac{N}{N_o}\right) = \frac{-kk't^m}{m} \tag{6.6}$$

The rate constant, k, is for $\log_{10}$, and m is the reaction kinetic constant for the m-order and n-order reaction where $m \neq 0$ and $n \neq 0$. The constant "m" is determined by plotting log $[\log(N/N_o \times 10^{10})]$ as a function of log (t).

Eliassen et al. (6) and Hess et al. (9) have proposed a model relating coliform densities in sewage as expressed by most probable number, and the

chlorine residual as determined by the orthotolidine test. The model is

$$\frac{1}{\log (MPN)} \qquad a + bR \qquad (7)$$

where MPN = most probable number of coliform organisms in 100 ml

a = constant

b = constant

R = the orthotolidine chlorine residual

Data for evolution of the model were obtained from over 100 sewage treatment plants and represented 5,000 sets of data. A high correlation for the model was achieved using the available data.

MATERIALS AND METHODS

Studies were carried out at the Fort Meade Sewage Treatment Plant No. 2. The plant is a conventional trickling filter plant. The chlorine stream was produced by passing tap water and chlorine gas through an ejector. The flow rate varied from an approximate minimum of 0.9 mgd (3,410 cu m/day), which was attained during the late morning or early afternoon hours. Wastewater streams investigated during the study were primarily those occurring between the hours of 0900 to 1800 during week days. During these hours BOD_5 of the secondary effluent was 20-25 mg/l, and the organic nitrogen and ammonia concentrations were 4-6 mg/l and 11-15 mg/l, respectively. Optimization of mixing in the pilot plant was accomplished using the pipe and Venturi mixers which were mounted and operated in a trailer located near the chlorine contact chamber. The trailer was equipped for the conduct of all chlorine and pH determinations and all coliform assay procedures.

Indigenous coliforms having a median density of 350,000 per 100 ml prior to disinfection were used as indicator organisms for the bacterial inactivation studies. The multiple tube fermentation technique given in Standard Methods (16) was used for determination of total coliform densities. Results were confirmed using brilliant green bile lactose broth.

For the plant condition studies, composited samples in replicate were taken in sterile bottles containing sodium thiosulfate. For mixer studies samples of the mixed stream for bacterial analyses were withdrawn immediately downstream from the mixer by means of a Cornwall syringe equipped with a three-way valve. At least two 5-milliliter portions were withdrawn for each sample and injected directly into a vial containing sodium thiosulfate. The chlorine contact time from chlorine introduction into the mixer until injection of the sample into the vial was about 2 to 4 seconds. The syringe was flushed several times with the sewage-chlorine mixture between samplings. Samples for contact periods of about 15 seconds or greater were collected at

the discharge point into the contact basin and were held for the required time period before neutralization of the disinfectant with sodium thiosulfate. Total chlorine and free chlorine residuals were determined, the latter qualitatively, using modifications of the leucocrystal violet procedure of Black and Whittle (16).

The sewage stream was pumped from the secondary effluent stream into the trailer and through a rotameter prior to introduction into the mixer. The chlorine stream likewise passed through a rotameter prior to introduction into the mixer as the disinfectant stream. The rotameters were calibrated by a positive displacement technique.

DISCUSSION

McKee's et al. (14) proposed mathematical model, previously discussed, is as follows,

$$\frac{N}{N_o} = \left(\frac{t}{t_o}\right)^m \tag{5}$$

The m values have been calculated for different plant and mixing conditions and are tabulated in Table 1. The value of m, a function of the amount of inactivation within a designated time period, is therefore also a function of other variables. A listing of the more important parameters includes the chlorine stream pH, the mixed stream pH, the chlorine species, the chlorine dose, and mixing. It is observed that the calculated m values differ over a wide range, presumably due to differing disinfection conditions. Thus, this is a poor model to be used for all but very well defined conditions.

For the case where m = 0, development of Hom's model was shown to yield,

$$\log\left(\frac{N}{N_o}\right) = -K_n \log\left(\frac{t}{t_o}\right) \tag{6.4}$$

K_n is determined by plotting log (N/N_o) as a function of log (t). An evaluation of K_n for representative mixing data was performed as is shown in Table 1. Where multiple observations were made as a function of time, the calculated values of K_n were quite dissimilar between each consecutive time interval. Thus, it is evident that Hom's model, when m = 0, for the mixing data is not linear as a function of log (t). The values of K_n vary with a number of system parameters not included in the model, the most important being mixing intensity and the pH of the chlorine stream. Therefore, as a general model this model is inoperative.

Similar conclusions may be drawn for the model where $m \neq 0$, and $n \neq 0$ which was shown to be expressed as

$$\log\left(\frac{N}{N_o}\right) = \frac{-kk't^m}{m} \tag{6.6}$$

TABLE 1

EVALUATION OF McKEE'S AND HOM'S MODELS FOR COLIFORM DISINFECTION

	Test Condition	Time, t (min)	Log (t)	Inactivation N/N_0	Log (N/N_0)	t/t_0	McKee's m	Hom's K_n m=0, n≠0	Hom's m m≠0, n≠0
I.	Plant Conditions[a, b]	2	0.30	1.6×10^{-2}	-1.80				
	17.4 mg/l dose	11	1.11	9.8×10^{-6}	-5.01	5.50	-4.32	-3.97	-0.55
		16	1.26	1.2×10^{-5}	-4.92	8.00	-3.46	0.60	0.01
II.	Plant Conditions[a, b]	2	0.30	1.4×10^{-1}	-0.85				
	4.5 mg/l dose	13	1.04	2.2×10^{-2}	-1.66	6.50	-0.98	-1.10	-0.39
		18	1.20	3.2×10^{-3}	-2.50	9.00	-1.72	-5.25	-1.11
III.	Venturi Mixer[a, b]	.03	-1.52	7.9×10^{-4}	-3.10				
	17 mg/l dose	.37	-0.43	2.5×10^{-4}	-3.60	12.3	-0.46	-0.46	-0.06
		.60	-0.22	7.2×10^{-5}	-4.14	20.0	-0.80	-0.26	-0.29
		15.6	1.19	$\leq 7.2 \times 10^{-6}$	≤ 5.14	520.	-0.75	-0.71	-0.07
IV.	Venturi Mixer[a, b]	.03	-1.52	2.3×10^{-1}	-0.64				
	4.3 mg/l dose	.37	-0.43	1.7×10^{-2}	-1.77	12.3	-1.04	-1.04	-0.05
		.60	-0.22	1.2×10^{-2}	-1.92	20.0	-0.99	-0.72	-0.17
		15.6	1.19	1.4×10^{-2}	-1.85	520.	-4.48	-0.05	-0.11
V.	Pipe Mixer (1 in. diam)[a, c]	1.0	0.00	6.5×10^{-1}	-0.19				
	17 mg/l dose	15.0	1.18	1.0×10^{0}	0.00	15.0	0.16	0.16	0.61
VI.	Pipe Mixer (1 in. diam)[a, b]	1.0	0.00	1.7×10^{-1}	-0.77				
	4.3 mg/l dose	15.0	1.18	3.8×10^{-3}	-2.42	15.0	-1.41	-0.62	-0.91

a. Coliform results confirmed.
b. Chlorine stream pH and mixed stream pH were both 7.0.
c. Chlorine stream pH and mixed stream pH were 2.1 and 6.8 ± 0.2, respectively.

Equation (6.6) may be verified by a linear relationship when a plot is made for log log $(N/N_o \times 10^{10})$ as a function of log (t). The rate constant, m, is determined from the slope of the relationship. The rate constant, m, is not linear as a function of log (t) as shown in Table 1. Intuitively this model has some validity for it expresses the coliform inactivation as a function of both contact time and chlorine residual. However, once again the model does not include the important system parameters of mixing intensity and chlorine stream pH, and therefore it is not ideal as a general model.

The mathematical model originally proposed by Eliassen et al. (6) was shown to be,

$$\frac{1}{\log (MPN)} = a + bR \qquad (7)$$

Table 2 contains an analysis of data together with constants reported by Eliassen et al. Eliassen's model gives a reasonable estimate of the expected coliform inactivation if the constants a and b are evaluated for the system under field operating conditions. As a change in the sewage or chlorine characteristics will effect a change in the attainable coliform inactivation, such a change will also change the constants. It should also be noted that exceptional mixing increases the attainable coliform inactivation and thus affects the constants and consequently the resulting curve. Eliassen's model through careful use and evaluation of constants offered a good approximation of the Ft. Meade coliform data.

The mathematical model proposed by Collins et al. (3, 4) was shown to be,

$$\frac{N}{N_o} = (1 + 0.23\ ct)^{-3} \qquad (1)$$

The model was applied to data for Ft. Meade plant conditions and the Venturi mixer, and the results are presented in Figure 1. The Ft. Meade No. 2

TABLE 2 DATA ANALYSIS FOR MATHEMATICAL MODEL OF ELIASSEN AND COWORKERS

Contact Time, min	Eliassen's Data Constants a	Eliassen's Data Constants b	Ft. Meade Data Analysis Constants a	Ft. Meade Data Analysis Constants b	No. of Observations	Correlation Coefficient
2			.20	.014	12	.818
5	.17	.32				
10	.21	.74				
11			.27	.14	10	.722
15	.28	1.04				
16			.38	.22	11	.826
16 (Venturi Mixer)			.28	.72	10	.893
20	.65	1.02				

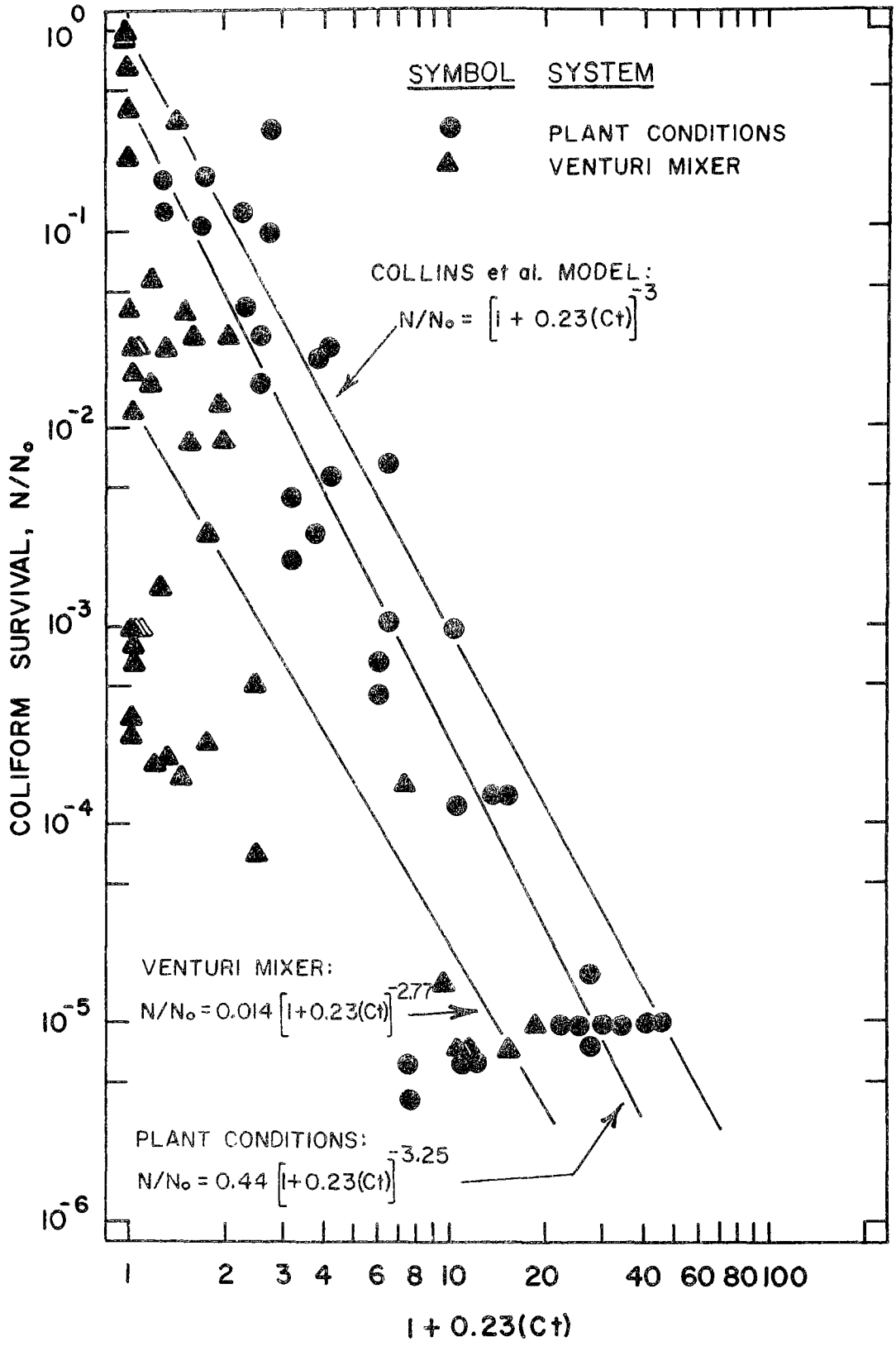

FIGURE 1. Plot of mathematical model for confirmed coliform disinfection according to Collins and Selleck (3) and Collins et al. (4).

conventional disinfection facilities generally attained a slightly greater degree of coliform removal than that shown by the Collins' model as represented in Figure 1.

For conventional disinfection facilities having hydraulic characteristics approaching plug flow and treating domestic sewage, the Collins' model may present a good approximation of the amount of expected coliform removal. The model has the additional advantage that it contains a term representing contact time. However, the model is very conservative when mixing is optimized as is shown by the results for Venturi mixer experiments. The Venturi mixer, with improved mixing of the chlorine stream, achieved significantly greater initial coliform inactivation of approximately 1.5 logs. The subsequent disinfection rate achieved using the Venturi mixer was slightly less than the disinfection rate achieved using conventional plant conditions. The experimental models are shown below.

$$\text{Venturi model:} \quad \frac{N}{N_o} = 0.014 \left[1 + .23\ (ct)\right]^{-2.77} \tag{8}$$

$$\text{Plant model:} \quad \frac{N}{N_o} = 0.44 \left[1 + .23\ (ct)\right]^{-3.25} \tag{9}$$

Regression coefficients for the Venturi model and the plant model were -0.694 and -0.886, both significant at the 99 percent level.

In order to describe rapid mixing quantitatively for both design and operational considerations, a good descriptor of the mixing process must be identified and evaluated with inactivation data. Accordingly, bacterial inactivation data were evaluated as a function of mean velocity gradient, Prandtl eddy frequency, and Reynolds number. Generally, log-log transform of the data yielded the best fit. Data fit, evidenced by the statistics in Table 3, are best for the higher chlorine dose, 17 mg/l, and for the data evaluated as a function of mean velocity gradient and Prandtl eddy frequency. The non-significant correlation coefficients and unremarkable t statistics for 4.3 mg/l chlorine dose data expressed as a function of Reynolds number may be due, in part, to the fact that Reynolds number is a direct function of the mixer diameter, whereas, the time required for chlorine transport across a transverse section of the mixer is inversely related to the mixer diameter. Both mean velocity gradient and Prandtl eddy frequency show promise of being adequate rapid mixing descriptors to be used in conjunction with the design and evaluation of disinfection facilities though considerable additional disinfection data must be evaluated to establish firmly any relationship which may exist. The mean energy gradient is an easily calculable quantity which incorporates the design parameters of power, flow rate, and head loss, the knowledge of which are essential to the designer. However, through the development of the Prandtl eddy frequency theory and related concepts, relationships may be developed which will incorporate material transport factors, rather than momentum transfer, and the decay of the free chlorine species as a function of sewage characteristics. This type of relationship is necessary to describe adequately a rapid mix, disinfection system.

TABLE 3

REGRESSION ANALYSIS*** OF COLIFORM AND f_2 VIRUS INACTIVATION FOR MIXER STUDIES AS A FUNCTION OF MIXING DESCRIPTORS

Independent Variable, X	Dependent Variable, Y	Chlorine Dose, mg/l	Number of Observations	Intercept, a_o	Regression Coefficient of Y on X, a_1	Correlation Coefficient
Mean Velocity Gradient	Coliform	4.3	19	0.57	-0.31*	0.32
	Inactivation	17	17	1.92	-1.10**	0.85
Prandtl Eddy Frequency	Coliform	4.3	19	0.71	-0.37*	0.34
	Inactivation	17	17	2.26	-1.27**	0.82
Reynolds Number	Coliform	4.3	19	1.27	-0.37	0.11
	Inactivation	17	17	6.25	-1.68**	0.55

* Significant at 95% level
** Significant at 99% level
*** $\log Y = a_o + a_1 \log X$

Reynolds number appears to have limited value as a descriptor for mixing conditions necessary to achieve a required degree of bacterial inactivation.

The high correlation coefficient of 0.98 and t statistics significant at the 99 percent level were achieved when Prandtl eddy frequency was regressed as a function of mean velocity gradient as shown in Table 4. These statistics require further evaluation. With the assumption of a direct relationship between these two turbulence descriptors based on the statistics the following expression can be derived where the subscripts G and f denote those terms attributable to mean velocity gradient and Prandtl eddy frequency, respectively

$$\left[\frac{\theta \gamma V_m^3}{\mu}\right]_G \propto \left[\frac{V_m^2}{a^2}\right]_f \qquad (10)$$

where γ = density of water

θ = Moody friction factor

and the other terms have been previously described.

The density and viscosity of water can be closely approximated with a constant over the range of water temperatures encountered during the study. Therefore, the above relationship may be further simplified as

$$\left[\theta \quad V_m\right]_G \propto \left[\frac{1}{a^2}\right]_f \qquad (11)$$

Application of the continuity equation shows that for a given plug flow mixing system and constant flow rate, V_m will vary inversely with a^2. Under the same condition θ will decrease slowly with increasing V_m. This relationship is, therefore, expected but significant since the use of phenomenological relationships of mean velocity gradient and Prandtl eddy frequency allow a close correlation to be developed between disinfection efficiency, material transport parameters, and energy input to the system.

Reynolds number as a function of mean velocity gradient and Prandtl eddy frequency, respectively, is shown in Table 4. The bivariate, linear regression analysis of the log-log transformed data yielded non-significant correlation coefficients of 0.53 and 0.39 for the regression of Reynolds number on mean velocity gradient and Prandtl eddy frequency, respectively. The only practical use to which the Reynolds number may be applied for the evaluation of disinfection data, as discussed herein, is the determination of the friction factor necessary for the derivation of the mean velocity gradient for a given mixing system.

TABLE 4

REGRESSION ANALYSIS*** OF MIXING DESCRIPTORS

Independent Variable, X	Dependent Variable, Y	Number of Observations	Intercept, a_0	Regression Coefficient of Y on X, a_1	Correlation Coefficient
Mean Velocity Gradient	Prandtl Eddy Frequency	11	0.29	0.86**	0.98
Mean Velocity Gradient	Reynolds Number	11	3.46	0.36	0.53
Prandtl Eddy Frequency	Reynolds Number	11	3.53	0.35*	0.39

* Significant at 95% level
** Significant at 99% level
*** $\log Y = a_0 + a_1 \log X$

CONCLUSIONS

Analysis of data collected in studies for improving disinfection of sewage effluent from the Fort Meade Sewage Treatment Plant No. 2 justifies the following conclusions:

1. Rapid and substantial bacterial inactivation may be achieved by chlorination of wastewater under highly turbulent, plug flow conditions.

2. Rapid mixing of chlorine with wastewater may achieve a required degree of disinfection by using less chlorine. Added benefits would be material (chlorine) savings and possible decreased formation of chloro-organics.

3. Mean velocity gradient and Prandtl eddy frequency are highly correlated parameters for coliform inactivation, and they require further investigation and development as descriptors for the rapid mixing, disinfection process.

4. Reynolds number is not a universal descriptor for the rapid mixing, disinfection process.

5. The Collins' model adequately predicted coliform disinfection using conventional disinfection practices. Modification of the coefficients used in the Collins' model permitted use of the model to accurately predict disinfection for an improved mixing system.

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2. UPGRADING EXISTING CHLORINE CONTACT CHAMBERS

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ABSTRACT

Most wastewater treatment facilities in the United States use chlorine for disinfection. It has been estimated that over 200,000 tons of chlorine are discharged through municipal wastewater treatment plant effluents each year.

Recent studies strongly suggest that chlorine disinfection represents a potential environmental health threat because of unwanted chlorinated organic synthesis. Clearly, an alternate means of wastewater disinfection should be made available. It must be realized, however, that significant time will be needed to adequately develop, design and then install alternate disinfection systems in municipal wastewater facilities throughout the United States. During that time period, methods of improving existing chlorine disinfection system efficiencies should be considered. An upgraded system will require less chlorine and will therefore lessen the total burden of chlorine pollution.

An inexpensive method of increasing the contact period in a serpentine flow chlorine contact chamber was developed through hydraulic model studies. This modification scheme (a series of perforated baffles), when installed into existing full scale units with a length of flow to width ratio of 8/1, was found to parallel the performance of a unit with a length of flow to width ratio of 25/1. Disinfection efficiency analysis of the modified and unmodified unit operating at equal conditions at a facility in Maynard, Massachusetts demonstrated that approximately eight percent less chlorine is needed for the modified chamber.

Because savings from this chlorine dose reduction compares favorably to the material cost for these perforated baffles, it may be concluded that the modification scheme is cost effective. In addition to obvious financial benefits, this proposed modification scheme represents the potential for significant pollution reduction and should be considered a practicable interim solution to dangers resulting from present wastewater disinfection practices.

INTRODUCTION

Wastewater disinfection with chlorine is a well established practice that traditionally enjoys favor with design engineers because it is proven in terms of hardware technology and operations manageability. Existing wastewater treatment plants designed and installed over the past couple of decades are typically equipped with a chlorine feed system and a chlorine contact chamber for final effluent disinfection. In effect, most municipalities are committed to the chlorine disinfection process. Unfortunately, many studies (11) indicate that residual chlorine and chlorinated organic compounds released in wastewater effluents represent a potential threat to water quality. It seems inevitable, therefore, that alternative disinfection methods such as ozone or ultraviolet light will replace chorine disinfection systems at wastewater treatment facilities. This replacement process will not only require financial commitments for research and development projects, engineering design activities and installation, but will also require time. Although progress in these areas is being made, the total time required to accomplish widespread replacement of alternate disinfection processes is significant.

At present, more than 200,000 tons of chlorine are discharged each year through municipal wastewater effluents in the United States (3). Numerous studies have noted that efficiency of a chlorine disinfection system largely depends on the chlorine contact chamber's hydraulic character (2,7,9,10). A chlorine contact chamber (referred to as CCC in this paper) with short circuiting currents will not provide the necessary time for disinfection reactions to approach completion. Consequently, a higher chlorine dose is used to obtain the necessary degree of disinfection (7). If a method of improving the efficiency of existing CCC units is available, hazard from wastewater chlorination could be alleviated until more permanent solutions are implemented. Because improvements to existing CCC units are an interim solution, they must be relatively cheap, easy to install and versatile.

Model studies conducted by the author (4) developed a method for improving the hydraulic character of the commonly used cross baffled serpentine flow CCC unit by installing a combination of perforated baffles. These modifications were specifically designed to meet the above mentioned requirements of low cost, simplicity and versatility. This paper reports on observations made at two wastewater treatment facilities in Massachusetts after installing these baffles.

MATERIALS AND METHODS

Figures 1 and 2 illustrate the baffle schemes installed at the Marlboro, MA Easterly Wastewater Treatment Plant and the Maynard, MA Wastewater Treatment Plant respectively. The modification scheme installed at Marlboro, MA is exactly similar to the model modification scheme while the modification scheme installed at Maynard, MA varies from the original model scheme because of differences in tank geometry.

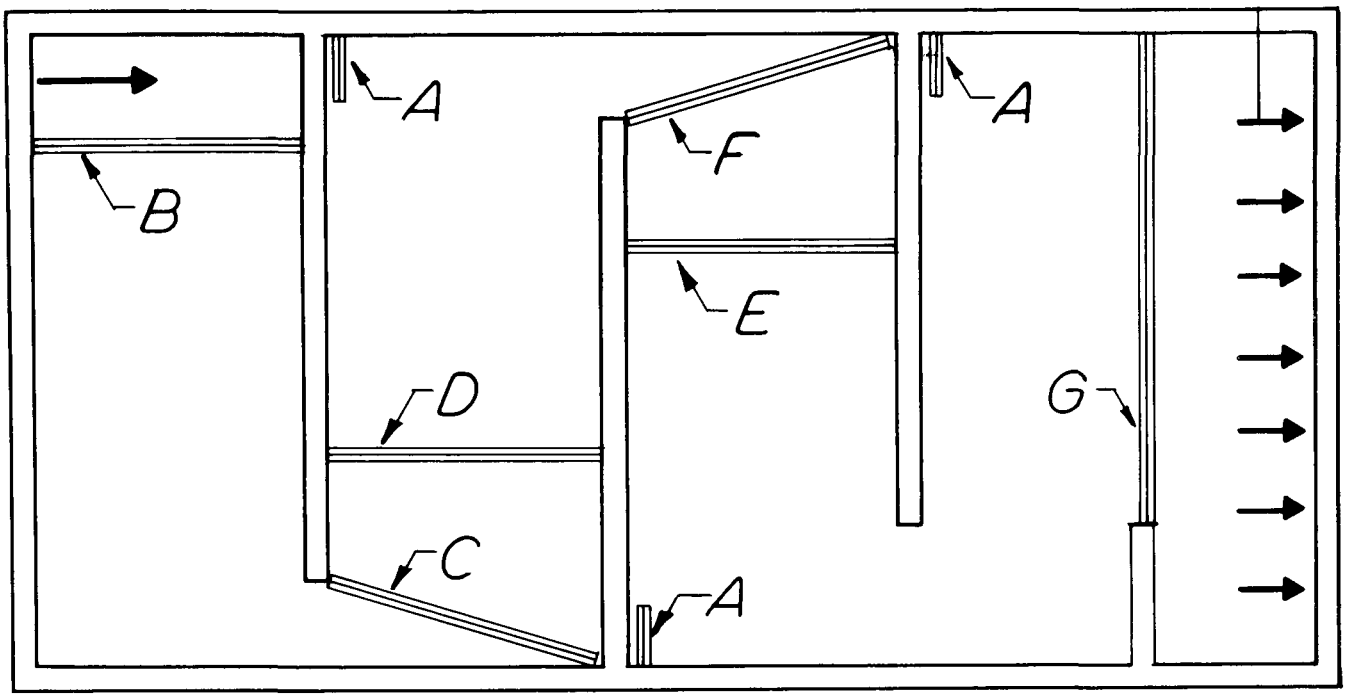


Figure 1. Modified CCC Unit at Marlboro, MA

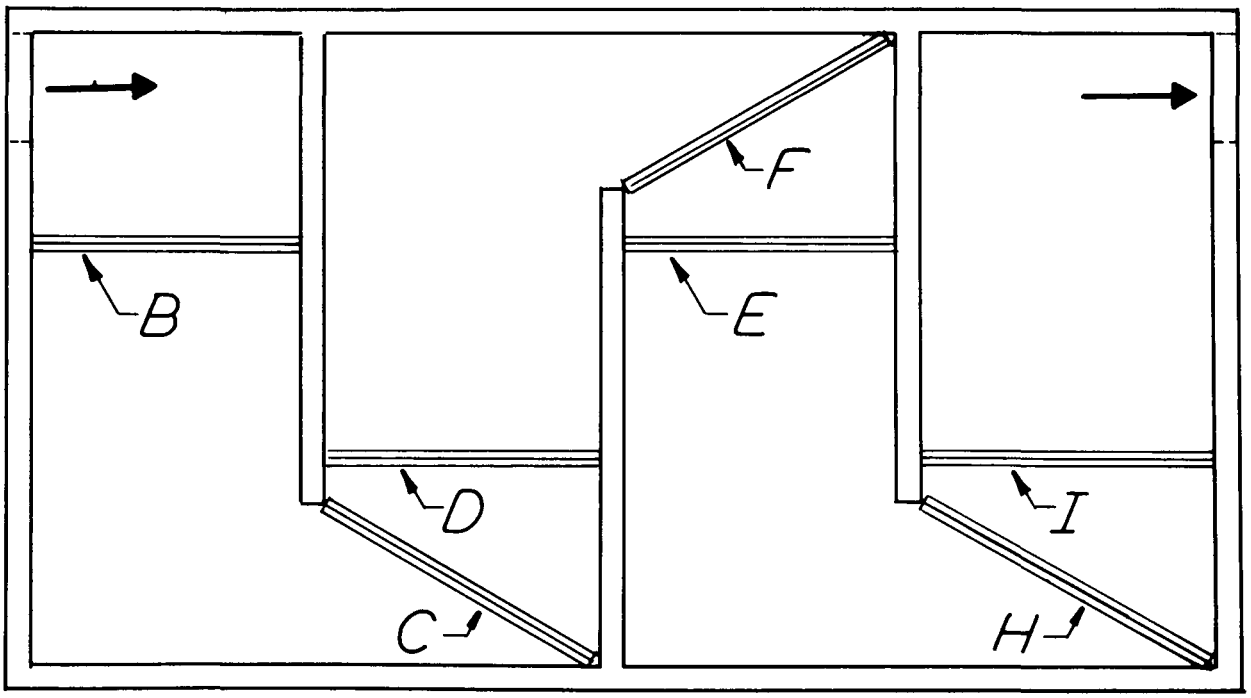


Figure 2. Modified CCC Unit at Maynard, MA

Further description of the baffle configuration and design may be found in other papers (4,5,6). Table 1 lists specifications for both field units.

Because both treatment facilities were equipped with dual CCC units, simultaneous evaluation of modified and unmodified systems were possible as baffles were only placed in one side. This method of field testing helped eliminate the inclusion of many uncontrolled variables (particularly wastewater characteristics) because both modified and unmodified systems were subjected to those variations during the experiment period.

Table 1. CCC Unit and Baffle Specifications

Specification	Marlboro, MA	Maynard, MA
CCC		
Length	15.50 m	6.25 m
Width	7.30 m	3.05 m
Depth	3.00 m	2.44 m
L/W (flow)	8/1	8/1
Baffles B,D,E,I		
Length	3.66 m	1.40 m
Depth	3.05 m	2.44 m
Hole Diameter	15.24 cm	6.35 cm
% Open Space	17% (except B=3.4%)	17% (except B=3.4%)
Baffles C,F,H		
Length	3.76 m	1.70 m
Depth	3.05 m	2.44 m
Hole Diameter	15.24 cm	6.35 cm
% Open Space	17%	17%
Baffle G		
Length	6.10 m	-
Depth	3.05 m	-
Hole Diameter	15.24 m	-
% Open Space	17%	-
Material Cost	$599.00	$250.00

Tracer experiments using Rhodamine WT fluorescent dye were conducted when chlorine was not in use. A single pulse of dye was injected directly upstream before entrance into the CCC unit. Measurements of the effluent dye were made in the field with a Turner Model 111 Fluorometer until all dye was recovered. C-curves (C/C_o vs t/T), and dispersion index values (d) were

generated from the tracer data. Equations used to calculate the dispersion index are as follows:

$$\sigma_t^2 = \frac{\Sigma t^2 c}{\Sigma c} - \left(\frac{\Sigma tc}{\Sigma c}\right)^2 \qquad 1$$

where: t = time
c = tracer concentration at time = t

$$t_g = \frac{\Sigma tc}{\Sigma c} \qquad 2$$

$$\sigma^2 = \sigma_t^2 / t_g^2 \qquad 3$$

$$2d + 8d^2 = \sigma^2 \ . \qquad 4$$

Expression 4 identifies the relationship of the C-curve variance to the dispersion coefficient for an open vessel.

A further development of the dispersion index expression to describe the C-curve for an open vessel is as follows (8):

$$E_\theta = \frac{1}{(4\P\theta d)^{\frac{1}{2}}} \exp - \left(\frac{(1-\theta)^2}{4\theta d}\right) \qquad 5$$

where: $E_\theta = C/C_o$

$\theta = t/T$.

Expression 5 is derived on the assumption that flow is minimally disturbed at the inlet and outlet zones. Use of this expression is explained later in this section.

Total coliform populations were measured by the Membrane Filter test as described in Standard Methods (1). Samples were collected in sterilized bottles containing a 10 percent sodium thiosulfate solution and were immediately placed on ice. Initial MF screening tests were made in the laboratory to predict coliform numbers in order to increase the likelihood of successfully bracketing the required dilutions. This step was found necessary because the coliform population numbers varied considerably. Final incubation was always conducted within 24 hours of sample collection.

Disinfection response data generated from field units and laboratory batch reactors were fitted to an expression introduced by Collins (2) as follows:

$$\frac{N}{N_o} = \left(\frac{b}{ct}\right)^n \qquad 6$$

where: N = coliform population leaving the CCC unit or at time t in a batch reactor

N_0 = coliform population entering the CCC unit, or at initial time in a batch reactor
b = lag coefficient (mg x min/L)
n = velocity coefficient
t = time (detention time of a CCC unit)
c = chlorine dose

Coliform population values entering and leaving the CCC unit, chlorine dose levels and hydraulic retention times were applied to this expression to quantify the disinfection capability of both modified and unmodified systems. After solving these expressions for both systems, a comparison of their relative efficiency difference was calculated by setting both expressions to identical conditions of N/N_0.

A simulation of disinfection response under ideal plug flow conditions was obtained through batch reactor experiments as illustrated in Figure 3. These data were fitted to expression 6 for subsequent prediction of disinfection efficiency in a CCC unit defined through tracer experiment data. Such a method of CCC unit examination was presented by Trussell and Chao (10) with the following expression:

$$N/N_0 = \int_0^\infty (N/N_0)_{batch} \, E_\theta d\theta \qquad 7$$

where: (N/N_0) batch = expression 6 fitted to batch reactor data
E_θ = expression 5

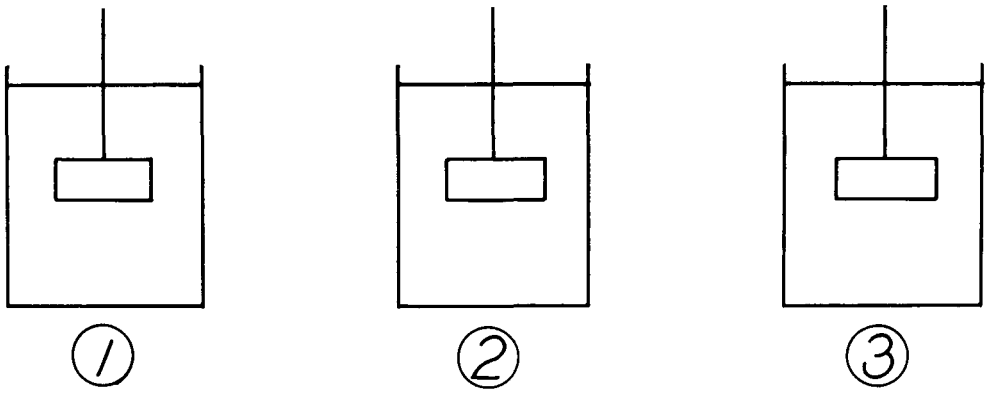
A value of N/N_0 for a given chlorine dose (c) can be calculated with equation 7 if the dispersion index (d), batch reactor data (b,n) and C-curve point (E_θ,d) are known. As with expression 6, expression 7 is used in this study to quantify the relative difference in disinfection capability of a modified and unmodified system.

PRESENTATION AND DISCUSSION

Because wastewater flow received at the Marlboro, MA facility during the field test period (nine months) was unusually low, simultaneous operation of the modified and unmodified CCC units could not be accomplished without allowing hydraulic retention times beyond the typical range. In addition, coliform population concentrations entering the CCC process were low. Consequently, disinfection efficiency data obtained at this facility were not considered reliable and will therefore not be presented here. Information obtained at this facility regarding the cost, durability and handling of the modification baffles, however, was valuable to this study and will be discussed later.

Tracer Response

C-curve plots for the modified and unmodified CCC units are presented in



SAMPLING:			
T = 0	N_o	N_o	N_o
DOSE:	2 MG/L	4 MG/L	10 MG/L
SAMPLING:			
T = 2 MIN.	N 1	N 2	N 3
T = 5 MIN.	N 1	N 2	N 3
T = 10 MIN.	N 1	N 2	N 3
T = 20 MIN.	N 1	N 2	N 3
T = 30 MIN.	N 1	N 2	N 3

Figure 3. Batch Reactor Experiment

Figure 4. As can be seen, an improvement in the chamber's hydraulic character was obtained from the baffle modification scheme. The dispersion index (d) and C-curve variance (σ^2) for these curves are 0.082 and 0.218 for the unmodified unit and 0.035 and 0.076 for the modified unit. A comparison of σ^2 data for various l/w configurations as presented by Marske and Boyle (9) indicates that the unmodified unit responds very closely to the expected performance of a unit with an 8/1 l/w configuration, while the modified unit responds closely to the performance of a unit with a 25/1 l/w configuration. In terms of hydraulic characteristics, therefore, the modified unit responds more efficiently.

Disinfection Response

Log-log plots of N/N_0 vs ct from field experiments at the unmodified and

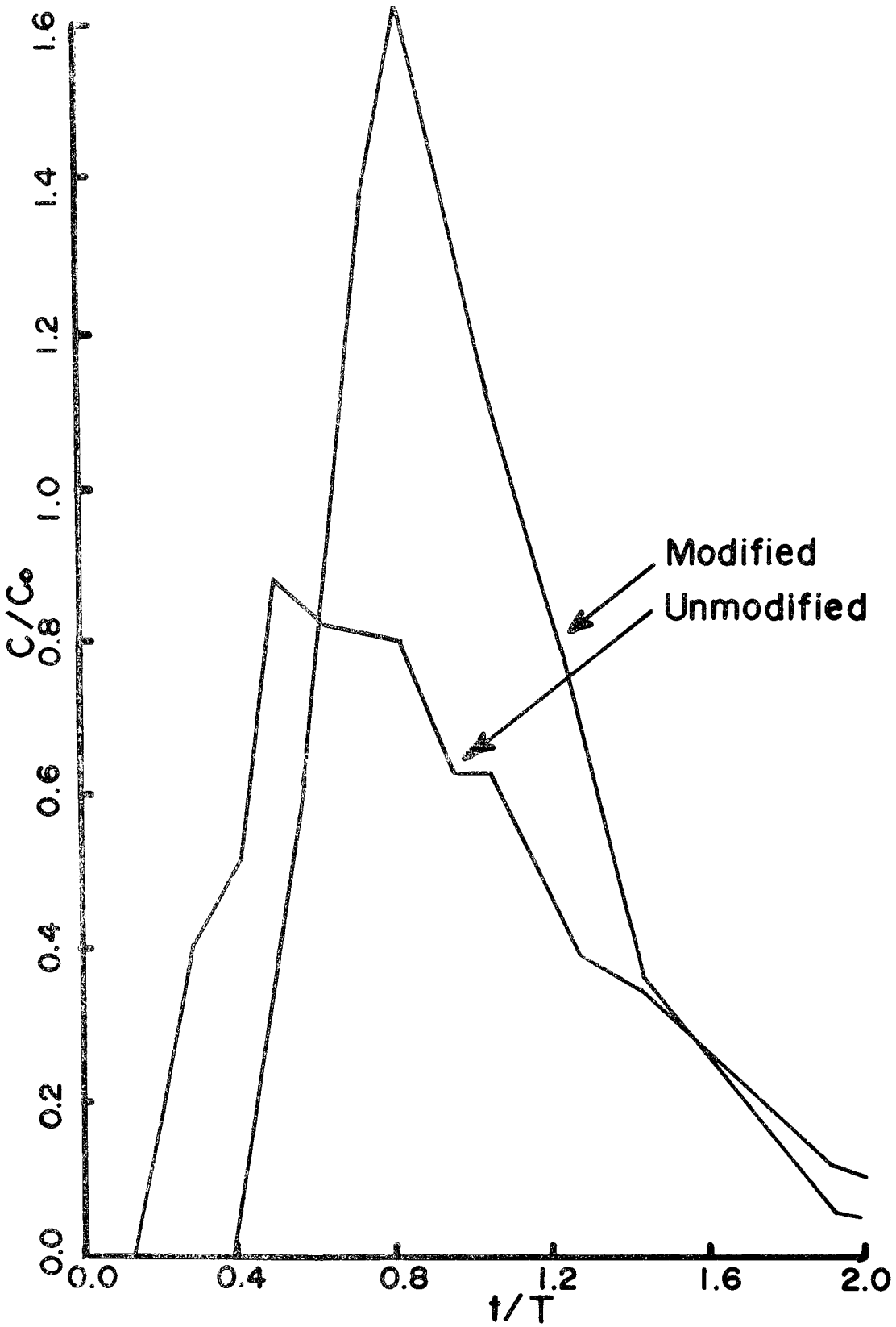


Figure 4. C-curves for Modified and Unmodified Units

modified units are presented in Figures 5 and 6. Each data point represents the mean of a two hour field experiment. Tests were conducted during mid-day periods when influent rates were relatively constant. These points when applied to a regression analysis fitting expression 6 yield:

Unmodified Unit

$$N/N_0 = (22.75/ct)^{2.96} \qquad 8$$

$$r^2 = 0.82$$

Modified Unit

$$N/N_0 = (21.26/ct)^{2.98} \qquad 9$$

$$r^2 = 0.86$$

Setting expressions 8 and 9 to an equal degree of disinfection, (N/N_0 modified = N/N_0 unmodified) demonstrates that a seven percent decrease in required chlorine dose is needed for the modified unit.

Figure 7 presents a plot of batch reactor data used to simulate ideal plug flow conditions. These data yield the following coefficients when fitted to expression 6:

$$N/N_0 = (19.00/ct)^{3.04} \qquad 10$$

$$r^2 = 0.83$$

Applying coefficients obtained for expression 5 and 10 to equation 7 yields a nine percent chlorine reduction requirement for the modified unit. Field tracer data and laboratory batch reactor data, therefore, indicate that the modified CCC unit should require nine percent less chlorine to obtain the same degree of disinfection.

Field Observations

In addition to disinfection performance evaluations, these field studies were conducted to evaluate the cost, durability and versatility of the baffles. Table 1 indicates that the material costs are relatively low and, as will be noted in the conclusion, the costs are reasonable when compared to potential savings from reduced chlorine use. Baffles at both facilities were constructed from standard size corrugated plastic sheets attached together with an epoxy resin and braced with 1" x 3" wood strips. The baffles were lightweight and could easily be placed into the chamber by two people. During the nine month experimental periods, no damage to the baffles was noted.

Higher accumulations of solids and floating materials were noted in the modified units. Such a drawback was expected because short circuit currents capable of carrying these materials through the CCC unit and out the effluent

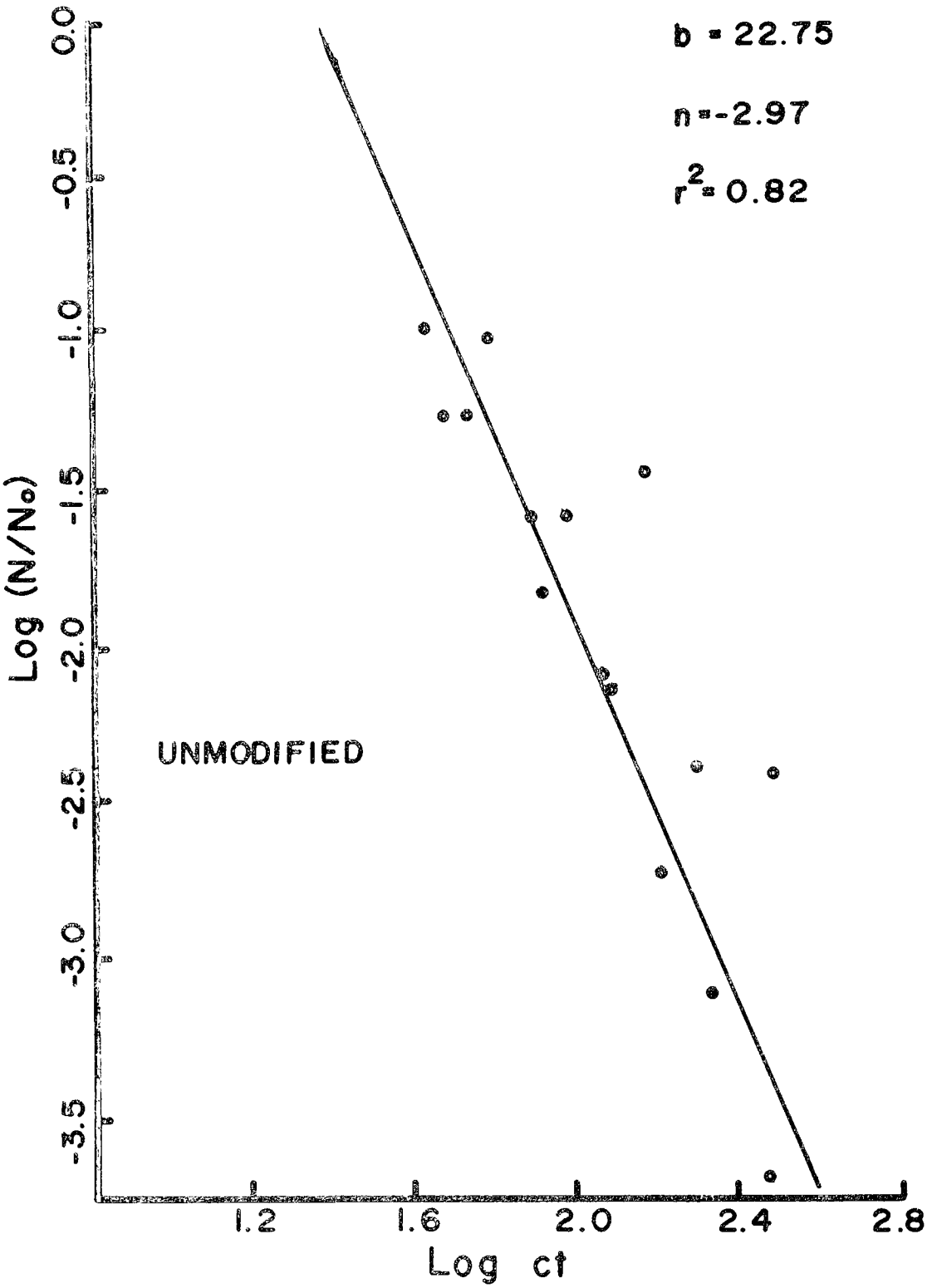


Figure 5. Unmodified Unit Disinfection Response

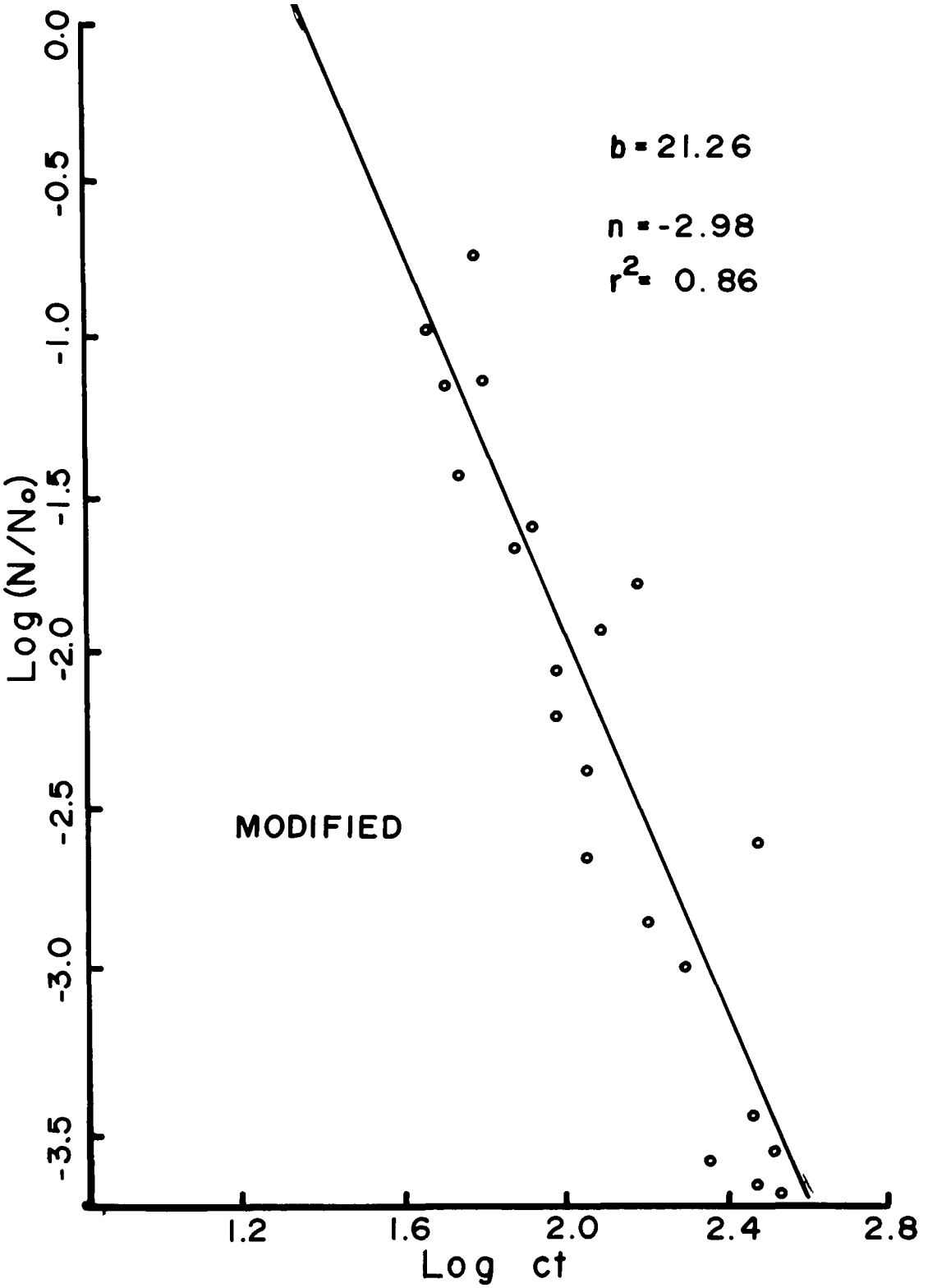


Figure 6. Modified Unit Disinfection Response

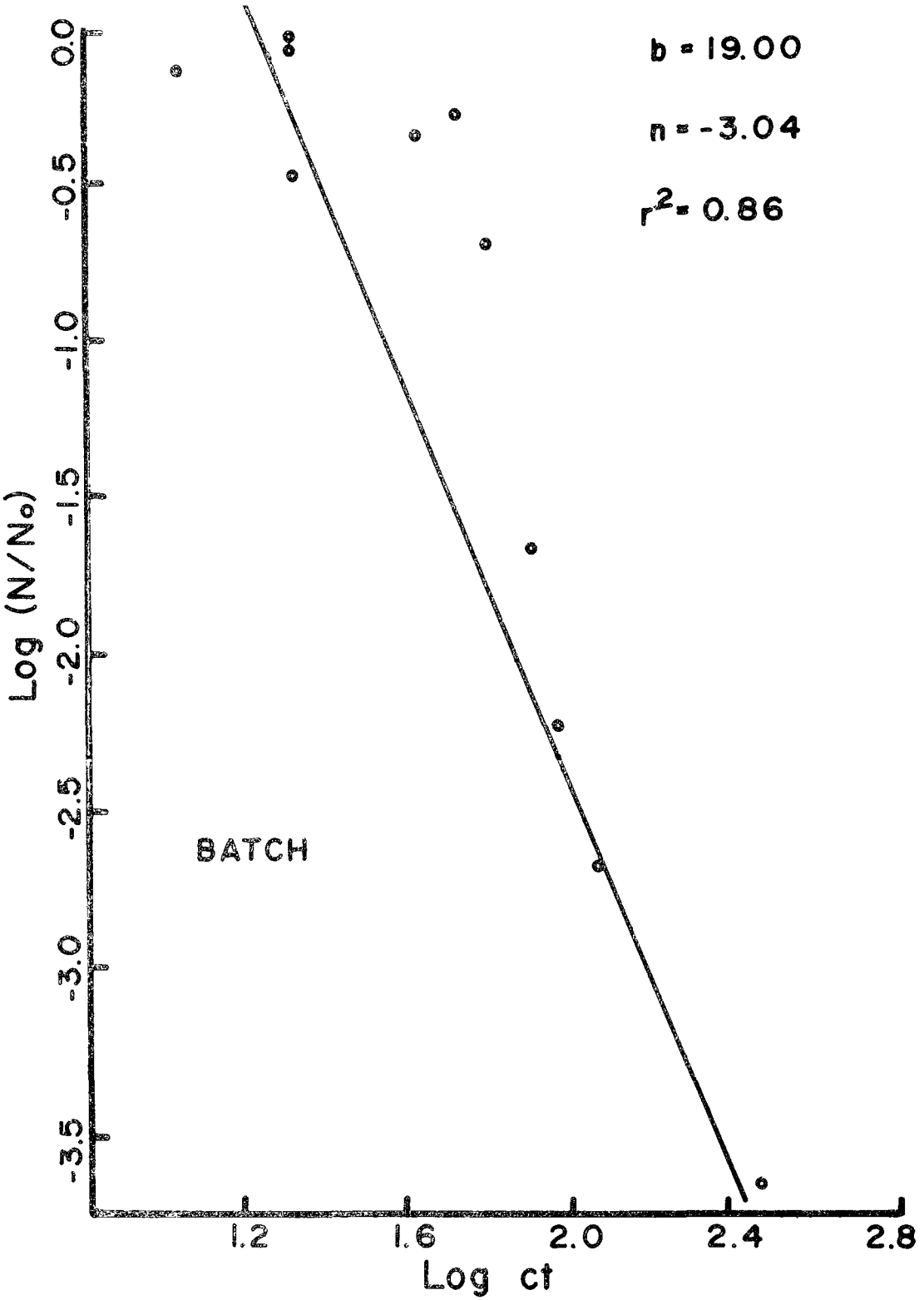


Figure 7. Batch Reactor Disinfection Response

were eliminated. Periodic cleaning of the modified unit was possible by removing the baffles. This was possible because the baffles were lightweight and were secured to the CCC wall by being placed between two guides. Permanent connection of the baffles to the CCC wall is not recommended.

Versatility of this modification scheme was partially demonstrated by these field tests because the two facilities are different sizes (see Table 1) and have slightly different CCC unit configurations (see Figures 1 and 2). Radically different configurations, however, may not respond similarly. It is anticipated that a very poor hydraulic design should benefit significantly from this type of baffle modification scheme. It should be noted that the configuration illustrated in Figures 1 and 2 was chosen because it is very common, not because extremely poor hydraulic conditions were expected.

CONCLUSION

Data presented in this paper indicates that the modified CCC unit performs at a more efficient level than the unmodified unit and will require about eight percent less chlorine to meet the same degree of disinfection. Assuming that this drop in required chlorine dose is achievable under field conditions, a decrease in chlorine cost savings could be realized. Using a chlorine cost figure of $0.22 per kg, and an average chlorine dose of eight mg/L, a 3800 m^3/d WWTP will save $243.00 per year. A comparison of this figure to the cost for constructing these baffles (see Table 1) indicates that these simple modifications are economically justifiable.

A more significant effect of improving the efficiency of existing CCC units, however, is the resulting potential for reduced impact to the aquatic environment. A thorough analysis of this potential, however, was beyond the scope of these projects. Specific areas that remain to be explored include the influence on chlorinated organic synthesis resulting from more efficient CCC units and the degree of efficiency improvement possible for different CCC unit configurations.

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3. PROBLEMS OF DISINFECTING NITRIFIED EFFLUENTS

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ABSTRACT

Several wastewater treatment plants in California have experienced some unexpected problems when trying to achieve the NPDES requirement of 2.2/100ml MPN coliform concentration in nitrified and filtered effluents. These problems do not exist in non-nitrified effluents or those containing 1.5 to 2.5 mg/l of ammonia nitrogen, or more. The nitrified effluents in question contain only trace amounts of ammonia nitrogen and nitrites.

The San Jose/Santa Clara Water Pollution Control Plant conducted a six month evaluation of the disinfection process. This included chlorine demand studies and a coliform profile of the various unit treatment processes. The chlorination - mixing - contact chamber system was designed to achieve a 2.2/100 ml MPN total coliform concentration in the plant effluent.

When the tertiary plant (nitrified and filtered) effluent went on line the chlorine required to achieve the 2.2/100 ml MPN coliform in the effluent was enormous compared to a non-nitrified effluent. Laboratory studies were made adding ammonia-N to the nitrified effluent. Considerably less chlorine was required to achieve the NPDES requirement of 2.2/100 ml MPN total coliform. Laboratory results were transferred to plant operation and were confirmed as follows: the nitrified and filtered effluent required a minimum dosage of 17 mg/l chlorine, which produced a residual of 9 mg/l at the end of 49 min. at peak dry weather flow (PDWF) (this residual contained 50-60% free chlorine). This compared to the same effluent fortified with 2 mg/l ammonia-N

requiring only 12 mg/l chlorine dose that resulted in a 7 mg/l combined chlorine residual at the same contact time.

The surprising factor in this investigation is that the combined chlorine residual was extremely more reliable in its germicidal efficiency than the free chlorine residual.

INTRODUCTION

This paper discusses the ramifications of the disinfection process in use at wastewater treatment plants required to turn out an effluent containing a maximum of 0.1 mg/l unionized NH_3-N and a total coliform concentration not exceeding 2.2/100 ml MPN. These requirements are the result of the guidelines formulated by the California State Department of Health, the State Water Quality Board and the State Fish and Game Commission. There are about fifteen treatment plants in California that are subjected to the 2.2 coliform requirements. However, not all of these plants are required to produce a completely nitrified effluent. Those that do not nitrify do not experience difficulty with the disinfection process.

The data presented here were developed over a twelve month period at the San Jose/Santa Clara Water Pollution Control Plant located on the southerly edge of San Francisco Bay. This treatment plant was first constructed as a primary plant in the early 1950's. About ten or so years later the plant was expanded into a secondary plant. The secondary effluent disinfection requirements were set at 240 MPN per 100 ml total coliforms. Nitrogen removal was not required. Disinfection to meet this requirement was achieved with about 12 mg/l dosage and a residual of 5-6 mg/l at the end of the contact chamber. The detention time at PDWF was approximately thirty minutes.

A clue to solving the problems of the future tertiary effluent occurred during the canning season when the secondary effluent lost its ammonia nitrogen content. During this period the disinfection process fell into disarray. There was not enough chlorinator capacity to achieve the 240/100 ml coliform NPDES requirement. Supernatant liquor from the digesters was added to the raw sewage at the headworks in sufficient quantity to produce a predominantly monochloramine residual. As soon as this was done the disinfection process returned to normal.

THE TERTIARY PLANT

In February 1979, the San Jose tertiary plant was put into operation. These additions to the secondary plant which composed the tertiary treatment process consisted of a nitrification unit (suspended growth system) and dual

media filters. A new chlorine contact chamber was also part of this construction project. The chlorine control system was a combination of flow pacing and chlorine residual control commonly described as compound loop control. Adjacent to the chlorine diffusers were turbine mixers followed by a specially designed serpentine chlorine contact chamber. The contact chamber had a 49 minute contact time at peak flow established by the first appearance of a dye entering at the chlorine diffusers (t_i). It was fully expected that the tertiary effluent would be of such superior quality to the secondary effluent that the chlorine required to achieve 2.2/100 ml MPN coliform concentration in the tertiary effluent would be less than that required to achieve 240/100 ml in the secondary effluent. This was based upon the assumption of the presence of a free chlorine residual in the nitrified effluent and a much lower coliform concentration (Y_o) in the filtered effluent.

It developed that the tertiary effluent exhibited an abnormal free chlorine demand, for which there was no ready explanation. At first it was thought that this was due to the presence of nitrites (one mg/l nitrite-N will consume 5 mg/l HOCl). Combined chlorine (chloramines) will not oxidize nitrites to nitrates within the time frame of wastewater treatment systems. The quality of the tertiary effluent is shown in Table 1. Examination of these data eliminates nitrites as the cause of high free chlorine demand.

Table 1. Tertiary Effluent Quality*

Parameter	Concentration, mg/l
Hardness	245
TOC	11-14
TDS	800-900
Org. N	1.3-2.3
NO_2-N	0.02-0.03
NH_3-N	trace

*In spite of this high quality filtered effluent the disinfection system was not able to meet the 2.2 coliform requirement. Table 2 illustrates this dilemma.

Table 2. Final Effluent Total Coliform Concentration After 49 Minutes Contact Time

Total Cl_2 Residual	Total Coliform MPN/100 ml
6.5	79
6.5	23
6.5	23
7.4	4
7.4	2
7.4	2
9.1	2
9.1	2
9.1	2
5.1	2
5.1	13
5.1	7

The residuals shown in Table 2 were measured by the forward amperometric procedure. These residuals contain about 60 percent HOCl and the rest titrates as "dichloramine." The latter species is probably composed of a variety of non-germicidal organochloramines as a result of the organic-N present.

INVESTIGATION OF TERTIARY EFFLUENT

Owing to the poor performance of free chlorine residuals it was decided to investigate the following characteristics of the disinfection process.

a) Verify the t_i contact time at PDWF (t_i = first appearance of dye at the exit of the contact chamber).

b) Determine the chlorine demand for various chlorine dosages and contact times. Compare with chloramine species.

c) Establish a coliform profile (without chlorination) beginning with the secondary effluent and continuing to the final effluent.

d) Determine chlorine dosage required to achieve 2.2/100 ml MPN total coliforms at contact time t_i.

e) Compare germicidal efficiency of free chlorine versus combined chlorine residuals.

f) Determine the benefit, if any, of applying the postchlorination dose at two separate points in the treatment train.

RESULTS

Contact Time

The contact chamber dye test revealed a t_i of 49 min. at PDWF. The contact times used for all samples analyzed in the laboratory were 5, 30, 49 min. and 24 hours.

Chlorine Demand Studies

The secondary effluent residuals were examined for total chlorine residual. This was done by the back titration method using an amperometric titrator.

The nitrified effluent was examined for free chlorine, mono, and dichloramines. This was done with a separate titrator using the forward titration procedure.

Figure 1 illustrates the chlorine demand of the secondary effluent which contains only combined chlorine residual. Figure 2 illustrates the same for the nitrified filtered effluent which contains about 60 percent free chlorine. Figure 3 illustrates the same for a nitrified filtered effluent that has been fortified with enough NH_3-N to provide a 6:1 chlorine to nitrogen wt. ratio.

Coliform Profile - A summary of the coliform levels in various stages of the San Jose plant is presented in Table 3.

Table 3. Coliform Profile of San Jose Plant

Location	Total Coliform MPN/100 ml		
	Max	Min	Median
Secondary Effluent	9.2×10^6	49,000	1.7×10^6
Nitrified Effluent	1.6×10^6	23,000	110,000
Filtered Effluent (no prechlor)	160,000	200	23,000
Filtered Effluent (with 8 mg/l prechlor, Cl_2 res. 1.2 mg/l, contact time 17 min)	1,300	<20	80

Coliform Kill Study

This study was performed concurrently with the chlorine demand studies. Each sample was divided into three replicates. Each of these replicates was then transferred to five tubes for four different dilutions: 10 ml., 1 ml 0.1 ml and 0.01 ml. This amounts to 20 tubes for each of three replicate samples. The secondary effluent and the filtered effluent were all subjected to this same examination. The discussion of these results follows below.

a) Secondary Effluent. The objective was to find if possible the chlorine dosage to provide a 2.2/100 ml MPN effluent using 49 min. as the contact time. This study provided a most important clue. Both 12 and 15 mg/l chlorine dosages at 49 min. contact time were investigated. Some of the 12 mg/l dosages resulted in 2.2/100 ml MPN coliform and some resulted in counts as high as 33/100 ml MPN. The 15 mg/l dose was more consistent owing to a higher residual at 49 min. contact time. At this dosage the residual that achieved 2.2/100 ml MPN coliform were on the order of 8 mg/l. This fits the Collins model.

b) Nitrified Filtered Effluent. The quality of this effluent is considerably superior to the same effluent without filtration. The Y_o coliforms are much lower and the organic nitrogen is significantly less. The latter means that the combined chlorine residual will contain less non-germicidal organochloramines. In spite of the superior quality of this filtered effluent the germicidal efficiency of the free chlorine residual was

disappointing to say the least. For example: the median Y_o from April through May 1980 was 23,000/100 ml MPN coliforms. This calculates to a ct = 91 in the Collins model. So, for a 49 minute contact time, disinfection should be possible with a 2 mg/l total chlorine residual. Laboratory and plant results have shown that the total residual for the nitrified filtered effluent must be on the order of 9-10 mg/l for consistent results. The San Jose plant does not have enough sulfonator capacity to dechlorinate this much residual.

Comparison of Germicidal Efficiency of Free Versus Chloramine Residuals

Owing to the above dilemma it was decided to experiment with artificial chloramine residuals and compare their efficiency against the free chlorine residuals. This was done by adding ammonia nitrogen in various Cl to N ratios to the nitrified effluent. Chlorine dosages used were 10, 12, and 15 mg/l. Cl to N ratios investigated were 6 to 1, 8 to 1, and 10 to 1. All of the dosages using chlorine to ammonia N at 6:1 produced an effluent coliform concentration of 2.2/100 ml or less without exception. From these tests it was patently clear that a chloramine induced residual can outperform a free chlorine residual by a wide margin at the San Jose plant. The breakpoint curve for this ratio is shown on Figure 4.

Effect of Mixing

The San Jose investigation has put the subject of mixing as it effects disinfection efficiency into an entirely different perspective. It appears that the most important reason for superior mixing in wastewater disinfection is to convert as soon as possible the free chlorine in the chlorine solution to chloramines. This minimizes formation of organic-N compounds which have low disinfection efficiency.[3] Laboratory experiments demonstrated the difference between good mixing and poor mixing.

The results shown in Table 4 are average residuals of several experiments with wastewater containing 2 mg/l artificially added ammonia-N and subsequently dosed with 12 mg/l Cl and a contact time of 60 minutes.

Table 4. Average Chloramine Residual in Wastewater Effluent as a Function of Degree of Mixing

	Monochloramine	Dichloramine
Good Mixing	7.25	1.25
Poor Mixing	3.25	3.45

In plant practice, good mixing is considered achieved when the velocity gradient, G, in the mixing chamber approaches 1000 (3).

DISCUSSION

Collins Model

The Collins mathematical model (3) is used to establish chlorine dosages at given contact times for combined chlorine residuals (non-nitrified effluents). It is a good basis for comparison of free versus combined residuals.

The Collins equation (1) is as follows: $y/y_o = (1 + 0.23\ ct)^{-3}$

where: y = 2.2/100 ml MPN (NPDES limitation),
y_o the median coliform concentration before chlorination
c - chlorine residual (mg/l) at the end of time t (minutes)
$t = t_i$ first appearance of dye at the end of contact chamber.

This investigation was not an exercise to prove or disprove the Collins model, which has served so well in evaluating the efficiency of disinfection systems of non-nitrified effluents. It has been used here to compare the efficiency of combined chlorine residuals versus combined residuals that are measured as predominantly free chlorine. The conclusion based upon the San Jose study is that the Collins model is not applicable for combined residuals that are measured as predominantly free chlorine, e.g., 50 percent or more. The above example for the nitrified filtered effluent indicates that the Collins model predicts a total chlorine residual of 2 mg/l at 49 min. contact to achieve a 2.2/100 ml coliform MPN. However, in reality it was found that the required total residual to achieve the 2.2 figure was closer to 9 mg/l. See Table 2. These residuals contained about 60 percent free chlorine.

Obviously the Collins model does not fit nitrified effluents. This is indeed a surprising development. The reason for this lies somewhere in the chemistry of the higher reactivity of free chlorine, hence its higher consumption. However, analyzing the 9 mg/l residual referred to above, this contained about 5 mg/l free chlorine. The remainder titrated as dichloramine.

Chloramine Residuals

Chloramine residuals occurring in wastewater always contain a mixture of monochloramine and dichloramine. The dichloramine is most probably due to the presence of significant concentrations of organic nitrogen (1-3 mg/l). It is presumed the chlorine residual species that titrates as the dichloramine fraction in a wastewater is probably a variety of organochloramines having little or no germicidal efficiency (4). Therefore, the objective is to get a chloramine residual with the highest percentage possible of the monochloramine fraction. Fast and thorough mixing of the chlorine with the wastewater is the key factor to achieve this result.

Compared to the germicidal efficiency of free chlorine, chloramines have been thought of as inferior. Some researchers in the 1970's, however, have discovered that if given enough time (40-60 minutes), chloramines are nearly as effective as free chlorine (4). Selleck et al (2). have shown that the most germicidal combined chlorine residual (chloramine) appears to occur when the chlorine to ammonia-N ratio is on the "breaking " side of the B-P curve. This is between points A and B on Figure 4. At point A the ratio at the hump of the curve is nominally 5 to 1 Cl to N by weight.

The coliform kill study revealed the maximum kill of coliform organisms with the least chlorine dosage occurred at a Cl to N ratio of 6:1 for the available contact time of 49 minutes at PDWF.

Comparison With Other Plants

An integral part of this investigation was to visit other plants with similar effluent requirements for coliforms and ammonia nitrogen. Including San Jose, a total of ten plants were visited. All of them were in California, and all but three were in the San Francisco Bay area. The plant processes and operation varied considerably, depending upon whether water reclamation was involved and whether or not the receiving waters or the end use of the effluent could tolerate ammonia-N in the effluent. (The NPDES requirement for ammonia-N is for the receiving waters and not the effluent.)

At one nearby plant, a 2.8×10^5 m^3/d capacity investigation was begun in 1980 to find out how energy might be saved if nitrification were not complete (1). It was found that NH_3-N concentration above 2 mg/l resulted in a 25 percent reduction in the chlorine demand. The investigation did not support the dogma that free chlorine is a better disinfectant than combined chlorine. Moreover, it was revealed that the final effluent dosage could be reduced without adversely affecting the bacteriological quality of the effluent. When the effluent contained 2-3 mg/l ammonia-N, the 2.2/100 ml MPN coliform concentration could be achieved on a consistent basis. When complete nitrification was practiced it was not uncommon to require final effluent chlorine doses from 14 to 20 mg/l. The contact time was one hour

Four plants with one hour contact times required between 18 and 25 mg/l chlorine to achieve the 2.2. These dosages resulted in total chlorine residuals of 9 to 14 mg/l. The free chlorine residual fraction varied from 45 to 85 percent of the total. One small plant required a 50 mg/l dose which resulted in a 35 mg/l residual.

Another plant was experiencing similar high chlorine dosage requirements but was further plagued by intermittent ammonia spikes in the effluent. This resulted in the conversion of the free chlorine to combined chlorine. An attempt has been made to try and control the nitrification process to leave a 2-4 mg/l ammonia-N residual. This has proved to be difficult. Owing to a limitation of chlorinator capacity a savings in chlorine consumption has not been realized. However, where the ammonia peaked as the plant flow increased the coliform kill increased so that compliance was achieved (2.2 MPN).

In every investigation there is always an exception. One plant, with a flow range of 34,000-53,000 m^3/d, using suspended growth reactors for nitrification and dual media filters, produced a completely nitrified effluent and achieved 2.2/100 ml MPN coliforms (7 day median) in the effluent with a dosage of 7-8 mg/l that resulted in a 3 mg/l total chlorine residual after about 60 minutes contact time.

Another plant was found to be unique because the effluent was not filtered and the chlorine dosage control was based upon a free residual (in the presence of combined residual) and the plant consistently turned out a completely nitrified effluent that met a 2.2/100 ml MPN coliform concentration. The chlorine dosage was 8 mg/l, contact time at peak flow was 49 minutes (as determined by tracer studies) and the total chlorine residual at the end of the contact chamber was about 3-4 mg/l. This was a well oxidized effluent (activated sludge) and the coliform concentration before chlorination was on the order of 140,000/100 ml MPN. The Collins model predicted a residual of 3.45 total chlorine residual. This plant was in a suburb so that effluent was primarily domestic wastewater. All of the industrial discharges were pretreated before entering the collection system.

CONCLUSIONS

a) A nitrified effluent, in spite of filtration, demonstrates a much higher chlorine demand than a non-nitrified, non-filtered effluent.

b) The higher demand described above is probably due to the higher reactivity of free chlorine compared to combined chlorines.

c) Both the laboratory and plant scale investigations determined that a 6:1 Cl to N ratio with a 12 mg/l chlorine dose proved to be the most germicidal ratio.

d) Plant scale operation proved that the addition of a 12 mg/l dose of chlorine added to the effluent containing 2 mg/l of ammonia-N can produce an effluent which will consistently meet the NPDES requirement of 2.2/100 ml MPN total coliforms.

e) The most germicidal combined chlorine residual proved to be one that is composed of about 75-80 percent monochloramine. The remainder titrates as "dichloramine" which is considered to be organochloramines of low germicidal efficiency. These chloramines are probably a result of the organic nitrogen present in wastewater effluents.

f) The laboratory experiments proved that good mixing was required to achieve residuals containing 75-80 percent monochloramine.

g) Adequate mixing should occur when the velocity gradient G approaches 1000.

h) Where mixing is poor the monochloramine species drops to about 50 percent of the total residual. This results in lower germicidal efficiency together with a higher consumption of chlorine.

i) Plant scale operation also proved that the 2 mg/l addition of ammonia-N to the effluent did not jeopardize the NPDES requirement of 0.025 mg/l un-ionized ammonia nitrogen (NH_4OH) in the receiving waters (lower San Francisco Bay).

j) The chlorine dosage and residual requirement to achieve an MPN coliform concentration of 2.2/100 ml in the nitrified effluent containing 2 mg/l ammonia-N was demonstrated to be 5 mg/l and 2 mg/l respectively less than for the nitrified effluent, without any ammonia-N. See Figure 5.

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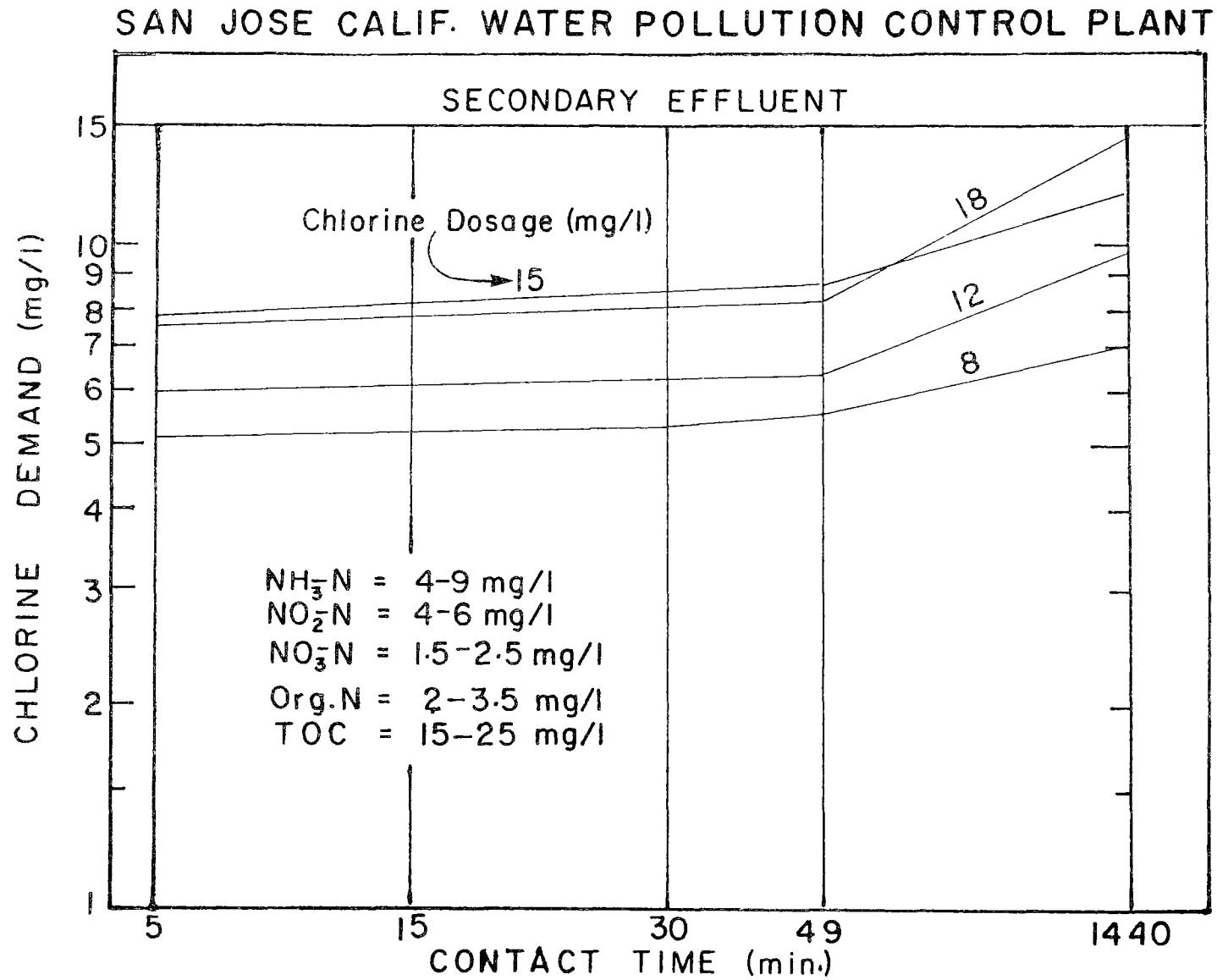

Figure 1. Chlorine Demand Secondary Effluent

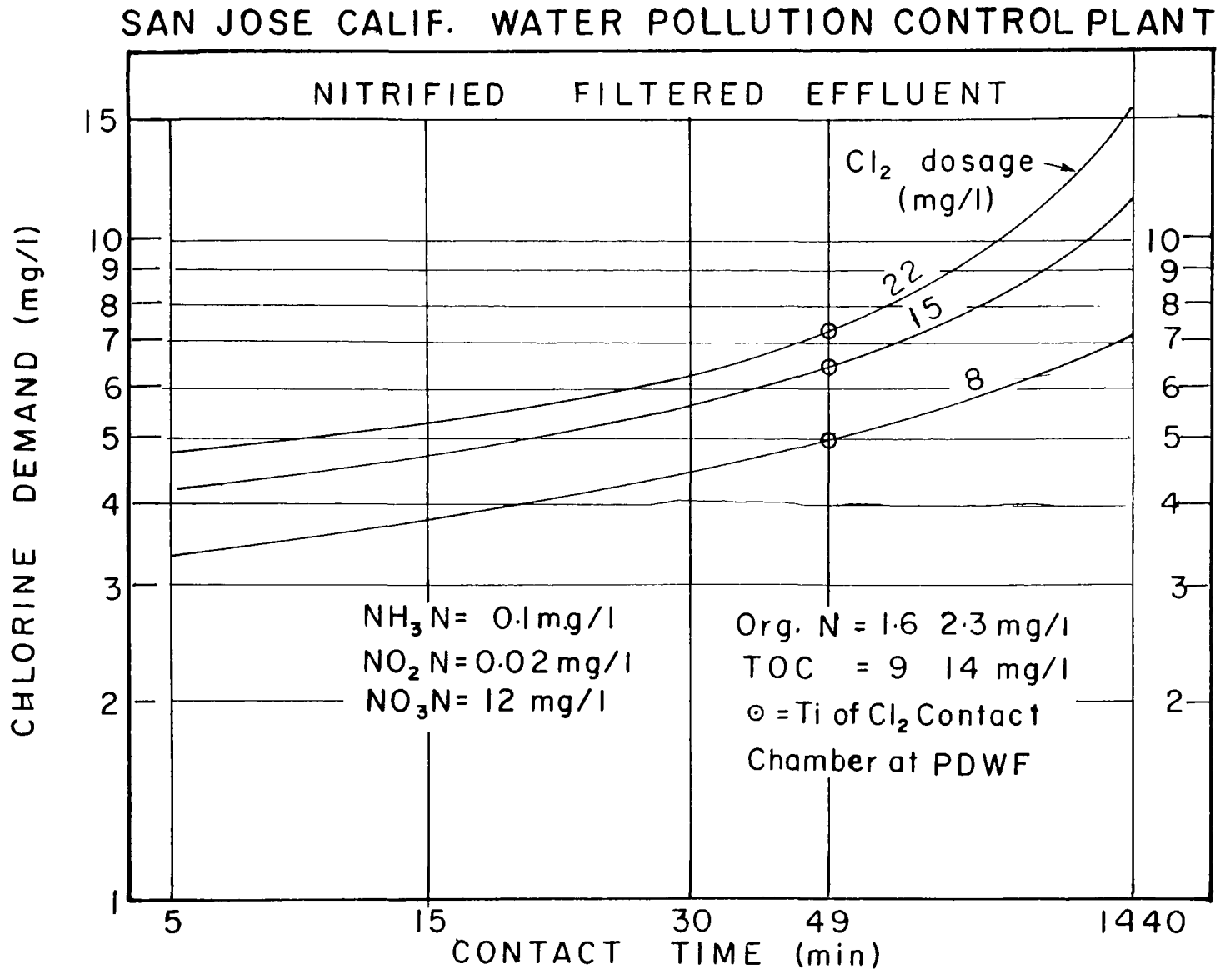

Figure 2. Chlorine Demand Nitrified Filtered Effluent

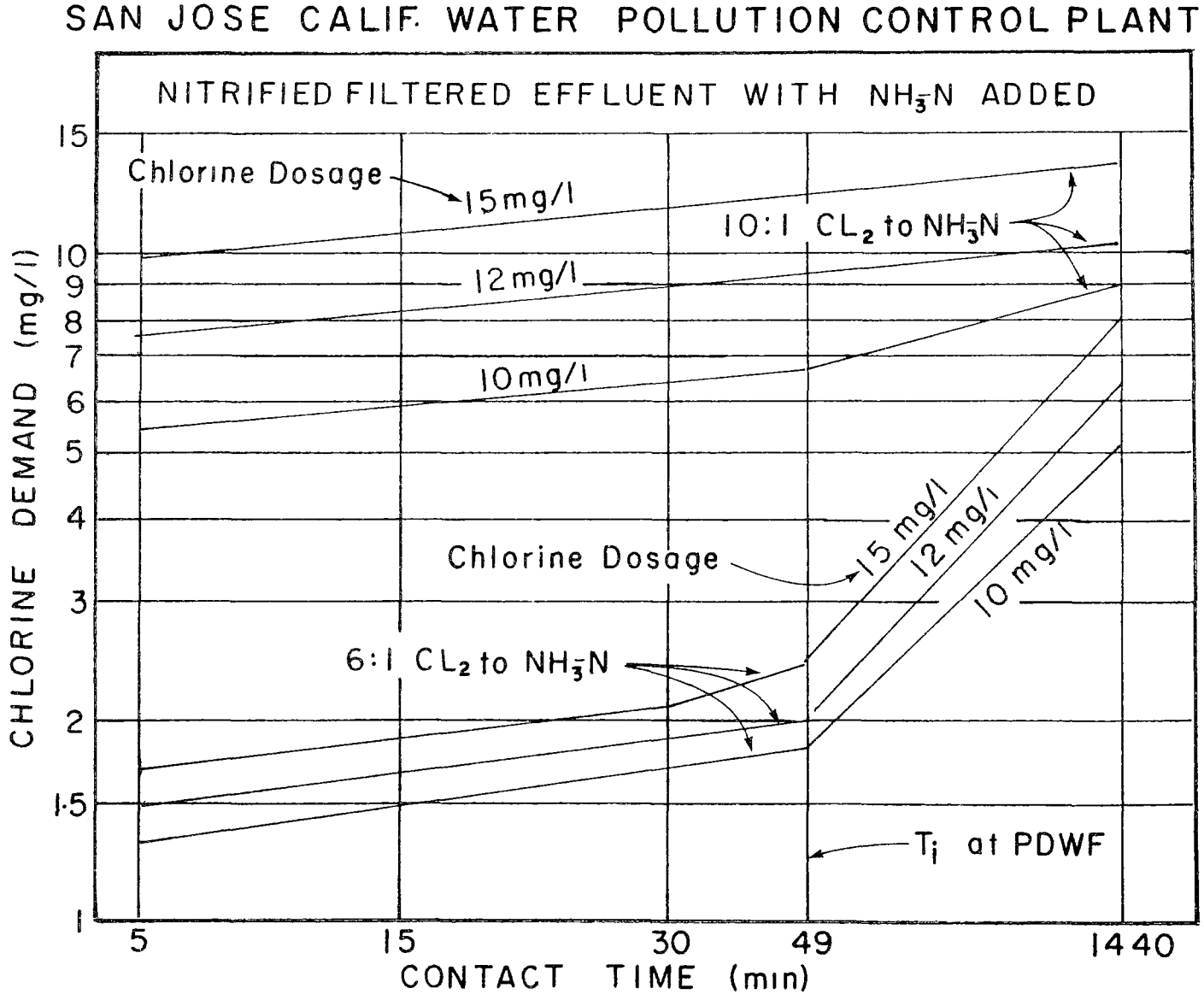

Figure 3. Chlorine Demand Nitrified Filtered Effluent with Ammonia-N Added

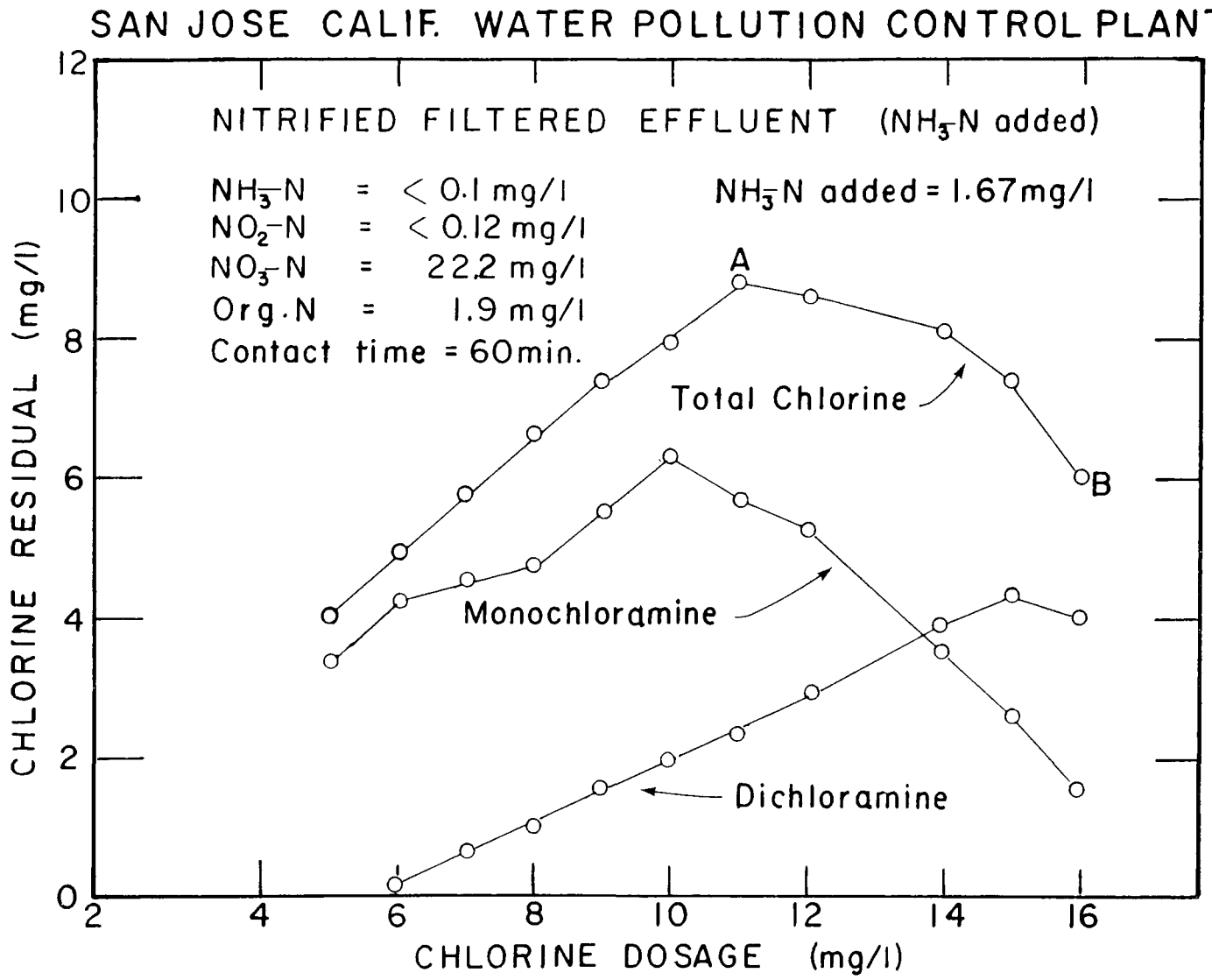

Figure 4. Breakpoint Curves for Chlorine to Nitrogen Ratio 6:1 by Wt.

SAN JOSE CALIF. WATER POLLUTION CONTROL PLANT

SUMMARY

Chlorine dosage and residual requirements to achieve an MPN coliform conc. of 2.2/100ml

	Dosage	Residual
Nitrified and Filtered Effluent	17	9*
Same as above except that 2mg/l NH_3-N has been added	12	7

Note: Residuals are those measured at the end of the contact chamber. This amounts to 49m contact time at PDWF.

* These residuals are 50 to 60% free. About 90 to 95% of the remainder titrates as dichloramine, the rest as monochloramine.

Figure 5. Chlorine Dosage and Residual Requirements to Achieve MPN Coliforms of 2.2/100 ml

4. OPERATING EXPERIENCE DISINFECTING SECONDARY EFFLUENT WITH PILOT SCALE ULTRAVIOLET UNITS

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ABSTRACT

The effectiveness of disinfection of secondary effluent from an activated sludge wastewater treatment plant was tested using pilot scale units from four different manufacturers. Each unit was operated to maintain an effluent fecal coliform concentration of less than 200 per 100 ml. Each unit was capable of maintaining the coliform standard when it was clean. However, keeping the units clean was the most serious problem observed with each unit. Flushing with citric acid proved to be an adequate method of cleaning the units. Units employing an ultraviolet sensor provided an early warning of impending failure. Results of this study will be used for the possible design of a full scale system.

INTRODUCTION

The Madison Metropolitan Sewerage District (MMSD) operates the 50MGD Nine Springs Wastewater Treatment Plant in Madison, Wisconsin. This plant provides primary and secondary treatment with anaerobic digestion and land application of residual solids. Plans are currently being developed to upgrade the treatment at the plant to Advanced Secondary Standards. A portion of this upgrading deals with the replacement of the obsolete chlorination equipment.

In the original Environmental Impact Statement the Environmental Protection Agency (EPA) commented adversely on the continued use of chlorine for disinfection. Therefore, the District's consultants considered the use of ozone and ultraviolet light in the update to the Facilities Plan. Preliminary calculations indicated that ultraviolet light (UV) would be more cost effective. Since at this time there was very little documented experience with the use of ultraviolet light to disinfect secondary effluent, the District decided to pilot test at least one manufacturer's unit. This first unit was put on line in December 1979. By the end of the test period, September 1981, units from four manufacturers had been tested.

METHODS

The purpose of the pilot tests was to determine what operational and maintenance problems could be expected and how to design to overcome these problems. Features offered on the various units were to be compared

and evaluated for inclusion in a full scale facility. No attempt was made to compare theories of operation or to measure the ultraviolet dose applied or the power used. Values measured on these test units would have been invalid for scale-up since each manufacturer was continuing to modify and refine his design.

Secondary effluent was pumped into each unit. Because of lack of sufficient ancillary equipment all four units could not be evaluated simultaneously. Each unit was initially operated at the flow rate recommended by its manufacturer. Based on the disinfection results achieved, the flow rate was varied accordingly. The effluent from the treatment plant will be required to attain a monthly geometric mean fecal coliform count of less than 200 per 100 ml. As a measure of reliability it was the goal of the tests to operate the pilot units so the value of 200 fecal coliforms per 100 ml was never exceeded. The unit was defined as being in the failure mode when this value was exceeded.

Samples of the influent and effluent of the units were analyzed daily for fecal coliforms by the membrane filtration method. The total and volatile suspended solids concentration of the influent were also analyzed as was the absorbance of the influent at 254 nm wavelength.

RESULTS

Unit A

The first unit evaluated, Unit A, was a standard production model rated at 100 gpm. Twenty-four UV lamps in an array four high and six wide comprised the disinfection chamber. Each lamp was enclosed in a quartz tube with an outside spacing of three-fourth inch between the tubes. Flow through the unit was perpendicular to the longitudinal axis of the lamps. The inlet to the unit was baffled as was the discharge with the free water surface being controlled by the effluent baffle. Teflon discs encircling but not touching the tubes were used as mechanical wipers. These discs were attached to a rack which slid along the longitudinal axis of the tubes at adjustable time frequencies. Light emitting diodes (LED's) on the outside of the unit indicated lamps which were operating properly. Also included in this unit was a sensor which measured the amount of light transmitted to it at 254nm. This sensor was housed in a quartz tube similar to those housing the UV lamps. Three circumstances could be responsible for a decreased reading from the sensor: 1) an increase in the UV absorbance of the water, 2) reduced output from the UV lamps, or 3) coating of the quartz tubes. A receiver on the side of the unit indicated the sensor reading on a scale labeled "relative transmittance".

The results obtained from this unit are shown in Figures 1 and 2. During the first four weeks of operation the disinfected effluent exceeded 200 fecal coliforms per 100 ml on only one occasion. After three weeks of operation the relative transmittance reading began to decrease. By the fourth week the transmittance reading was less than 50 percent of full scale,

and the effluent fecal coliform reading was consistently above 200 per 100 ml. As seen in Figure 1, the decline in the relative transmittance reading seems to correspond to an increase in the UV absorbance of the water. However, this was not the only cause of the higher effluent fecal coliform counts. Upon draining the unit it was discovered that the tubes were coated with a white substance. The tubes were removed from the unit and washed with hydrochloric acid. Further investigation of the coating indicated that it contained calcium, magnesium, and iron. Compared to the concentrations in the water, the iron seemed to be depositing in a higher ratio. Although it is known that iron readily absorbs UV light, no explanation could be found for its deposition on the tubes.

After cleaning the tubes they were placed back in the unit, and adequate disinfection was again attained. However, the tubes continued to scale. The results of this situation are shown in Figure 2. When the unit was returned to service after cleaning on February 23, March 7, and March 15, the reading on the relative transmittance meter rose to 100 % and good disinfection results were obtained. A method to adequately prevent the inhibitory coating of the tubes was not found.

Unit B

Two units were tested from manufacturer B. The first was a 10 gpm upflow unit containing four lamps which were enclosed in quartz tubes. Flow through this unit was parallel with the longitudinal axis of the tubes. Cleaning was provided by an ultrasonic system. An ultrasonic transducer was mounted in the bottom of the unit with the ultrasonic energy being generated in parallel with the longitudinal axis of the tubes. Three ports were spaced along the length of the unit to accept a removable sensor. This sensor was similar in function to the one on Unit A. On most occasions the resistance in ohms was read across the sensor at each port. The sensor ports were spaced at varying distances from the ultrasonic transducer to determine the effective range of the ultrasonics. Results seemed to indicate that the ultrasonic cleaner was able to keep the quartz tubes clean, but did not keep the sides of the unit or the sensor ports clean. Figure 3 shows typical results for this unit.

The second unit to be tested by manufacturer B had six lamps and was a fully enclosed unit. As in the first unit the flow pattern was parallel to the longitudinal axis of the lamps. However, the flow through the unit was horizontal. An ultrasonic transducer was placed along the bottom of the unit so that the ultrasonic energy moved perpendicular to both the quartz tubes and the flow pattern. Only one sensor port was built into this unit. Samples were collected when the unit operated at flow rates of 20 to 40 gpm.

Both units performed well when clean. However, the quartz tubes became coated with a scale formation just as with Unit A. Whenever the scaling had increased to the point that the target fecal coliform level was not being achieved, the unit was cleaned with a citric acid

Solution. Run times between chemical cleanings ranged from two to eight weeks. Figure 4 shows the results of the longest run of eight weeks.

Unit C

A totally different design was presented by the manufacturer of Unit C. Six teflon tubes connected in series conveyed the secondary effluent through an array of UV lamps. Each 1-1/2 inch diameter teflon tube was surrounded by four UV lamps. Reflectors were attached to the lamps to direct the light to the teflon tubes. This unit did not have an ultraviolet sensor, although it did have an amperage meter to show total current draw by the lamps. Since the output of the UV lamps is dependent on the lamp temperature, a thermometer was installed to measure the temperature near one of the lamps. It was found that during the summer one of the housing panels had to be removed from the unit to reduce the lamp temperature to a satisfactory value.

The stated advantage of this unit was that the scale formation that plagued the quartz tube units would not affect the teflon tubes. Unfortunately, a coating also formed on the teflon tubes. As with the other units this coating was easily removed by circulating a solution of citric acid through the unit. The unit was operated at flow rates between 15-40 gpm. Allowable run times between chemical cleanings ranged from less than one week to nine weeks. Typical results are shown in Figures 5,6, and 7.

Unit D

The last unit to be evaluated was operated for only a short period of time. Unfortunately, the manufacturer supplied a unit designed for industrial rather than municipal use. The cylindrical unit contained 66 UV lamps enclosed in quartz tubes. Flow entered the unit and was split to allow it to run perpendicular to the longitudinal axis of the lamps. A mechanical wiping system composed of teflon washers around the quartz tubes was actuated on a variable time frequency. This unit did not contain a UV light sensor. Normal flow rate through the unit was 80 gpm.

Figure 8 presents the fecal coliform results obtained with Unit D. When the unit arrived the mechanical wiping system was jammed. The first set of data collected was obtained when the wiper system was not working. After the wiper was replaced, the second and third sets of data were obtained. The longest run in which reasonable results were achieved was three weeks.

DISCUSSION

As in any comparison of equipment each unit had its favorable and unfavorable features. Each unit was able to consistently achieve fecal

coliform concentrations below the target level when it was clean. Keeping the unit clean proved to be the major operational problem. Both units B and C were able to operate for over two months at one point without requiring cleaning. However, on other occasions both units only operated properly for a week. The cause of the scale formation could not be determined. Only water temperature correlated to any extent, with lower temperatures seeming to favor longer run times. Because of the varying frequency of scale formation a conclusion could not be drawn on whether the mechanical wipers, ultrasonics, or teflon tubes were effective in extending the run time between chemical cleanings. The only conclusion to be drawn was that citric acid was an adequate cleaning solution. Any full scale unit to be installed at Madison would be designed with a chemical cleaning system. Included in this system would be a cleaning solution mix tank, circulating pumps, piping connecting the solution tank to the units, and a drain line. Since a unit would have to be removed from service to clean it, the units would have to be built in modules to allow for adequate disinfection in the remaining units while one unit was being cleaned.

The UV sensors proved to be a valuable operational tool. Obviously they can not be used to measure ultraviolet dose, but they can give an indication of the results that can be expected. The fecal coliform results for Unit A were grouped according to a combination of the absorbance value of the wastewater and the relative transmittance reading of the UV sensor. The geometric mean value of each group was then calculated as shown in Figure 9. Although the resulting effluent fecal coliform values were dependent on both the absorbance of the water and the sensor reading, as long as the sensor was reading above 55% adequate results were obtained. It seems reasonable that a similar relationship could be developed for this type of unit at a different treatment facility. Since ultraviolet disinfection does not result in a measureable residual as in chlorination, some method is needed for the operator to determine if his unit is performing adequately. Use of the UV sensors may be a satisfactory method.

The same approach was used with the sensor readings of the first unit tested from manufacturer B. Figure 10 shows the increase in the sensor readings during a typical run. Since a sensor reading was being taken at three sites on the unit, they were added to obtain a "total resistance" reading. The fecal coliform results for all samples analyzed on this unit were then plotted against the total resistance reading as shown in Figure 11. This graph seems to indicate that when the total resistance was less than 18,000 ohms effluent fecal coliform values less than 200 per 100 ml were assured. When the total resistance was above 24,000 ohms poor results were always achieved. Between these two values the effluent fecal coliform number could not be assured. By using this type of monitoring system the operator would be able to know when he was approaching conditions which would require him to chemically clean his unit. He would when be able to react to the situation before he discharged effluent with fecal coliform values above his limit.

Units A and D were equipped with LEDs to indicate proper operation fo the UV lamps. A meter for each lamp performed this same function on the units from manufacturer B. Unit C was equipped with a meter indicating current draw by the unit. Although low current draw readings indicated that one or more lamps were out, the ammeter was not capable of indicating which specific lamp had failed. As a matter of practicality, the LEDs provided the necessary information at a much lower cost than the meters of Unit B. The meters showed the loss of efficiency of each lamp as it aged, but this information could be obtained by recording the operating time of each lamp.

CONCLUSIONS

1. Each unit was capable of consistently attaining an effluent fecal coliform count of less than 200 per 100 ml when it was clean. Each unit was plagued by a scale formation which limited typical run times from one to eight weeks.

2. Citric acid proved to be an adequate chemical cleaner. Any unit to be installed at the Nine Springs Wastewater Treatment Plant would be equipped with a chemical cleaning system.

3. Since a unit has to be removed from service during cleaning, an ultraviolet disinfection system should be designed in a modular fashion. This will allow for continued disinfection while one unit is being cleaned.

4. For the units which were equipped with ultraviolet sensors, a relation could be developed between sensor reading and effluent fecal coliform count. Each unit should be equipped with at least one sensor to allow the operator to develop a relationship which he could use to indicate if the unit was operating properly.

5. LEDs indicating operation of each lamp were useful and should be included on all units. Meters indicating lamp output are useful but probably could not be justified on a large scale.

ACKNOWLEDGEMENTS

The Madison Metropolitan Sewerage District wishes to thank the four manufacturers who provided units for the study. The author wishes to express his appreciation to the analysts in the Nine Springs Laboratory who performed the many analyses required to make these tests worthwhile.

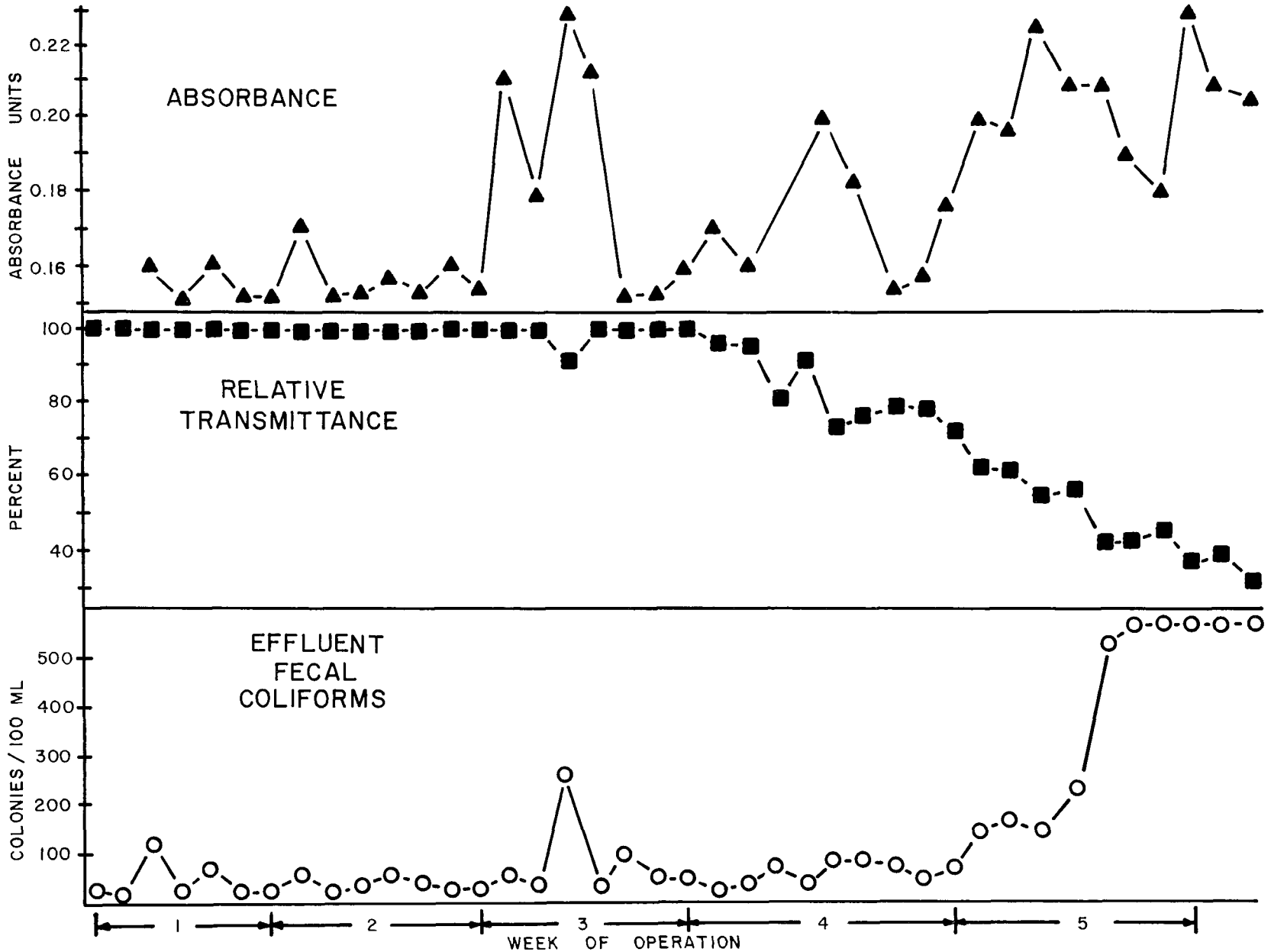

Figure 1. Unit A Results - Run 1

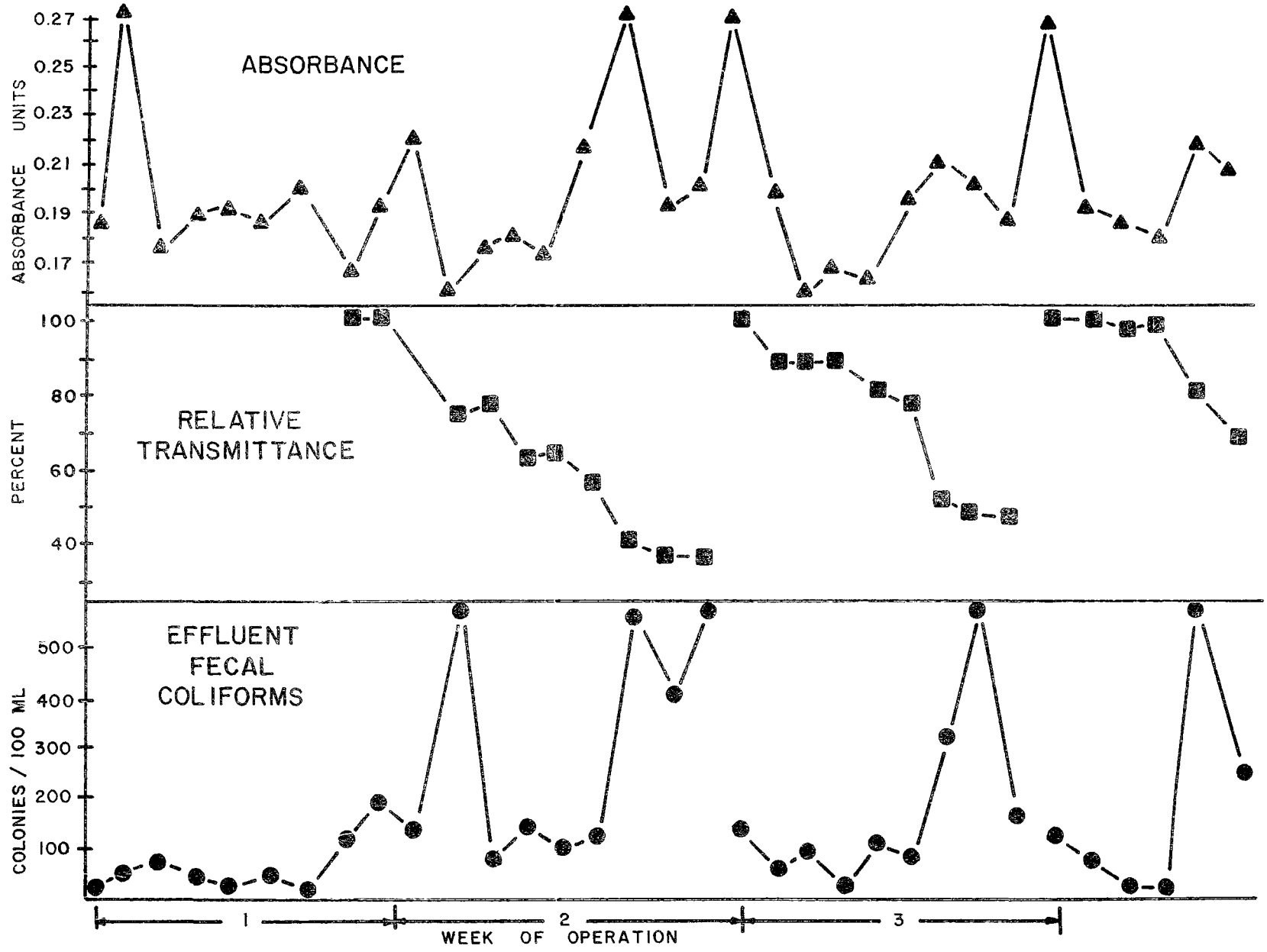

Figure 2. Unit A - Runs 2,3,4

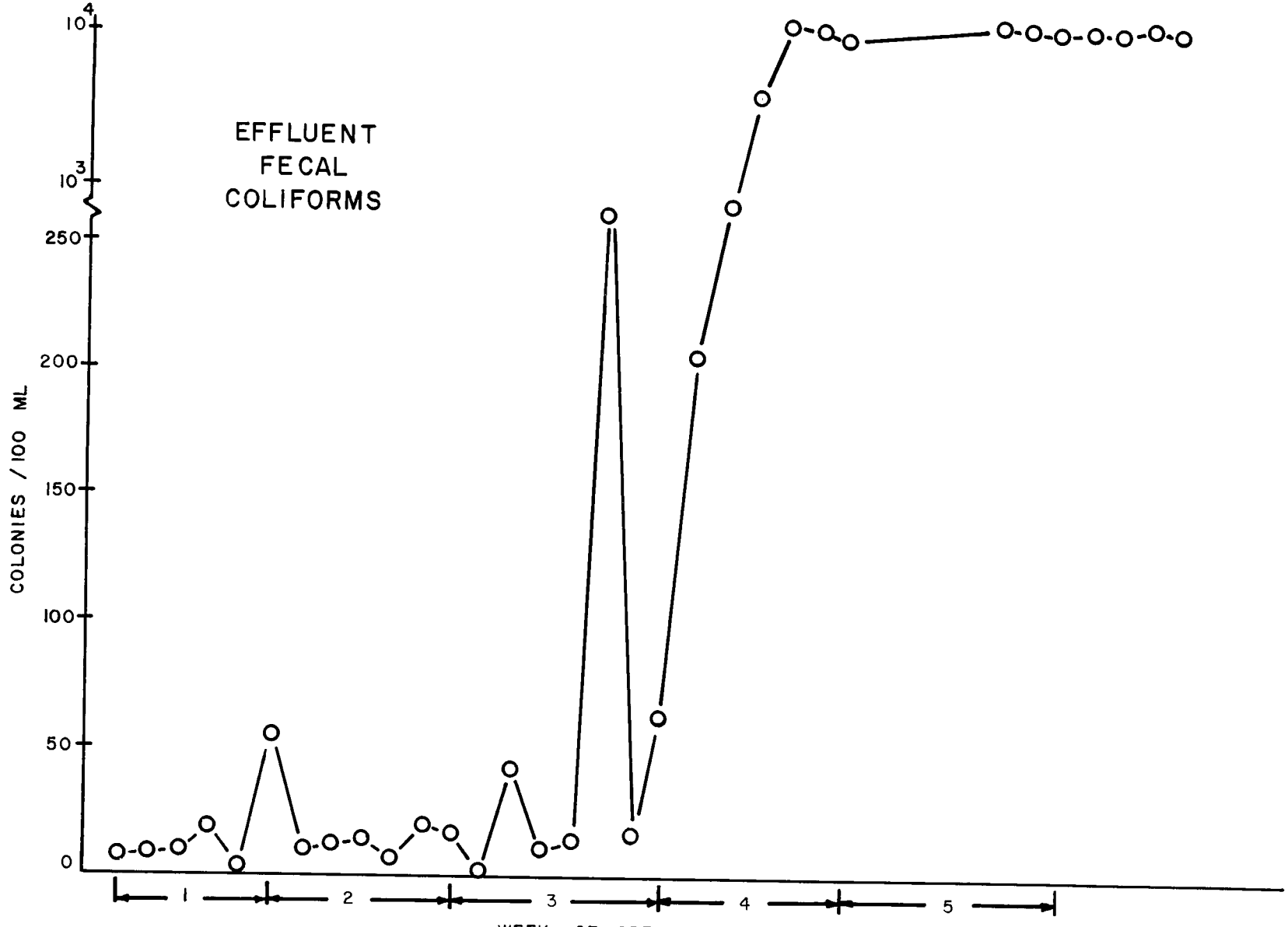

Figure 3. Unit B Results - Run 1

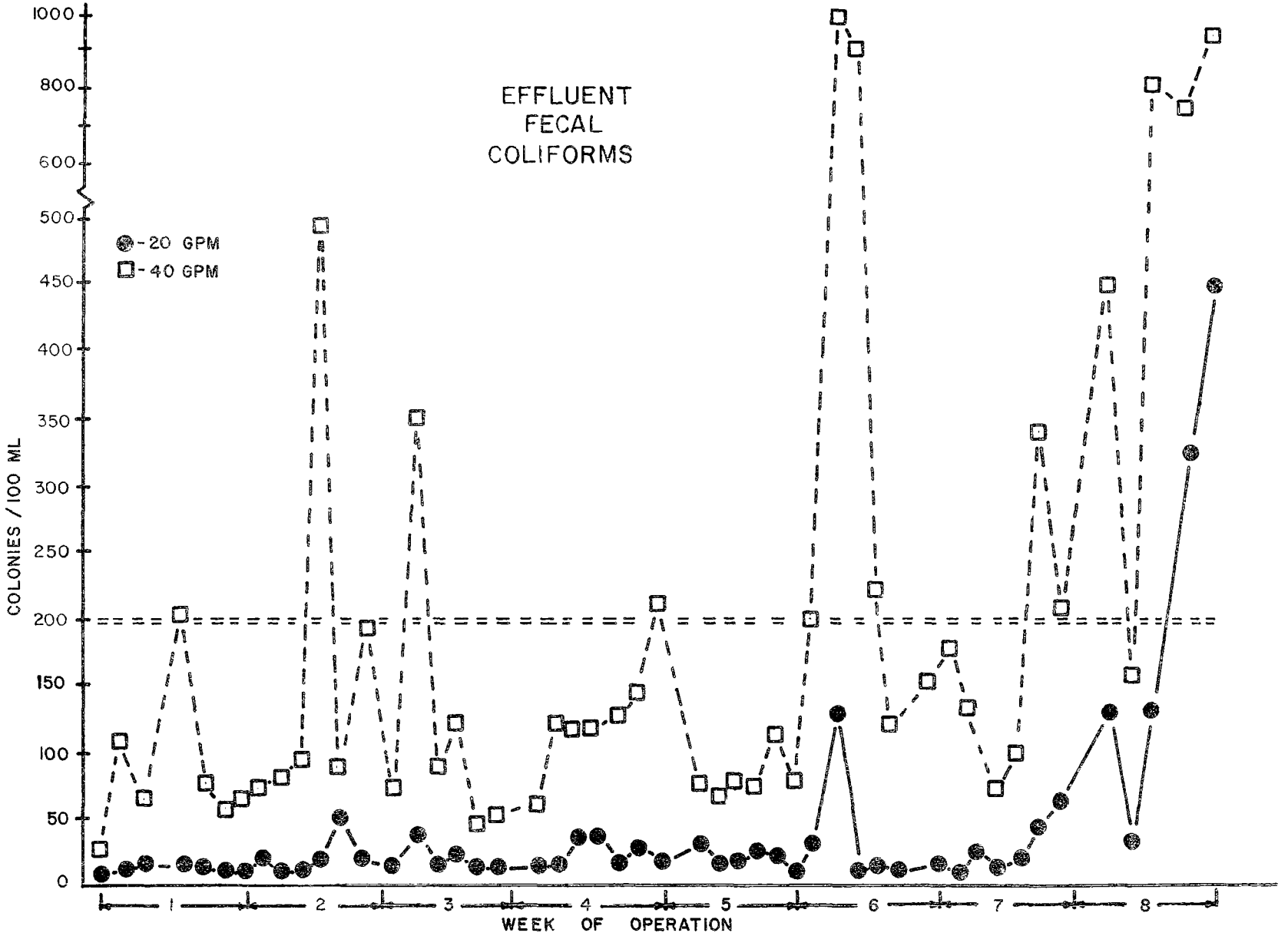

Figure 4. Unit B Results - Run 3

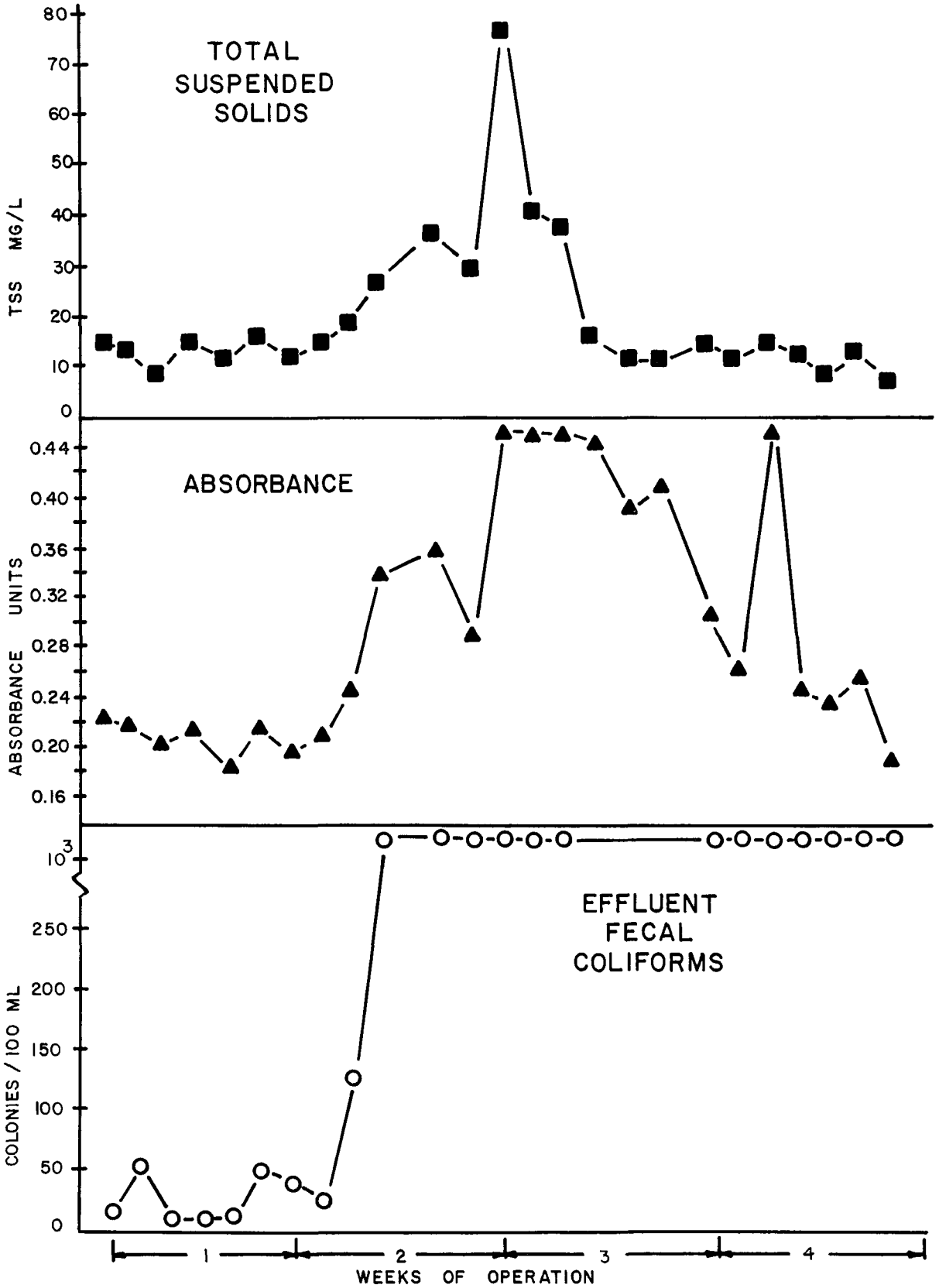

Figure 5. Unit C Results - Run 1

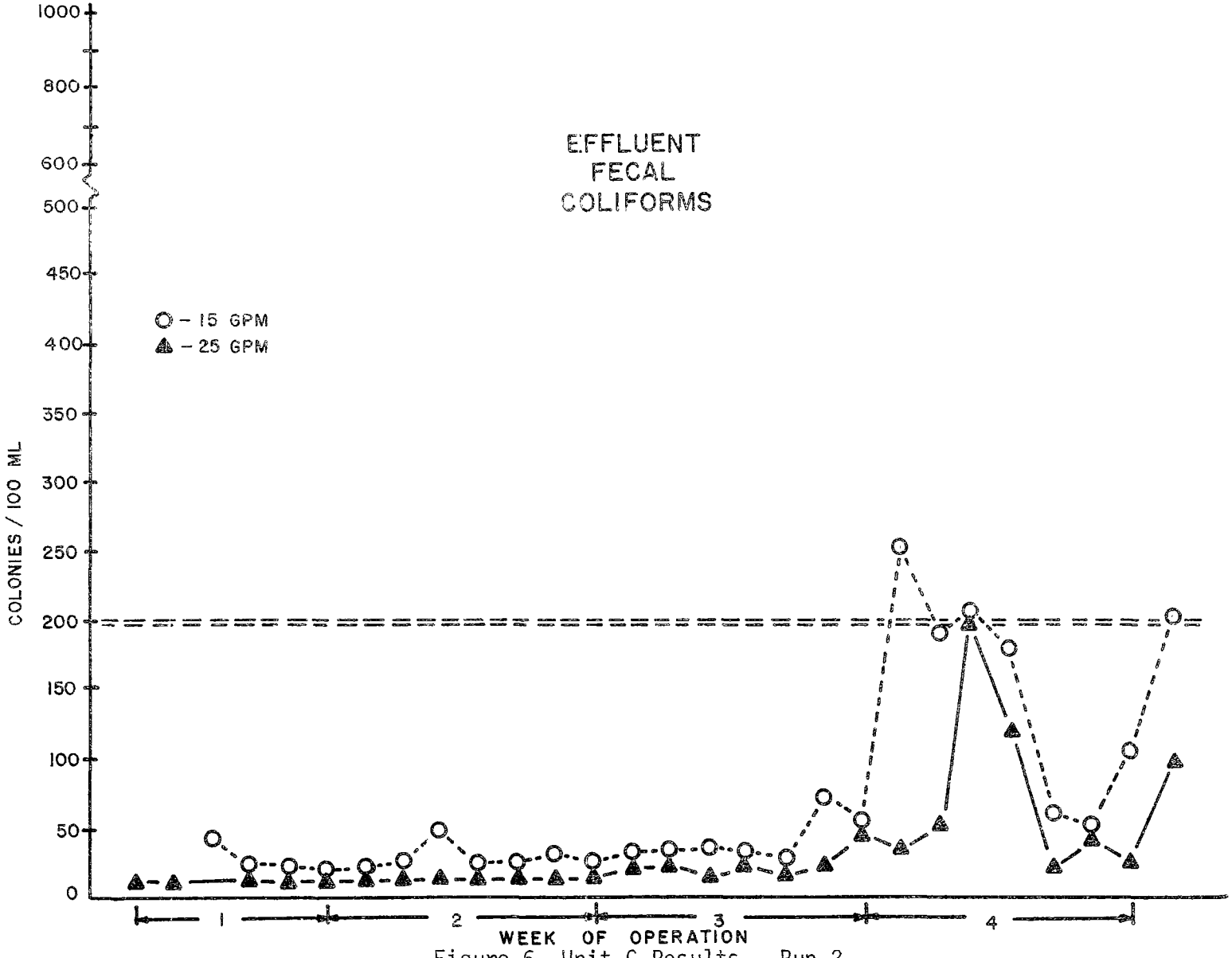

Figure 6. Unit C Results - Run 2

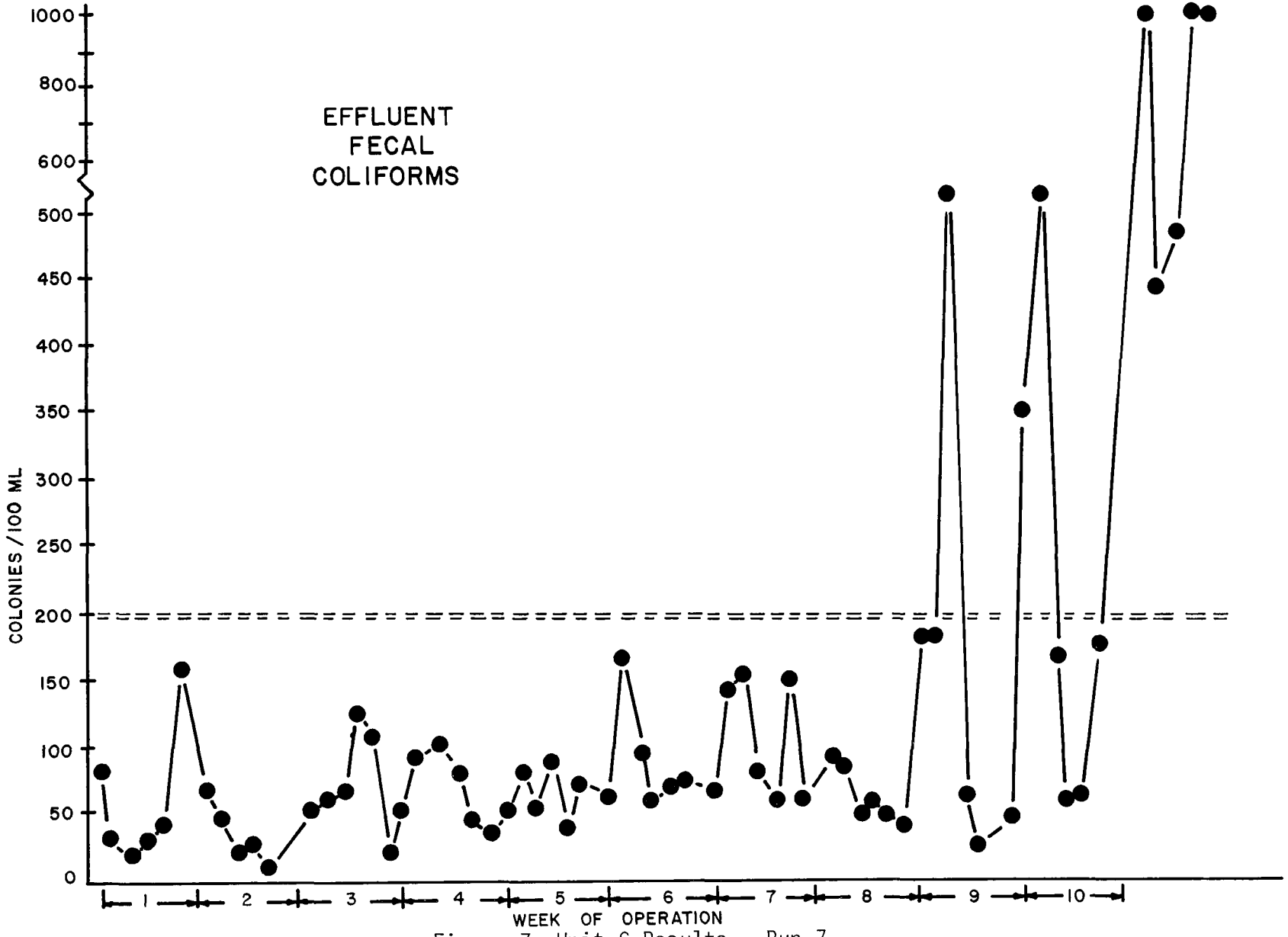

Figure 7. Unit C Results - Run 7

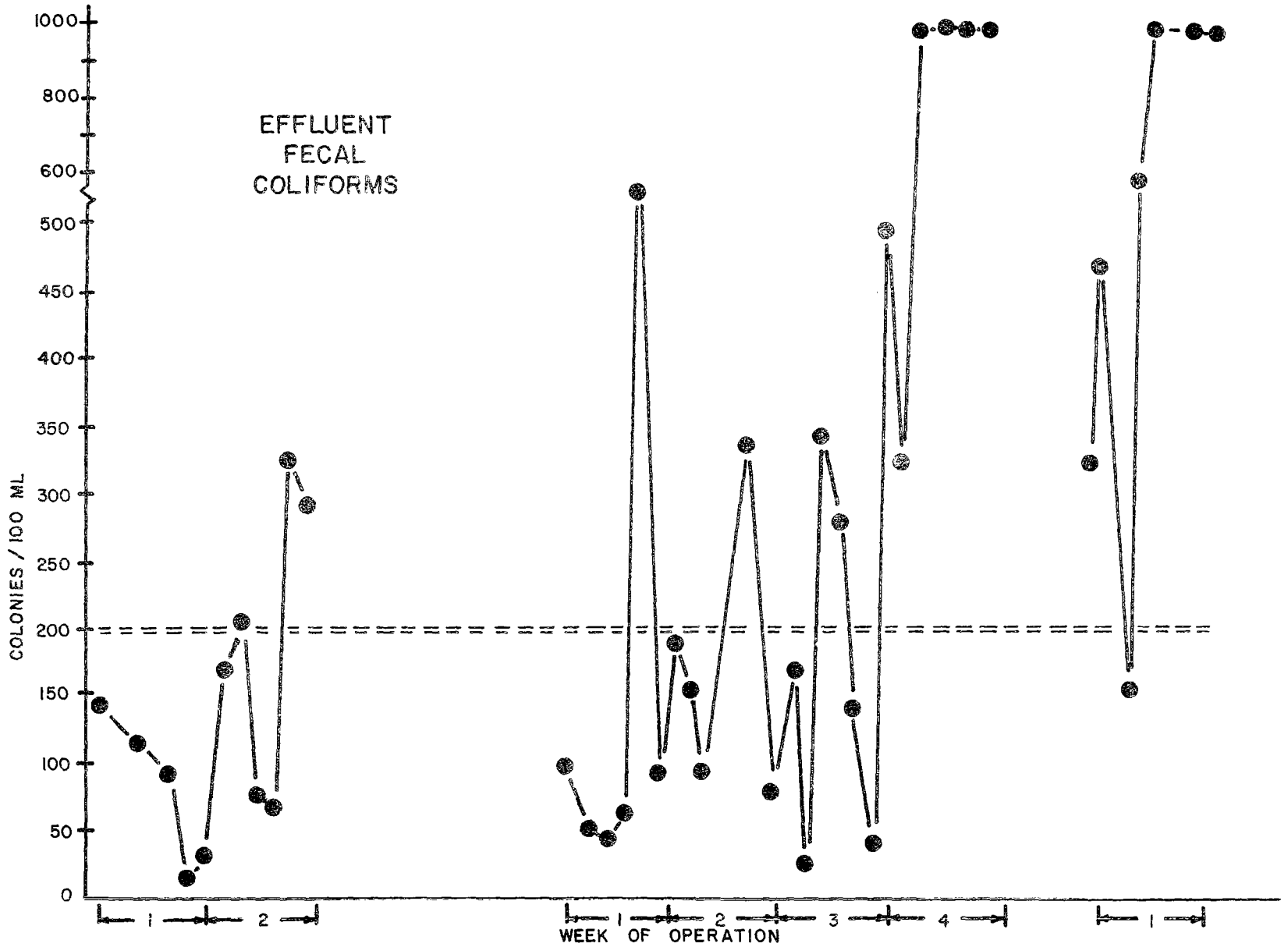

Figure 8. Unit D Results

GEOMETRIC MEAN FECAL COLIFORM RESULTS

RELATIVE TRANSMITTANCE	ABSORBANCE UNITS	
	<0.200	>0.200
<55%	410	552
>55%	35	80

Figure 9. Effect of Relative Transmittance Readings and Wastewater Absorbance Values on Fecal Coliform Results of Unit A

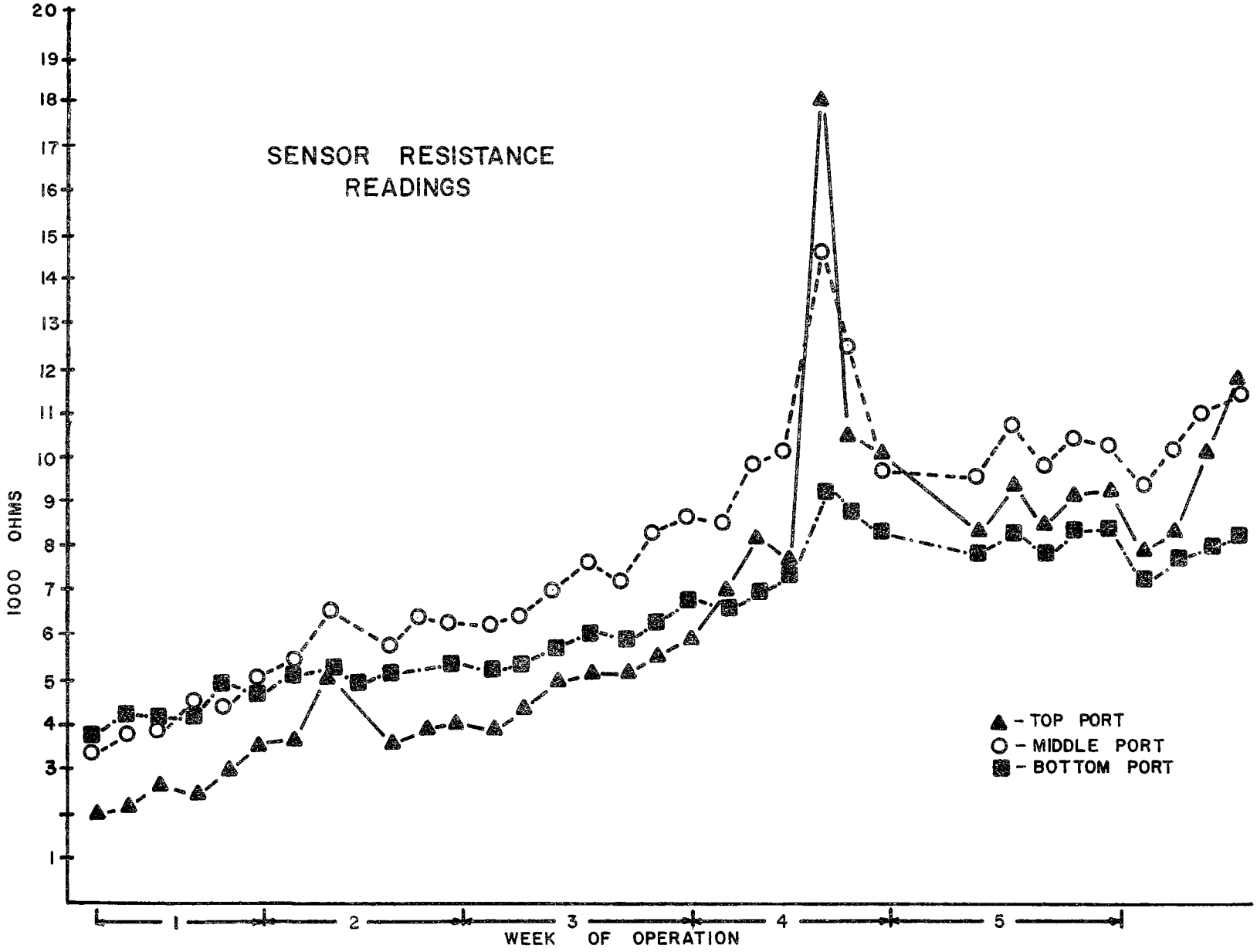

Figure 10. Unit B UV Sensor Readings

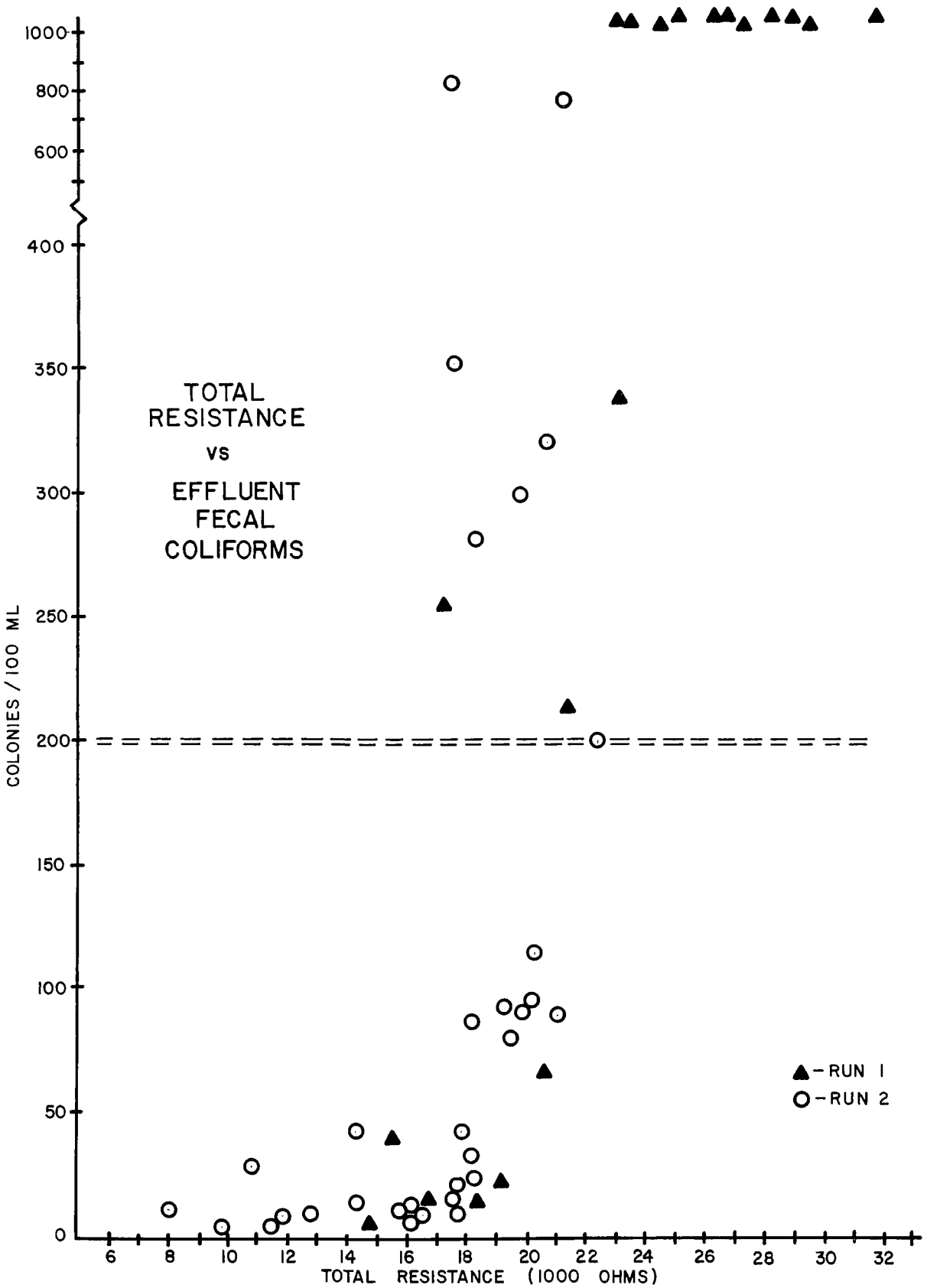

Figure 11. Unit B Sensor Reading vs Effluent Fecal Coliform Counts

5. UV DISINFECTION OF SECONDARY EFFLUENT: DOSE MEASUREMENT AND FILTRATION EFFECTS

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ABSTRACT

The first phase of this study involved an ultraviolet (UV) disinfection pilot plant study comparing: filtration, water quality parameters, and two reactors. The pilot plant study directed us to laboratory experiments involving: (1) the development of a method for *in situ* measurement of dose rate using a calibrated bioassay, (2) experimental verification of a method for calculating dose rates, (3) evaluation of the role of lamp spacing in dose efficiency, and (4) simulation of UV disinfection.

A bioassay method was developed to measure average dose rate (i.e., intensity) within a UV reactor. The survival of spores of *Bacillus subtilis* was determined as a function of UV dose in order to standardize the sensitivity of the spores. Spores were added to unknown systems and the survival could be used to determine the average dose rate. A modification was used for flowthrough reactors, in which spores were injected as a spike and collected at a known time from injection.

Spectrophotometric measurements were found to significantly overestimate the UV absorbance in wastewater because of scattering. A method to correct for scattering was tested. A point-source summation method for calculation of dose rate was verified by bioassay measurements in a simple cylinder. This calculation method was also applied to multiple lamp reactors. A method for simulating survival in complex flowthrough reactors was presented and a simulation of our pilot plant runs corresponded reasonably well with the observed survival. Mixed media filtration significantly improved disinfection in pilot plant media experiments. A laboratory experiment showed that a relatively small number of coliforms were protected inside particles, but they were the factor limiting disinfection at -3 or -4 logs survival.

INTRODUCTION

Environmental problems associated with chlorination have prompted research into alternatives for disinfection of wastewater effluents. Resi-

duals and by-products can be toxic to aquatic life in receiving waters (15) and they may form carcinogenic by-products (8). In addition, chlorination is less effective in killing viruses, spores and cysts than in killing bacteria. One disinfection process which would not be expected to produce undesirable by-products is ultraviolet light (UV).

The Environmental Protection Agency has funded several pilot or full scale investigations of UV disinfection of wastewater (5,9,11,12). While these pilot studies of UV disinfection have generally been successful at meeting disinfection goals, comparison, both within and between these and most other UV studies, has been limited because there has been no direct method of measuring UV doses, nor has there been a substantiated method of calculating doses in the complicated geometries of a practical reactor. In addition, lack of dose measurement methods has prevented the controlled evaluation of effects of variables such as UV absorbance of the water, filtration, reactor design and the varying sensitivity of different organisms.

The first phase of this study was a pilot plant study comparing: (1) the effects of mixed media filtration, (2) the effects of randomly varying water quality parameters, and (3) two UV disinfection reactors employing different lamp spacing. Experience from the pilot plant study directed us to a laboratory experimental second phase involving: (1) development of a method for *in situ* measurement of dose rate (i.e., intensity) using a calibrated bioassay, (2) experimental verification of a method for calculating dose rates or intensities, (3) separation of effects of absorbed and scattered UV light and its relation to spectrophotometer measurement, (4) evaluation of the role of lamp spacing in dose efficiency, and (5) simulation of UV disinfection.

The following are several problems with the dose estimation in previous studies of UV disinfection. (Studies exemplifying these problems are indicated in parentheses.)

1. UV radiometer detectors measure intensity on a planar surface. Thus, they don't correctly measure the 3-dimensional intensity (i.e., dose rate) to which a cell may be exposed near a long tubular lamp (3,9).
2. A UV radiometer detector positioned in the wall of a disinfection reactor can't be used to estimate the average dose rate within the entire reactor (3,2).
3. Wastewater contains particles which scatter UV light so that spectrophotometers tend to overestimate the UV absorbance (9).
4. Equations have been used which incorrectly calculate the dose rate near a tubular lamp in an absorbing solution (9,12,13).
5. In flowthrough systems there is a distribution of exposure times not simply related to volume and flow rate (9,12).

MATERIALS

All measurements of intensity at 254 nm were made with a calibrated International Light 500 radiometer. Measurements of UV output at 254 nm were

made by integrating intensity measurements, made far from the lamp, over a spherical surface centered on the lamp centroid (5). To obtain accurate dose-survival data, suspensions of bacteria were irradiated in a collimated beam apparatus (Fig. 1). To test calculations of UV intensity in a cylindrical geometry, suspensions of spores were irradiated for a fixed time inside the cylindrical apparatus shown in Fig. 2. A moveable paper tube was located between the lamp and the quartz tube so that the lamp could be warmed up and an exact exposure made. Cylinders of different radii were used. Suspensions were well stirred. Fulvic acid was used to vary absorbance.

Bacillus subtilis (ATCC 6633) spores were used for bioassays of UV dose. Preparation of spore stocks is described elsewhere (5). Spores were suspended in buffered water (1) and plated on Thermoacidurans agar. For laboratory experiments total or fecal coliform density was determined by the membrane filter technique (1); however, in pilot plant experiments the MPN procedure (1) was used, and both total and fecal coliforms were carried to the confirmed level. Methods used for water quality parameters are described elsewhere. Spectrophotometric UV absorbance (254 nm) was measured with a Cary 219 spectrophotometer. For some experiments, a special quartz cuvette, ground so as to be translucent on the side nearest the detector, was used to correct for scattering of UV light (14). For pilot plant experiments, two disinfection units were used: an Aquafine CSL-6, and a Pure Water Systems (PWS) I-75. Both filtered and unfiltered secondary effluent were disinfected. Filters were pressurized and contained sand-anthracite media.

RESULTS AND DISCUSSIONS

Bioassay Method for Measurement of Dose Rate

A bioassay method was developed to measure average dose rate in flow-through reactors as well as to verify a method of dose rate calculation. Dose is defined as:

$$\text{Dose} = (\text{dose rate})\ (\text{exposure time}) \tag{1}$$

or, in units:

$$\text{mW-sec/cm}^2 = (\text{mW/cm}^2)(\text{sec}) \tag{2}$$

The term "dose rate" has been used instead of the more familiar "intensity" because of the ambiguities in definitions of intensity. The survival (N_S/N_O) of organisms is usually a function of dose:

$$N_S/N_O = \text{fn(dose)} \tag{3}$$

where N_O and N_S are the density of organisms before and after irradiation, respectively. Equations 1 and 3 imply that dose rate and exposure time may be varied reciprocally to obtain the same survival.

The survival of spores of Bacillus subtilis was determined as a function

of the UV dose in order to "calibrate" the sensitivity of the spores. Since dose rate, as measured by a radiometer, was only applicable in a collimated beam, the spores were exposed for varying periods of time to a collimated beam of UV light in a stirred petri dish (Fig. 1). The dose rate at the surface of the suspension was measured. Since fluid depth and absorbance were minimal, the dose could be calculated based on the measured dose rate and the exposure time. In cases where absorbance was significant, the average dose rate was calculated using an integration of Beer's law over the fluid depth. Calibration curves of log survival vs. dose were constructed (Fig. 3) and found to be quite reproducible over several months. The dose rate may be determined in an unknown system by: (1) determining the survival (N_s/N_o); (2) reading the dose corresponding to the observed survival using the calibration curve (Fig. 3); and (3) using the known exposure time in eq. 1 to calculate average dose rate.

Separation of Effects of UV Absorbance and Scattering

Calculation of average UV dose rate requires an absorbance measurement. Wastewater effluents contain particles which may scatter as well as absorb the UV light. Bioassay experiments showed that scattered UV light was still effective for killing bacteria. Since the usual spectrophotometric measurements do not separate scattering and absorbance, we needed a way to separate the two. An established method using a frosted cuvette for both the blank and sample allowed a correction for most of the scatter (14). A piece of oil saturated paper placed on the cuvette face may also be used.

We tested this technique against a bioassay method to separate absorbance and scattering. A sample of tertiary effluent (14 NTU turbidity) was filtered through a 0.45μ filter. Suspensions of intermediate turbidity were made by mixing portions of the filtered and scattered sample. Thus, the soluble absorbing component was held constant and the particulate component varied. Samples were spiked with Bacillus spores and irradiated in a petri dish in the collimated beam apparatus. The average dose rate in the suspension was assayed. By using the integrated form of Beer's law (7) we determined the absorbance which would yield the observed assayed dose rate. The assayed absorbance for the suspensions of varying particulate content is shown as a function of the spectrophotometric absorbance (Fig. 4). The difference between the spectrophotometric absorbance and the assayed absorbance was the scattering component. The soluble absorbance, particulate absorbance, and scattering were 47 percent, 41 percent, and 12 percent, respectively, of the spectrophotometric absorbance. The frosted cuvette method showed a slightly lower scattering component. The scattering component was estimated to have averaged 9 percent in our pilot plant studies. The soluble absorbance was 60 percent to 80 percent of the spectrophotometric absorbance in most of the secondary effluents measured.

Calculation of Dose Rate

Common radiometer detectors cannot be used to measure dose rate near a tubular lamp because they measure energy flux on the planar surface of the detector. Light received at angles other than 90° to the surface of the

is attenuated since the surface of the detector intercepts a smaller cross section of the rays. The detector "sees" primarily the portion of the lamp directly in front of it. Biological cells in motion in a solution, however, present a 3-dimensional target and they respond to the 3-dimensional dose rate from all angles within a disinfection reactor (6).

To calculate the UV dose rate at a point near a tubular lamp in an absorbing solution, we used an equation which we call the point source summation (PSS) calculation (4,6,10). This equation assumes that a line segment source can be treated as the sum of a number of point sources. We can consider a cylindrical coordinate system around a line segment light source surrounded by a quartz sleeve (Fig. 5). The total line source of UV output OPT is divided into N point sources each of which has strength S (units in Watts).

$$A = OPT/N \tag{4}$$

The dose rate at a point $I_{(R,Z_C)}$ due to one point source (Z_L) can then be treated as the product of the spherical spreading times the attenuation due to absorbance over a definite path length ($P-P_1$).

$$I_{(Z_L),\ (R,Z_c)} = [S/4\pi(R^2 + Z_{LC}^2)]\exp[-a(R-R_1)P/R] \tag{5}$$

where a is the absorbance of the medium and the other geometry is shown in Figure 5. The total dose rate at point $I_{(R,Z_C)}$ is the sum of the contributions of each point source (at each Z_L) over the source length (Z_{LN}).

$$I_{(R,Z_c)} = \sum_{Z_{Ln}}^{Z_{Lo}} I_{(Z_L),(R,Z_c)} \tag{6}$$

The use of this calculation requires two measurements: absorbance of the water, and the lamp UV output (5).

To test the PSS calculation, we compared the calculated average dose rate inside a cylinder (Fig. 2) to that measured by the spore bioassay. We used the PSS calculation in a computer program to average the dose rates over the volume of a cylinder around a lamp. We did this for a series of cylinders of varying radii and for fluids of different absorbances. The survival of the spores was measured and the assayed average dose rate determined as outlined previously.

The PSS calculations were generally verified by the bioassay measurements. Figure 6 shows a comparison between the calculated PSS curves (solid lines) and the bioassay data (data points). The correspondence was good both for cylinders of different radii and for fluids of varying absorbances. The stirring device may have produced some shadowing loss in the 2.5 cm cylinder.

We also performed the same experiment using spores spiked in a secondary effluent, and PSS calculations were within 10 percent of the bioassay dose rates. We also applied the calculation methods which had been used in some previous studies (9,12) to these cylinders and those methods gave results which differed greatly from our experimental average dose rates (5).

Practical UV reactors are flowthrough systems and have a distribution of exposure times. To use the bioassay of dose rate in a flowthrough system we needed a way to determine a definite exposure time. To do this we used the spores in a manner analogous to a tracer injection study. To demonstrate this method we used a flowthrough tube surrounding a UV lamp. Spores were injected into the flowstream of water at the entrance to the tube and the outflow fractions were collected in a rotating sampling tray as a function of time from injection. The injection was performed with the light on and repeated with the light off. The density of the unirradiated spores (N_o) is shown in Fig. 7. The distribution of unirradiated spores reflects the retention time distribution (RTD). The density of surviving irradiated spores (N_s) is shown in Fig. 7. The survival (N_s/N_o) was calculated for each flow fraction separately by comparing spore densities in the corresponding irradiated and unirradiated fractions at a given time from injection. The average dose rate was then determined for each fraction by finding the corresponding dose from the calibration curve and dividing by the time from injection. The assayed dose for each flow fraction is also plotted in Fig. 7. The slope of the regression line of the assayed dose vs. time from injection was equal to the average of the assayed dose rates in the separate fraction. A modification of the spore injection bioassay may be used to measure average dose rate in full scale reactors.

The assayed average dose rates within the flowthrough tubes (Fig. 6, "injection expts." data points) corresponded well with the calculations of the PSS model (Fig. 6,lines). The distribution of unirradiated and irradiated viable spores in Fig. 7 also showed that nearly all of the surviving spores emerged from the tube before the average retention time. This illustrates the important effect that flow dispersion can have on the disinfection efficiency.

Calculation of Dose Rate in Multiple Lamp Reactors

To calculate average dose rate in multiple lamp reactors we used the following method: (1) dose rate at each point was considered to be the sum of the contributions from each lamp calculated by the PSS model; (2) dose rate was mapped at each point on a grid of the cross-sections of the reactor; and (3)dose rates were averaged over the cross-sections and along the length of the reactor.

We found that UV lamps transmit little of the UV light coming from adjacent tubes (5) or absorb nearly all UV output striking them from neighboring lamps. Thus, it was necessary to make calculations which took this shadowing into account. Our calculations also made these simplifications: that reflections from the reactor walls was negligible under actual operating conditions, and that reflection and refraction by the quartz sleeves were

negligible.

There are divergent views on the design of UV reactors. Some of these views are based on improper equations or conventional wisdom rather than calculation or experimental measurement. This is because of the lack of adequate and comparable methods for measuring or calculating UV dose (e.g., 13). Our models can be useful for research and development of reactor design. We applied our calculations to contrast the efficiency of the different schemes of lamp spacing in absorbing fluids. Any surface or object which absorbs UV energy (e.g., walls, baffles, other lamps), in addition to the unavoidable absorbance of the water itself, reduces its efficient use. The product of dose rate times reactor volume is a factor which is directly proportional to the effectiveness of the unit at treating fluid volumes of water at a given flow rate and flow conditions. This factor isolates the effectiveness of the dose rate regime or intensity distribution from the effects of flow dispersion or hydraulic characteristics and can be used to compare reactors of different lamp spacings and volume. At a given flow rate and number of lamps, a close lamp spacing gives a higher average dose rate or intensity but at the sacrifice of shorter detention time because of the smaller volume of the unit. We showed with calculations how the distance the light was allowed to penetrate, before being lost on a neighboring lamp or wall, affects the efficiency of light use. Figure 8 shows the dose rate-volume product in cylinders of radius R or fluid depth around a UV lamp. The point at which the lines level out is the radius at which most UV light has been absorbed and no further improvement in efficiency occurs. In other words, the decrease in intensity just balances the increase in detention time as the radius or fluid depth increases. For an absorbance representative of secondary effluent, 0.16, it can be seen that walls or other obstructions within 5 cm can absorb a significant amount of available UV light. Fluid depths less than 5 cm are less efficient at this absorbance. Two reactors used in the pilot plant experiments were compared on the basis of their dose rate-volume products (Table 1). The reactor with lamps placed close to one another and the walls (PWS unit) had an average dose rate or intensity almost twice as high as the other reactor (Aquafine). However, the PWS reactor had a much smaller volume (and shorter retention time) so the dose rate-volume products were almost equal. However, the PWS reactor used a greater lamp wattage. We used a term we called the dose rate-volume "efficiency" (dose rate-volume product/input wattage) to compare the efficiency of the use of the lamp wattage. The PWS was much less efficient because of the proximity of the lamps to the walls and the wall and neighboring lamp absorption of the light.

The dose rate-volume product does not consider the effects of non-ideal flow. Although the dose rate-volume products of the two reactors were nearly equal, the PWS reactor gave from 0.6 to 2.1 log units greater survival of fecal coliforms than the Aquafine at the same flow rate because the less ideal hydraulic characteristics of the PWS unit gave severe short-circuiting of flow in the PWS reactor. Thus, the effects of flow dispersion must be considered as well as the dose-rate or intensity regime in determining the ultimate disinfection efficiency or total dose produced by a given lamp wattage into a given volume of fluid.

We also used simulation of a full-scale reactor, operated in NW Bergen County, N.J. (12) to show the effect of varying lamp spacing on the UV light use efficiency and an analysis of the relative costs (5).

Simulation of Dose and Disinfection in Flowthrough Reactors

The second factor in calculation of dose, exposure time, can lead to as much error in calculations as dose rate or intensity. In flowthrough reactors there will be a distribution of retention time. Figure 7 shows clearly neither the retention time (RT) calculated from flow rate and volume nor even the average RT determined from dye studies can be used to predict the average survival. Since survival is not linearly related to dose, the average dose is insufficient to predict the average survival over the RT distribution,but the survivor density must be calculated for each flow fraction and then summed.

The following equations will show how the density of survivors (N_s) may be predicted from the following data: (1) coliform density in inflow (N_o), (2) average dose rate (DR), either measured or calculated, (3) retention time distribution, and (4) dose-survival curve (determined accurately, e.g., in collimated beam apparatus).

For an aliquot of volume V_t entering the reactor at time t_o, the aliquot will exit in n fractions of volumes V_i at times t_i. Survival in each fraction is some non-linear function (fn) of dose.

$$N_s/N_o = \text{fn(dose)} \tag{7}$$

$$\text{Dose for the ith fraction} = (DR)(t_i) \tag{8}$$

$$\text{Survival in the ith fraction} = N_{s_i}/N_{o_i} = \text{fn}[(DR)(t_i)] \tag{9}$$

$$\text{Average density of survivors, } \bar{N}_s = N_o \sum V_i(\text{fn}[(DR)(t_i)])/V_t \tag{10}$$

Data from a dye study on the RTD may be put in a form to use in these equations. The area under a curve of dye concentration vs. time to set equal to V_t (and may be thought of as a 1 ml aliquot entering the reactor). Then,

$$V_i = (\Delta t)(\text{relative dye concentration})/V_t \tag{11}$$

For a computer simulation of average survival, the RT distribution, and dose-survival curve data pairs were fed into arrays and intermediate values needed in eq. 10 were generated by linear interpolation.

As an example of simulation of survival in a flowthrough reactor, we simulated runs with the Aquafine reactor. These simulations we then compared to the observed survival in the pilot plant experiments. The average dose rates calculated by the PSS model for two levels of applied voltage and the input data in equation 10 were used. The RT distribution was measured with dye injection and adjusted to the correct flow rate. We lacked the methods at the time of the pilot plant runs to determine an accurate coliform dose-survival curve, so one was determined some time later for a sample from the same site.

The average log survival predicted by the simulation corresponded reasonably well with that observed in the pilot plant runs (Table 2). Some deviation might be expected since the dose-survival curve was based on one sample taken at a later date. Further research should involve simulation using data obtained simultaneously with full reactor runs.

Simulation takes into acount the factors of the dose rate-volume characteristics as well as the effects of flow dispersion and sensitivity of the target organisms. It can be useful tool for research and development of reactor design. For example, it can be used to find optimum lamp configurations and tradeoffs with flow dispersion. It can be used to predict the design parameters needed for a specific situation so that costly overdesign is not necessary. The predicted survival of a standard coliform sample at a given flow rate may be used to compare a number of different reactors. The simulations may also be used to prepare empirical curves of predicted survival vs. flow rate, operating voltage, water quality, etc., for a particular installation as a guide to continuous operation.

Protection of Cells Inside Particles and Effects of Filtration

In our pilot plant experiments, an extended aeration secondary effluent was subjected to mixed media filtration. Both filtered and unfiltered effluents were subjected to UV disinfection in two UV reactors, at two different flow rates and two levels of applied lamp voltage. The filtered effluents showed significantly better disinfection (Table 3). Total coliform log survival was 0.33 to 0.79 log units lower in the filtered treatments. The effect of filtration on UV absorbance was small and did not account for the disinfection differences. The differences in suspended solids, turbidity, and UV absorbances indicate that filtration tended to remove the larger particles which had relatively little effect on the absorbance. Average dose rate calculations and simulation supported the idea that the filtration effect was not due to the lower absorbance after filtration. We concluded that a relatively small number of coliforms were protected inside particles but that these tended to be removed by filtration.

We performed a laboratory experiment to support the hypothesis that particle protection is the major effect increasing disinfection after filtra-

tion. We determined the dose-survival curves of an unfiltered effluent sample and the same sample passed through a 70μ and 8μ pore size filter. Since coliforms are about 1-2μ in size, the 8μ filter allowed only single cells or very small aggregates to pass. The survival curve of this fraction (Fig. 9) shows disinfection continuing beyond -4.5 log units survival where survivors were undetectable. Curves for the 70μ filtered and unfiltered samples tend to level out after -2 or -3 log units survival. The coliforms not passing the 8μ filter were extremely resistant to UV. Since the curves were similar until less than about 10, or 1 percent, of the coliforms were surviving, the protected coliforms appeared to be a small minority but became the limiting factor to disinfection at levels needed to meet legal standards.

Other Pilot Plant Results

The Aquafine reactor met the disinfection goal of 200 MPN/100 ml in every case. However, the PWS reactor did not because of short circuiting of flow. Changes in applied lamp voltage and flow rate produced relatively small changes in survival because, as can be seen from the dose-survival curve in Fig. 9 for example, the dose-survival curves level out at -3 or -4 log units survival. Stepwise multiple regression of randomly varying water quality parameters on log survival of coliforms showed no consistant correlations. This lack of correction was probably due to the relatively small variation in UV absorbance and the lack of response of kill to dose increases at -3 or -4 log units survival. The significant correlations were spectrophotometric absorbance was predicted well by coliform densities, or if these were not considered, by COD, turbidity and suspended solids together.

CONCLUSIONS

If the disinfection of single coliform cells in wastewater under ideal flow conditions is considered as "ideal efficiency" then the results of this report show the following to be the chief factors limiting ideal efficiency in practice: (1) protection of cells inside particles, (2) flow dispersion and poor mixing across dose rate gradients, and (3) shadowing and absorption of UV light by walls within a reactor.

ACKNOWLEDGEMENTS

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Table 1. Comparison of Aquafine and Pure Water System units.

CHARACTERISTIC	Aquafine	PWS
Input wattage	240	350
UV output, total W	54.6	68.2
Calculated average dose rate[1] (mW/cm^2)	8.5	16.2
Dose rate-volume product (mW/cm^2)(l)	93.5	94.2
Dose rate-volume "efficiency" [(mW/cm^2)(l)/input wattage]	0.390	0.269

[1] at absorbance = 0.17

Table 2. Actual vs. simulated survival (S) of total coliforms in a Sandy Creek secondary effluent.

Lamp voltage	Ave. intensity (mW/cm^2)	Simulated log S	Pilot plant log S
60	5.1	- 3.00	- 3.29 ($\pm$ .13)
128	8.5	- 3.61	- 3.69 ($\pm$ .16)

Table 3. Inactivation shown as mean -log survival of fecal coliforms in unfiltered and filtered secondary effluent, broken down by filtration status, applied voltage and flow rate. Standard deviations (of log units) shown in parentheses.

		Aquafine	
		Unfiltered	Filtered
Flow rate (l/s)	Voltage	Fecal coliforms (-log survival)	
4.92	60	3.08 (.20)	3.88 (.19)
	128	3.41 (.23)	4.17 (.18)
2.27	60	3.91 (.23)	4.29 (.17)
	128	3.47 (.28)	3.92 (.24)

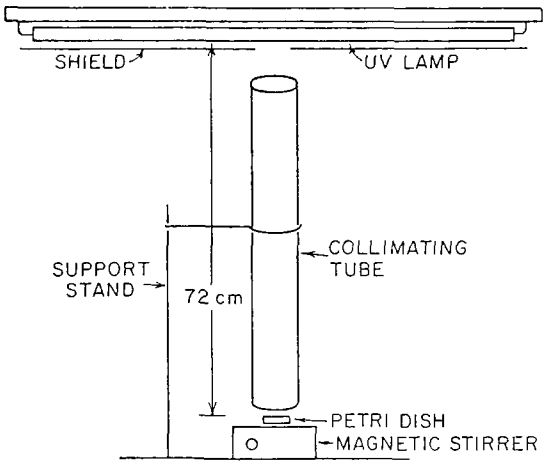

Fig. 1. Collimated beam apparatus

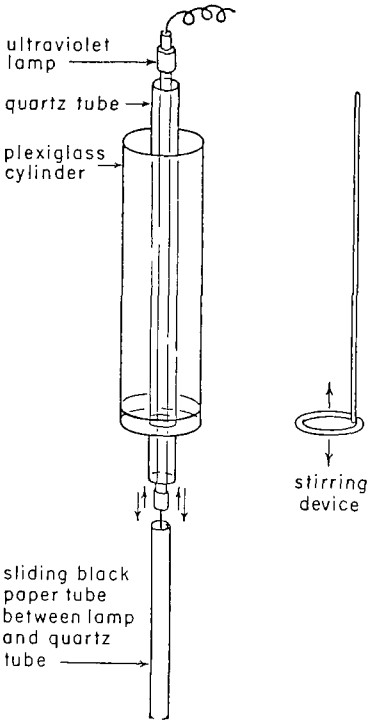

Fig. 2. Cylindrical batch irradiation apparatus

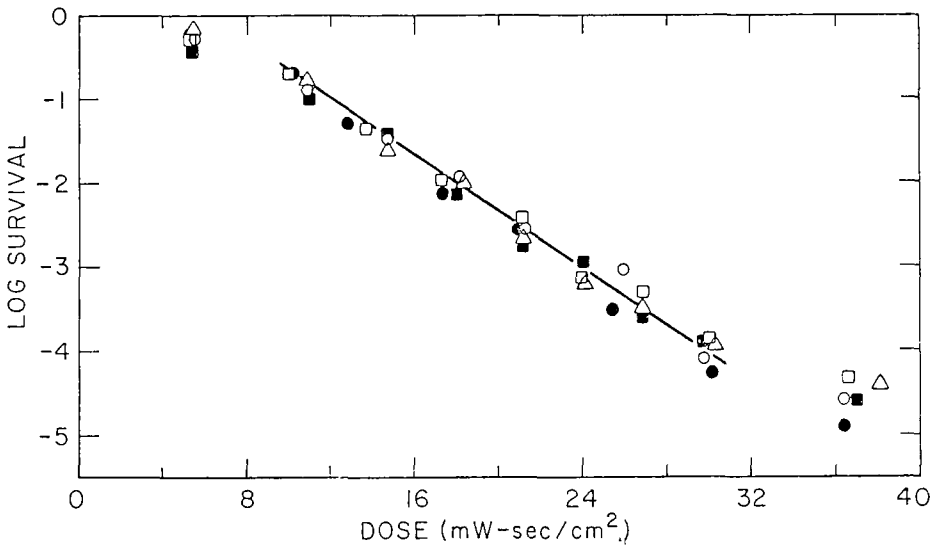

Fig. 3. Log survival of Bacillus subtilis vs. UV dose in a collimated beam of known dose rate. Different symbols represent 5 different runs. Data from doses of 10-30.5 mW/cm^2 appeared linear and fit the regression line Y = .167x + 1.01 (r = .98).

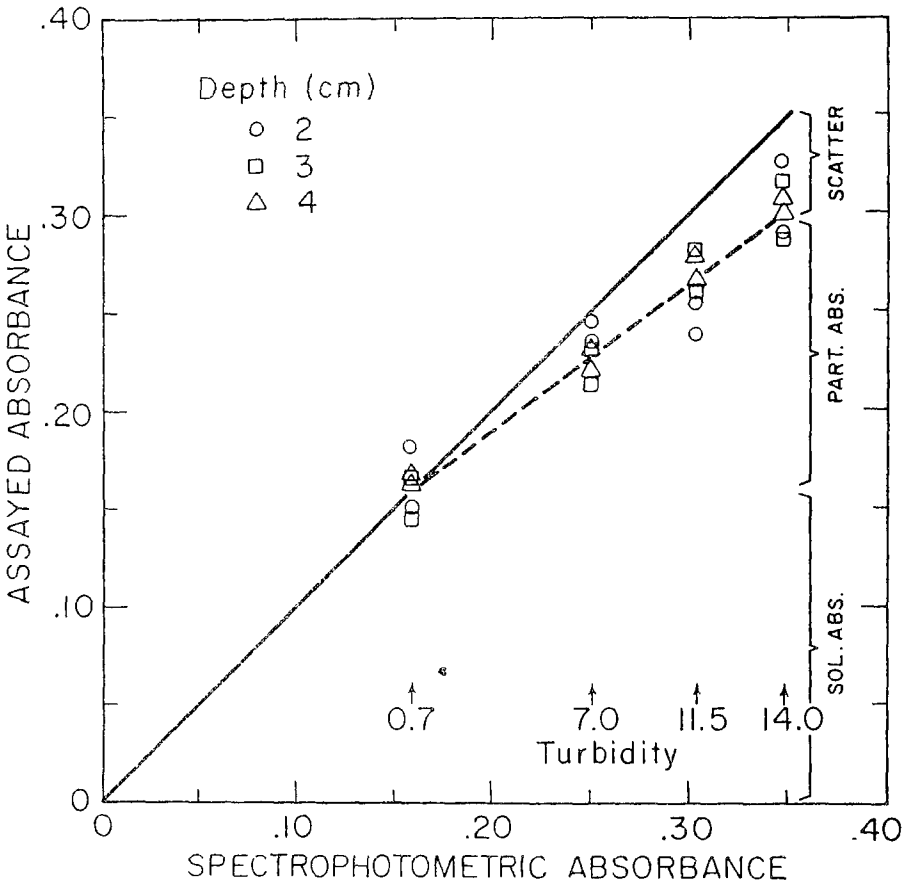

Fig. 4. Spectrophotometric absorbance vs. absorbance measured by the bioassay method for a Chapel Hill tertiary effluent sample. The soluble UV absorbance was kept constant and the particulate concentration varied by diluting the unfiltered (14 NTU) sample with filtered (.07 NTU) sample. The solid line represents an exact correspondence between the two methods. The dotted line is a regression through the data points. The soluble and particulate absorbance and scatter components of the spectrophotometric absorbance of the unfiltered sample are indicated.

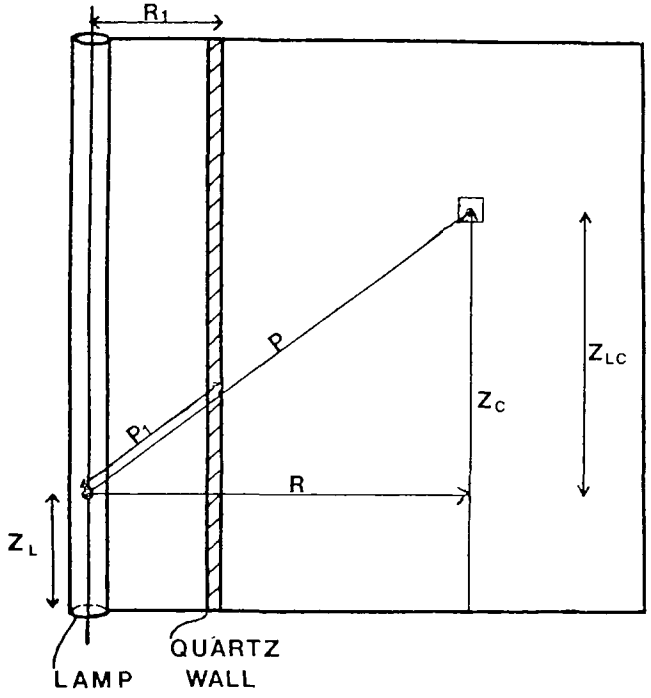

Fig. 5. Cylindrical reactor geometry for point source summation calculation. (Modified from Jacob and Dranoff [4]).

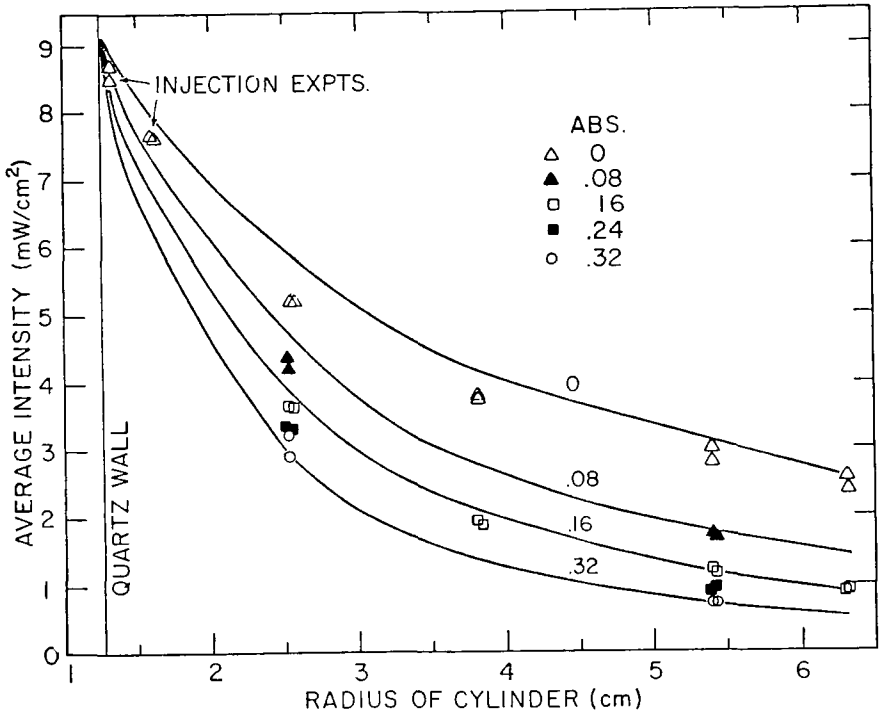

Fig. 6. Average dose within a cylinder of radius R. The solid lines were calculated by point source summation for several different absorbances. Data points represent bioassayed average dose rate within the cylinders of various sizes. Data points for 1.32 and 1.59 cm radius were obtained from flowthrough tubes rather than batch.

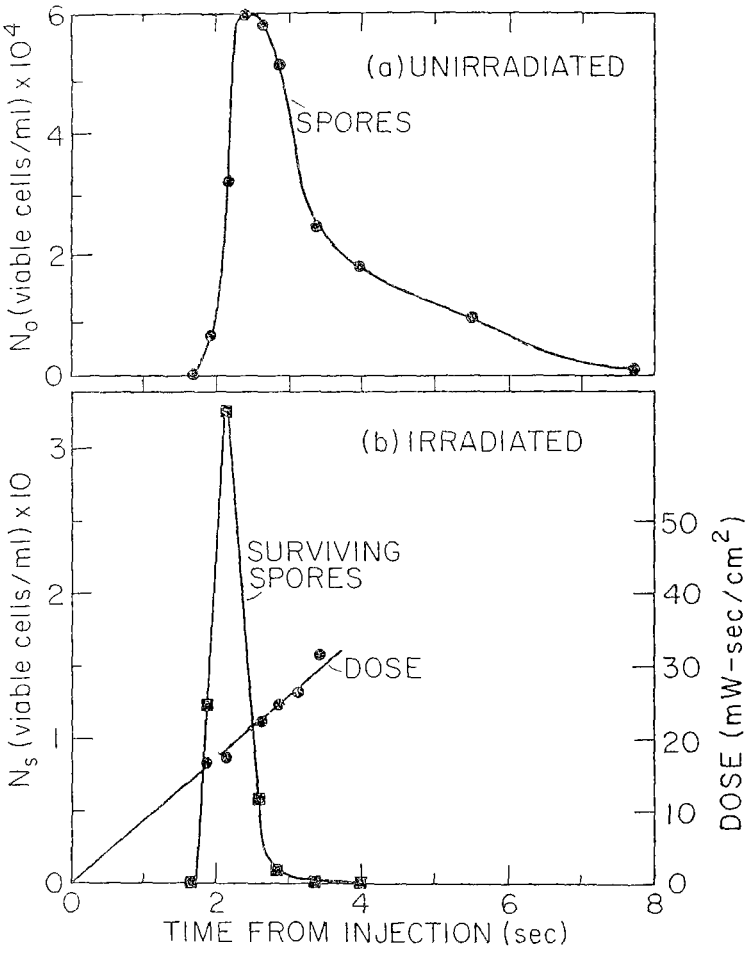

Fig. 7. Assay of average dose rate in 1.32 radius flowthrough tube by injection of spores and collection of separate fractions over time after injection. Fig. A shows the concentration of spores vs. retention time (time after injection) with no irradiation. Fig. B. shows the spore concentrations as a function of retention time when irradiated at the same flow rate. Also shown is the assayed dose calculated from the N_o,N_s of each fraction collected and the calibration curve. For clarity, the viable spore distribution curves are shown for only one experiment but the assayed dose rate for each point is, (the assayed dose)/(retention time), and the average corresponds to the slope of the regression line through all the points forced through the origin.

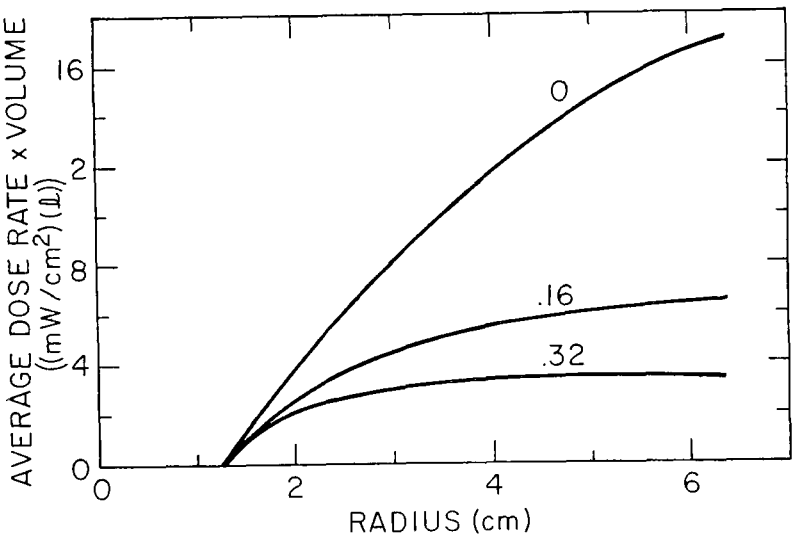

Fig. 8. Effectiveness of various fluid depths in cylinders of radius R around UV lamps. Calculated values of the product of average dose rate in a cylinder, of radius R, times the volume of that cylinder are shown vs. the other radius of the cylinder for fluids of absorbances 0, .16 and .32.

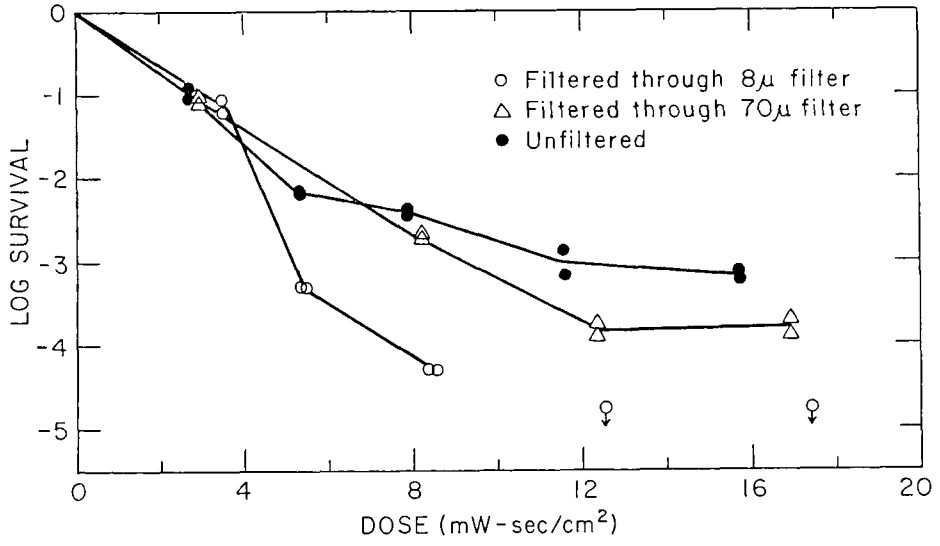

Fig. 9. Effect of filtration on survival of total coliforms in Sandy Creek with arrows indicating limit of detectibility for exposure in which no survivors were found.

6. PILOT INVESTIGATION OF ULTRAVIOLET WASTEWATER DISINFECTION AT THE NEW YORK CITY PORT RICHMOND PLANT

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ABSTRACT

A major EPA-NYC funded project investigating ultraviolet disinfection of secondary effluent and of CSO wastewaters has been started and is entering the experimental phase. This paper presents a progress report on the study.

The pilot plant is operational with two 100 lamp submerged bulb systems in place. A third unit, a non-contact teflon tube system will be in place by the Spring of 1982. Each unit will receive wastewater flows between 0.95 and 4.5 ML/d (0.25 and 1.2 mgd) under a controlled experimental program. The field evaluation was started in December 1981 and will continue for a period of 15 to 18 months. The major efforts which are currently underway are the development of a generalized mathematical model, a detailed characterization of the hydraulics through each system, and a direct comparison of the two submerged systems, which differ only in the spacing of the lamps.

INTRODUCTION

A large scale pilot investigation of wastewater disinfection by ultraviolet light irradiation (UV) is being conducted at New York City's Port Richmond Water Pollution Control Plant, Staten Island, New York. The project is jointly funded by the United States Environmental Protection Agency (Municipal Environmental Research Laboratory, Cincinnati, Ohio) and

the City of New York Department of Environmental Protection. HydroQual, Inc. is the Principal Investigator for the program.

This is a progress report. The experimental program has been underway for approximately three months of the anticipated 18 month schedule, and elements still remain in the construction of the pilot facility. Thus, little can be presented in the way of actual field data or conclusions regarding the operation of the facility. Rather, this presentation will center on a description of the facilities, the scope of work and a discussion of tasks which have been completed to date. Where appropriate, field data will be presented, although the reader is cautioned that these are preliminary and cannot be rigorously interpreted.

SCOPE OF WORK

The major objective of this project is to establish and demonstrate a rationally based protocol for the design of ultraviolet disinfection systems. This is an outgrowth of the conclusions and recommendations reached in the recently completed study at the Northwest Bergen County WPCP, Waldwick, NJ(3). Much of the investigative work in UV disinfection to date has been empirically based, making it difficult to compare systems or to test the sensitivity of a design to various operating variables. The Port Richmond project will involve the development of a rational design protocol and will then demonstrate the validity and application of the method by collection of actual field performance data. Three systems will be tested, each differing in their basic design configuration. The design method considers water quality, system hydraulics, and system geometry (lamp spacing), all of which will be study elements of the experimental program.

Other objectives of the field program will involve the evaluation of operation and maintenance requirements, photoreactivation, the impact of wastewater variability, and the development of capital and O&M costs. The experimental phase is expected to end in the Spring of 1983, with a formal report to be issued by late Summer, 1983.

PILOT FACILITIES

The Pilot Facility is located at the Port Richmond Water Pollution Control Plant (WPCP), one of New York City's twelve operating wastewater treatment plants. Port Richmond, on the northern shore of Staten Island, receives residential and industrial wastewater from a 62 square km drainage area. The WPCP is a step aeration activated sludge facility which was upgraded during the 1970s to provide secondary treatment. It is designed to treat an average flow of 227 million liters per day, ML/d, (60 million gallons per day, mgd) in the secondary system and a maximum of 454 ML/d (120 mgd) through the primary system. Present flows average 151 ML/d (40 mgd) during dry weather and up to 378 ML/d (100 mgd) during storm events. Flow in excess of the secondary design flow is bypassed directly from the primary tanks to the final effluent channel.

A schematic of the Port Richmond WPCP is shown in Figure 1. The UV test facility is located north of the building containing the secondary aeration tanks. A layout of the UV pilot plant is shown in Figure 2. Secondary plant effluent is pumped from the effluent channel via an existing spray water pump located in the sludge pump gallery. Primary plant effluent, which will simulate the quality of settled combined sewer overflow (CSO) wastewater, is pumped directly from the bypass channel during storm events. Both types of flows are pumped into a constant head tank just outside the temporary building (6.1 m x 7.6 m) housing the UV systems. From the head tank, the effluent flows by gravity through the UV units and is discharged to the bypass channel joining regular plant effluent prior to the outfall. Each UV system can receive a flow between 0.76 ML/d (0.2 mgd) and 4.5 ML/d (1.2 mgd). Palmer-Bowlus flumes have been inserted into the effluent channels, which, in conjunction with ISCO Model 1700 meters, are used to monitor the flow of each system.

A description of the UV units is summarized in Table 1. There are two UV systems inside the temporary building as shown in Figure 3. Each has an influent and effluent tank attached to the lamp units. Overall dimensions of each are 1.07 m wide by 2.74 m long and 3.05 m long, respectively. The only difference between the units is the lamp spacing: 1.25 cm and 5 cm (defined as the closest distance between the surfaces of two quartz sleeves). The lamp battery dimensions (internal) are 0.74 m long by 0.69 m high and 0.73 m wide for the widely spaced unit and 0.40 m long by 0.35 m high and 0.73 m wide for the narrowly spaced unit. Each system contains 100 lamps in a symmetrical (10 x 10) array perpendicular to flow. The lamps are Voltarc 40 Watt (nominal) G36T6VH units. Each is enclosed in 23 mm diameter quartz sleeves. The rated output at 253.7 nm for each lamp is approximately 14 W.

The lamps are cleaned by a mechanical wiper system. The wiper blade is cable driven at a variable stroke rate by a pneumatic cylinder. Each UV system has a separate power panel containing shutoff switches for each of three banks of lamps (divided into 30, 40, 30 lamps) and pilot indicators for each lamp. UV intensity monitors for each bank of lamps, elapsed time of operation totalizers, lamp ballasts and the wiper timing devices are also mounted in the power panel. The remaining equipment within the building includes an air compressor for the wiper mechanism, the variac control for modifying lamp intensity, flow meters and additional power distribution and lighting panels.

A third UV system is proposed for installation outside the temporary building. The unit will connect to the same influent and effluent lines as are currently used. It differs from the other two in that the wastewater flows through teflon tubes. The ultraviolet bulbs are parallel to the flow and are not immersed in the wastewater. The size of this unit is 4.9 m long by 0.91 m high by 1.52 m wide.

Construction of the UV facility was started in May, 1981. By October the facility was complete, including electrical work and installation of the two UV systems. A startup period followed during which various operational difficulties were resolved. The pilot facility was ready for

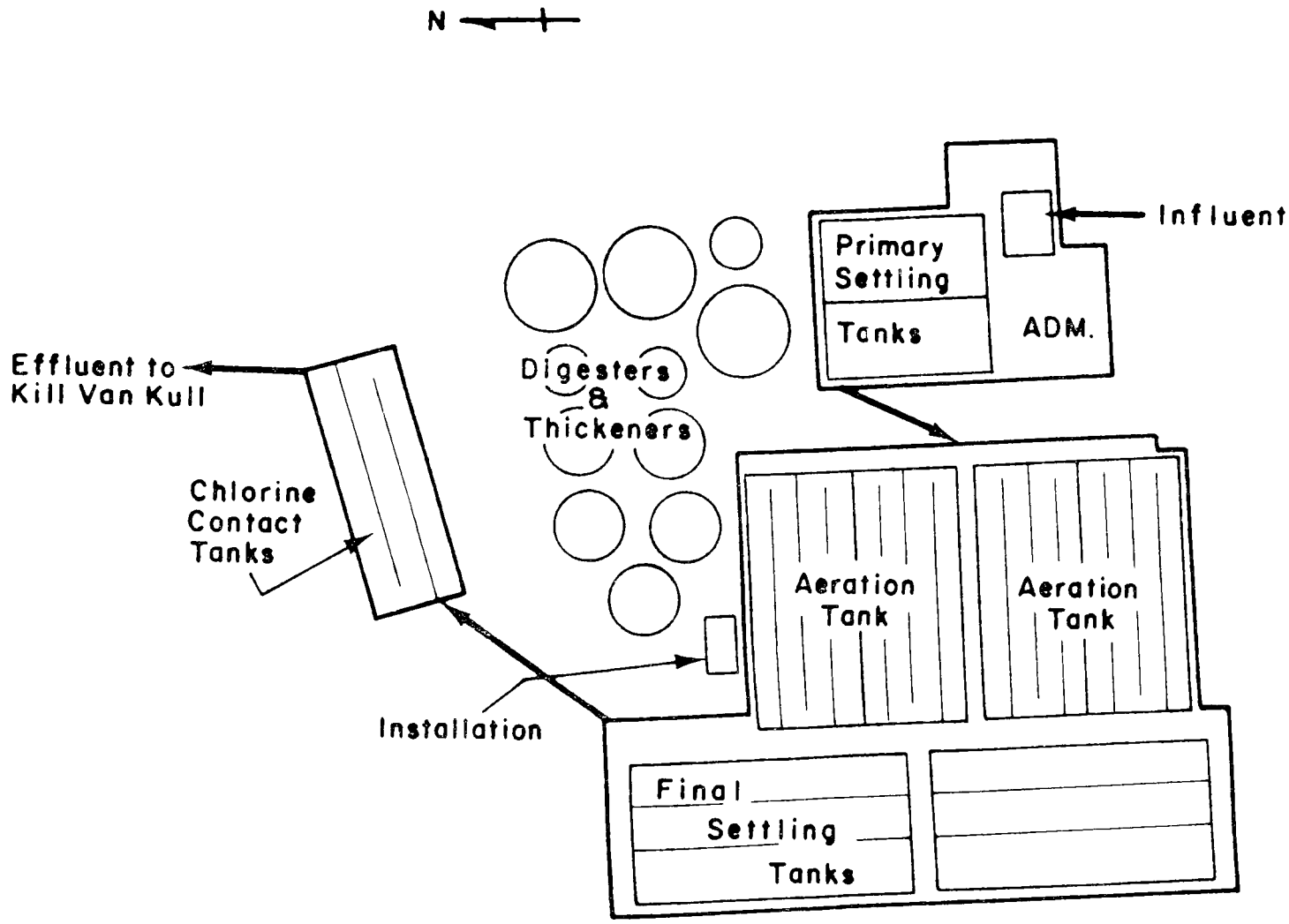

Figure 1.
Schematic Layout of Port Richmond WPCP

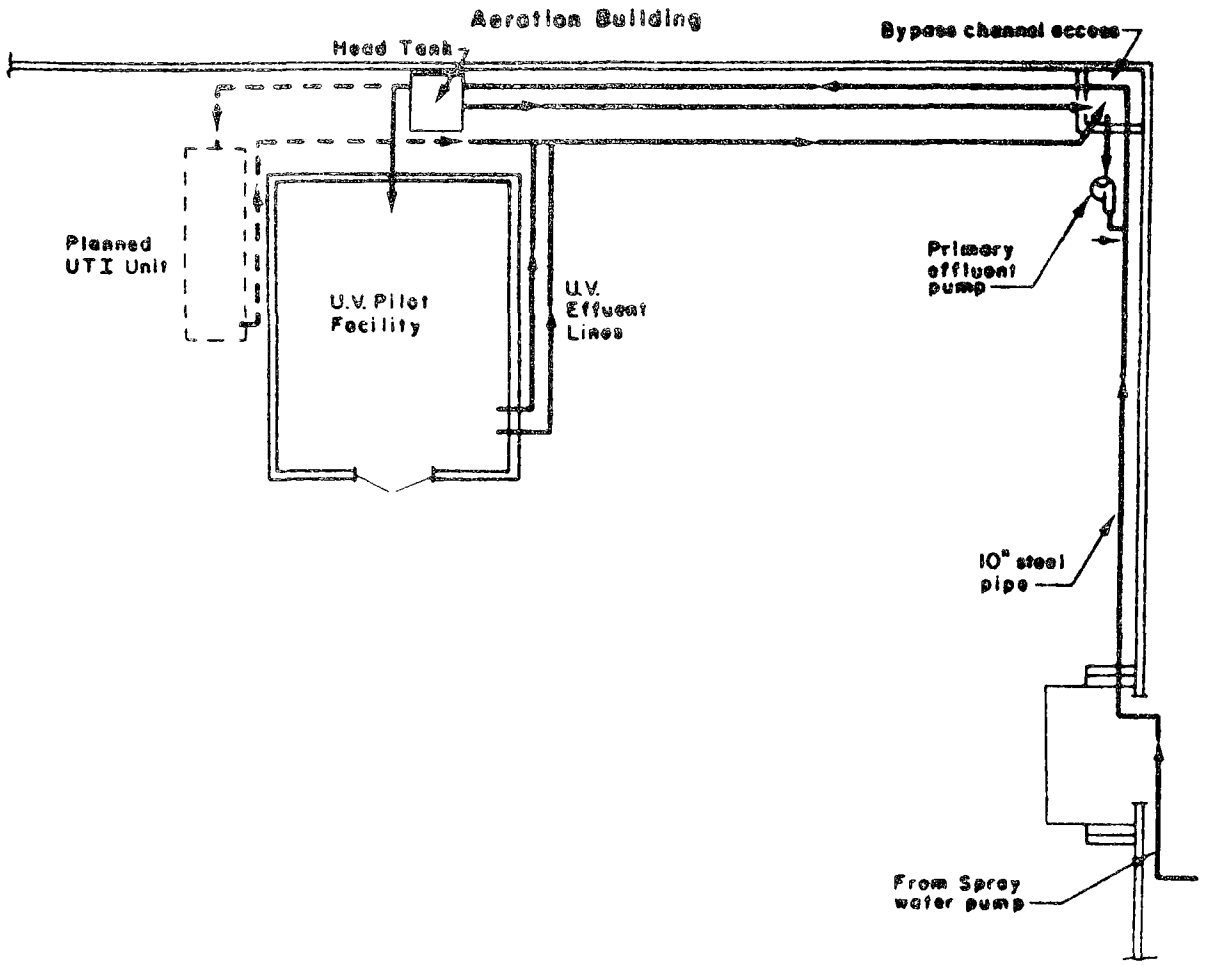

Figure 2.
Schematic of U.V. Pilot Plant Layout

TABLE 1

SYSTEMS INSTALLED OR PLANNED

	1 Contact	2 Contact	3 Non-Contact
No. Lamps	100	100	72
Type	G36T6VH	G36T6VH	G64T5
Power (Watts)	40	40	80
Teflon Tubes (No.)	-	-	32
Spacing	5.0 cm	1.25 cm	(1)
Flow Range (ML/d)	0.76-4.5	0.76-4.5	0.76-4.5
(mgd)	0.2-1.2	0.2-1.2	0.2-1.2
Manufacturer	PWS (2)	PWS (2)	UTI (3)
Void Volume (L)	378	76	(4)
(gal.)	100	20	(4)

(1) Lamps are 13 cm center to center.
Teflon tubes are 6.4 cm diameter, 13 cm center to center

(2) Pure Water Systems, Inc.

(3) Ultraviolet Technology, Inc.

(4) Liquid holding capacity is 9.8 liters/tube; total of 314 liters.

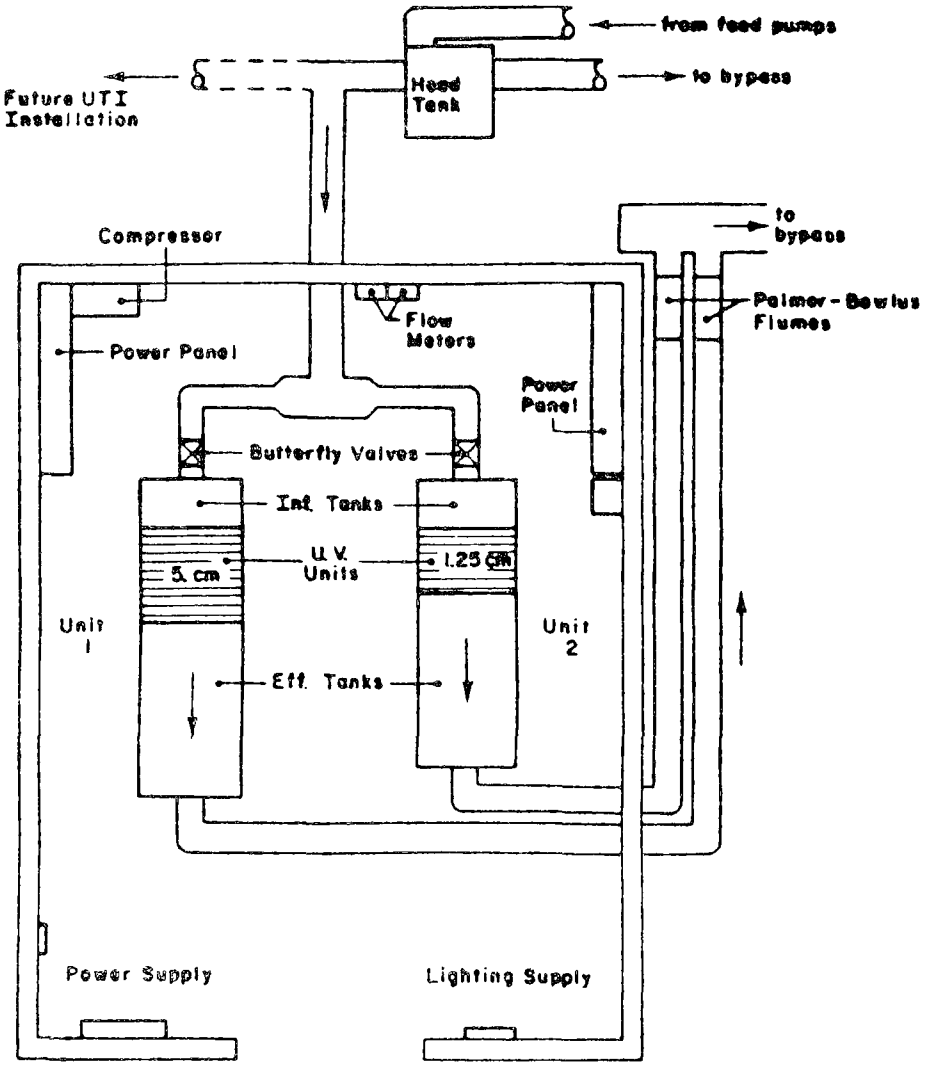

Figure 3.
U.V. Pilot Facility

continuous 24-hr operation and data collection by early December, 1981.

Laboratory facilities at the Port Richmond WPCP are used to support the sampling and monitoring program. Bacterial density measurements (total and fecal coliform, total plate count, fecal strep), suspended solids and turbidity analyses are performed at Port Richmond by HydroQual personnel. Other tests including UV absorbance, nitrogen (organic ammonia, nitrate, nitrite), total organic carbon (TOC), chemical oxygen demand (COD), and pH are performed at HydroQual's laboratory (General Testing Corporation), located in Hackensack, New Jersey.

EXPERIMENTAL PROGRAM

There is little in the way of actual performance data to report. What can be presented, however, is the basic model about which the experimental program is being constructed and two tasks which have been or are nearly completed which address two major elements of the proposed model. Additionally, water quality data which has been collected to date will be presented, indicating the characteristics of the secondary effluent discharged by the Port Richmond plant.

The modeling framework and calculations will be described only in general terms as they apply to Port Richmond. The reader is cautioned that these discussions are preliminary and subject to modification as the study progresses. The basis of the model and other calculations will be presented in more detail in subsequent conference presentations and in the final report.

Proposed Model

The proposed model to describe system performance for the disinfection of wastewater is expressed as follows:

$$\frac{L}{L_o} = \exp\left[\left(\frac{ux}{2E_x}\right)\left\{1 - \left(1 + \frac{4kE_x}{u^2}\right)^{1/2}\right\}\right] \qquad (1)$$

where L = residual bacterial density (colonies/100 ml)

L_o = initial bacterial density (colonies/100 ml)

u = fluid velocity (cm/sec)

x = distance (forward direction) traveled during exposure (cm)

E_x = dispersion coefficient in forward direction (cm^2/sec)

k = rate coefficient (sec^{-1})

The fluid velocity, u, is computed as

$$x(Q/V_v)$$

In the case of the PWS units at Port Richmond, x is the longitudinal dimension of the lamp battery (cm). The term V_v is the void volume of the lamp battery (cm^3) and Q is the wastewater flow rate (cm^3/sec).

Fluid flow is assumed to be completely mixed in the plane perpendicular to the direction of flow. The term E_x describes dispersion only in the forward direction. The rate coefficient (k) is a function of the intensity of ultraviolet light, i.e. the rate at which energy is being delivered to the wastewater. Intensity, in turn, is a function of the UV output of the lamps, the placement (spacing) of the lamps and the absorptive properties of the wastewater.

Hydraulic Evaluations

A series of tests has been performed to define the time distribution and flow characteristics through each of the submerged systems at Port Richmond. Prior to this each unit was evaluated to determine if there existed an acceptable approach condition, i.e. no shortcircuiting or significant velocity gradients across the front plane of the lamp battery. Significant gradients were found, particularly on the widely spaced unit. This was due (on both units) to the position of the inlet pipe and the relatively small size of the influent tank. Sufficient time and volume were not available to dissipate and equalize the velocity before entering the lamp battery. An overflow weir and stilling wall were subsequently installed in each influent tank to correct this problem. Further tests and observations indicated a good flow distribution across the plane of the lamp battery, with no evidence of shortcircuiting.

Mixing within each UV unit was evaluated using real time measurements of conductivity. Salt (NaCl) was used as the tracer. The normal procedure for developing detention time curves, i.e., taking discrete samples after an instantaneous tracer injection, was not possible given the system design at Port Richmond. The theoretical detention times (V_v/Q) were 2 to 30 seconds, leaving little time to practicably inject a sufficient quantity of tracer and collect an adequate number of samples to construct the trace. Additionally, the design essentially simulates an open channel with the lamp battery inserted across the width of the channel, making it very difficult to take representative discrete samples.

A new procedure was developed, as shown schematically on Figure 4. A concentrated salt solution was injected at a constant rate at a selected location immediately in front (approximately 3.8 cm in front of lamp plane) of the lamp battery. A conductivity probe was used to search for the point of maximum concentration on the exit side of the lamp battery. Note that the probe was also used to scan the entire exit plane in order to define the cross-section and location of the plume as it exited the lamp battery.

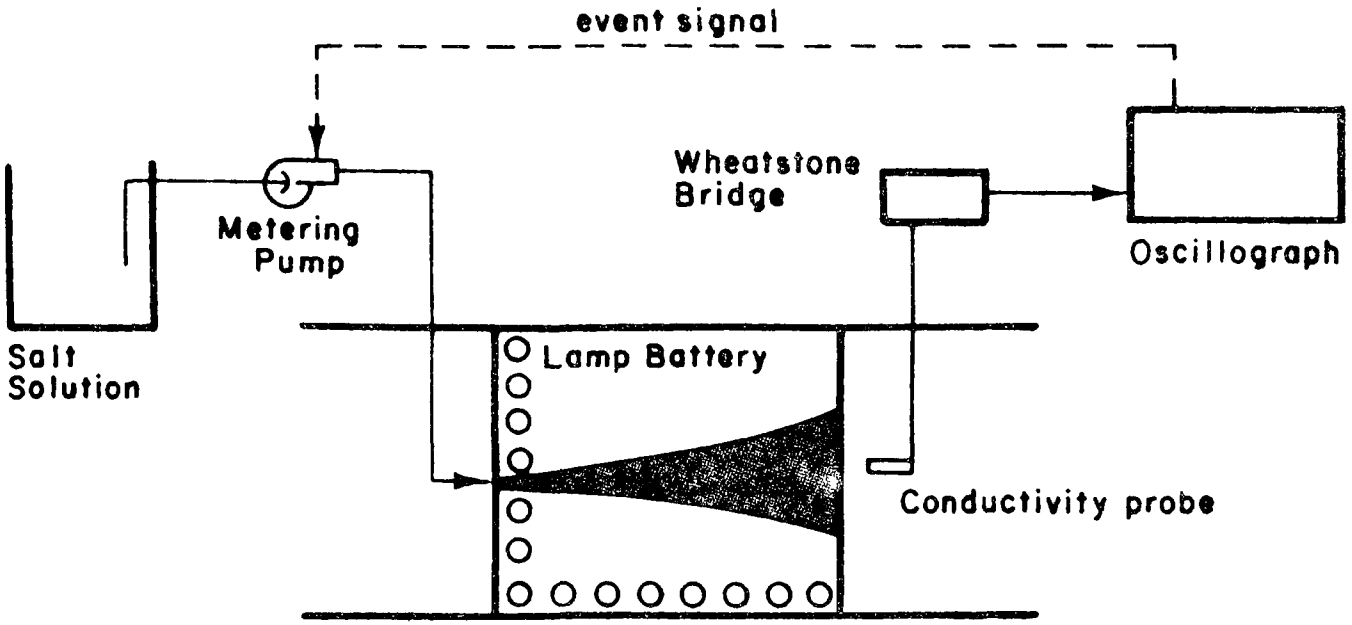

Figure 4.
Experimental Setup for Retention Studies

Once the probe was situated and fixed at the center of the plume, a steady state condition was allowed to develop at fixed wastewater and salt solution flow rates. The high frequency output from the conductivity meter was amplified and continuously recorded using an oscillograph. Continuous permanent tracings are made by a light beam onto light sensitive recording paper advancing at a rate of 0.64 cm/second. The recorder/meter was first calibrated by measuring the conductivity of known salt solutions.

Once steady state was indicated by the recorder, the salt solution pump was shut-off. This event was automatically signalled to the oscillograph and recorded. The die-away of salt was then monitored by the fixed probe and continuously recorded. Readings (conductivity) were then taken off the trace, converted to salt concentration (mg/l) and transposed to a plot of concentration against time.

Figure 5 graphically presents the method used to analyze the resulting trace. The upper plot shows concentration against time. The derivative of this curve is taken by plotting the slope (dc/dt) of tangents drawn at several locations along the curve. This is shown on the lower plot and resembles the typical curve derived from an impulse release.

Assuming the flow through the lamp battery, as shown on Figure 6, is completely mixed in the y and z plane and disperses only in the x direction, the response to an impulse release may be described as

$$\frac{dc}{dt} = \frac{W}{2A\sqrt{\pi E_x}} \frac{1}{\sqrt{t}} \exp\left[\frac{-(x-ut)^2}{4E_x t}\right] \qquad (2)$$

where

W = mass input (mg/sec)
A = cross sectional area (cm^2)
c = concentration (mg/l)
t = time (seconds)

At $t - t_o$, where t_o is the theoretical detention time, a constant, y, is defined

$$\frac{W}{2A\sqrt{\pi E_x}} = y\sqrt{t_o} \qquad (3)$$

This constant is then substituted into Equation (2),

$$\frac{dc}{dt} = y\sqrt{t_o} \exp\left[\frac{-(x-ut)^2}{4E_x t}\right] \qquad (4)$$

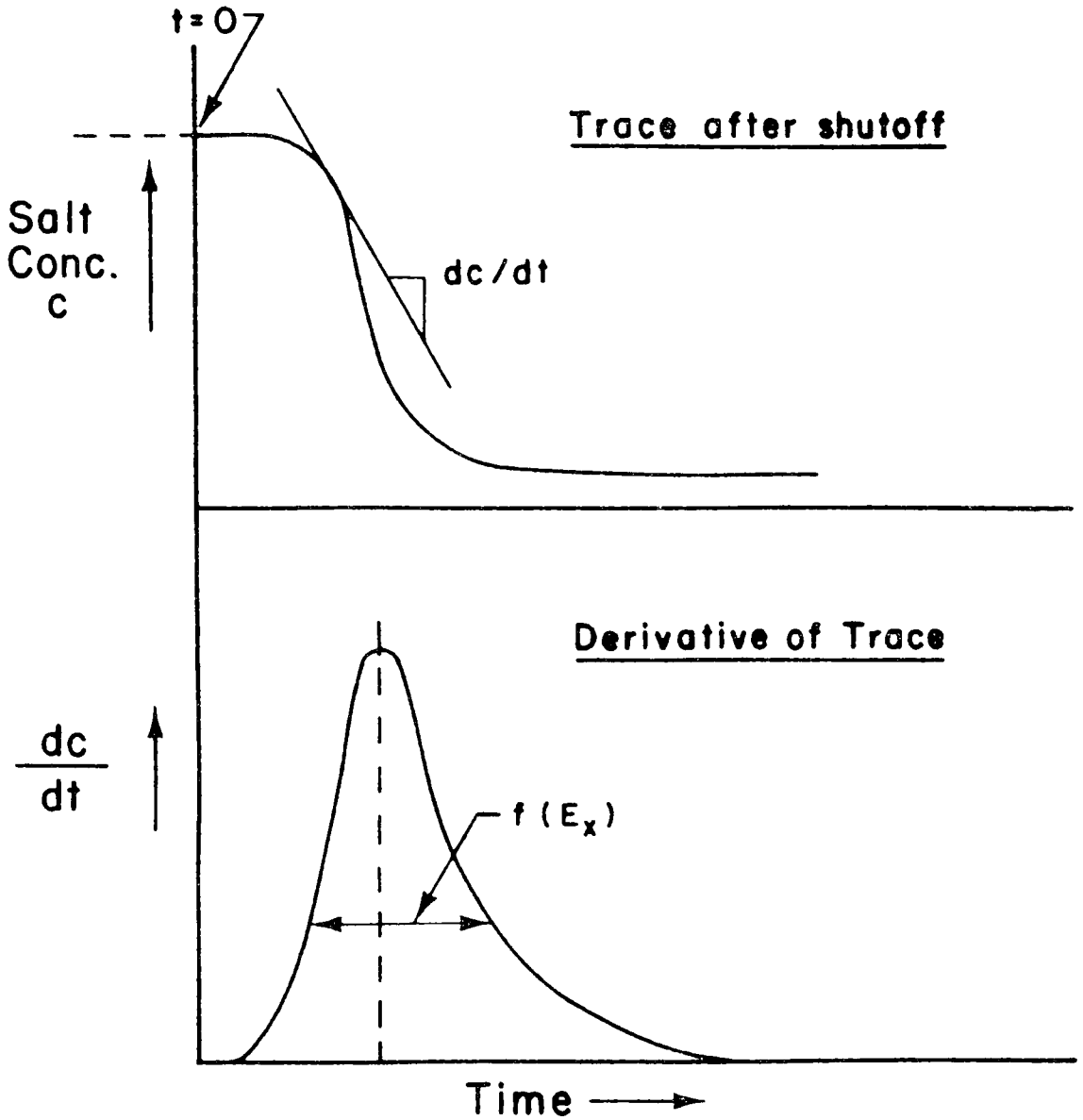

Figure 5.
Analysis of Hydraulics Data

Assumes completely mixed in y, z
dispersion in x

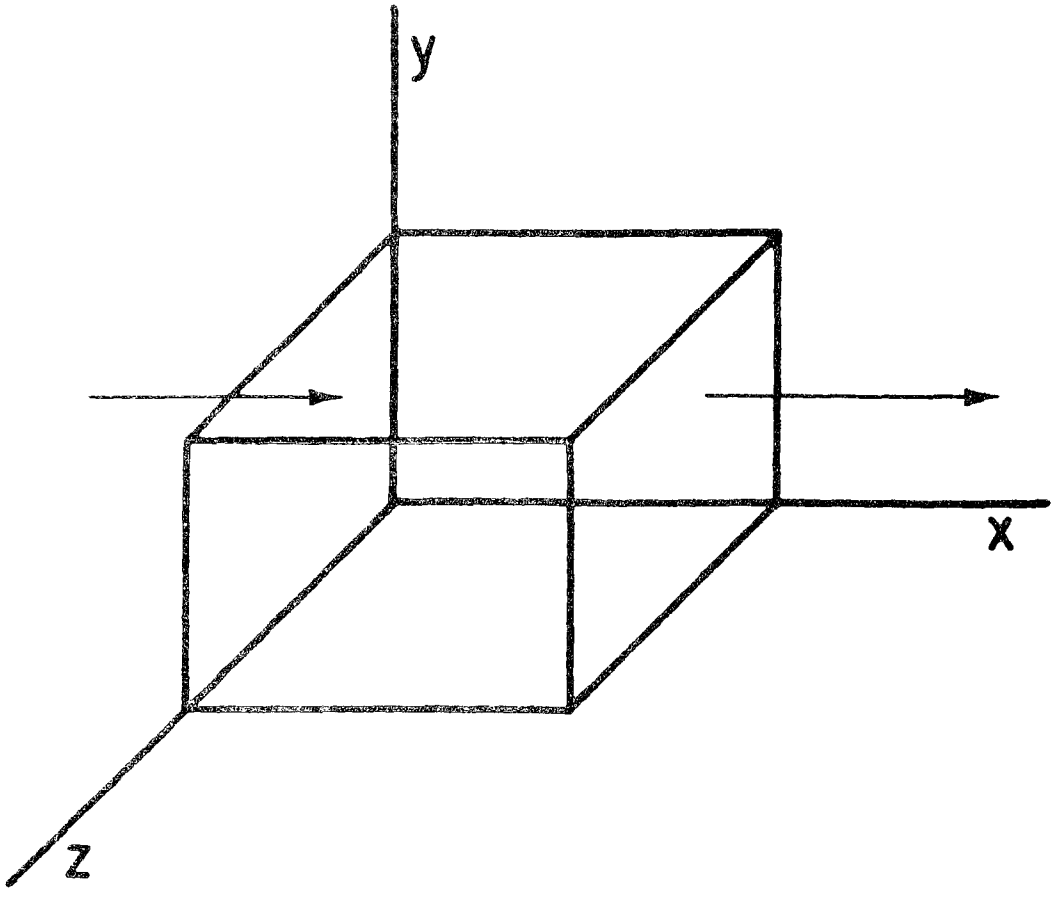

Figure 6.
Mixing Assumptions

Knowing the values of dc/dt, y, t_o, x, u, and t, the dispersion coefficient E_x can be estimated by a trial and error procedure to fit the data.

An example of the procedure is presented on Figure 7. These data are from a test run on Unit No. 2 (closely spaced lamp unit). The point of injection is shown on the upper display. The lower display presents the slope calculations as data points. The smooth line is a solution of Equation (2) at an E_x = 10 cm^2/sec.

The results of several runs on each unit, made at differing flows and injection point locations, indicated an average E_x of 1.5 cm^2/sec for Unit No. 1 (widely spaced unit) and 15 cm^2/sec for Unit No. 2 (closely spaced unit). This implies that the flow characteristics of Unit 1 correspond more closely to a plug flow condition (E_x approaches zero) than does Unit 2. It should be noted, however, that both units can be considered to closely simulate a plug flow condition relative to the opposing condition of complete mix, when E_x approaches infinity.

Figure 8 presents a series of solutions to Equation (1) which demonstrates the sensitivity to E_x. The log of the survival ratio (L/L_o) is plotted as a function of the rate coefficient (k) for various values of E_x. This is shown for both systems installed at Port Richmond at a flow of 1.9 ML/d (0.5 mgd). It is evident that significant deviation from the idealized flow (plug flow) condition can result in multilog increases in the survival ratio. This emphasizes the importance which must be placed upon the hydraulic characteristics of a system design.

ULTRAVIOLET INTENSITY

The rate of disinfection is directly related to the intensity of ultraviolet light, i.e. the rate at which energy is delivered to the wastewater medium by the UV source. Current system designs, which generally involve lamps or lamp bundles immersed or surrounding the receiving medium, have precluded any practical means to directly measure the true intensity at any point within a system.

A mathematical model has been developed as an element of this study which calculates the intensity at any point within a UV system and which can estimate the average intensity emitted by a specific unit. The calculations are based on the point source summation method described by Jacob and Dranoff (1) and recently applied to disinfection systems by Johnson and Qualls (2). The mathematical techniques rely on the basic physical properties of the ultraviolet lamps, the configuration of the multilamp chambers, and the properties of the aqueous medium.

UV energy emitted by a lamp is attenuated as the distance from the energy source increases. This attenuation occurs via two mechanisms: dissipation and absorption. Dissipation simply describes the dilution of the energy as distance from the source increases. The surface area over which the output of energy is projected increases with increasing distance,

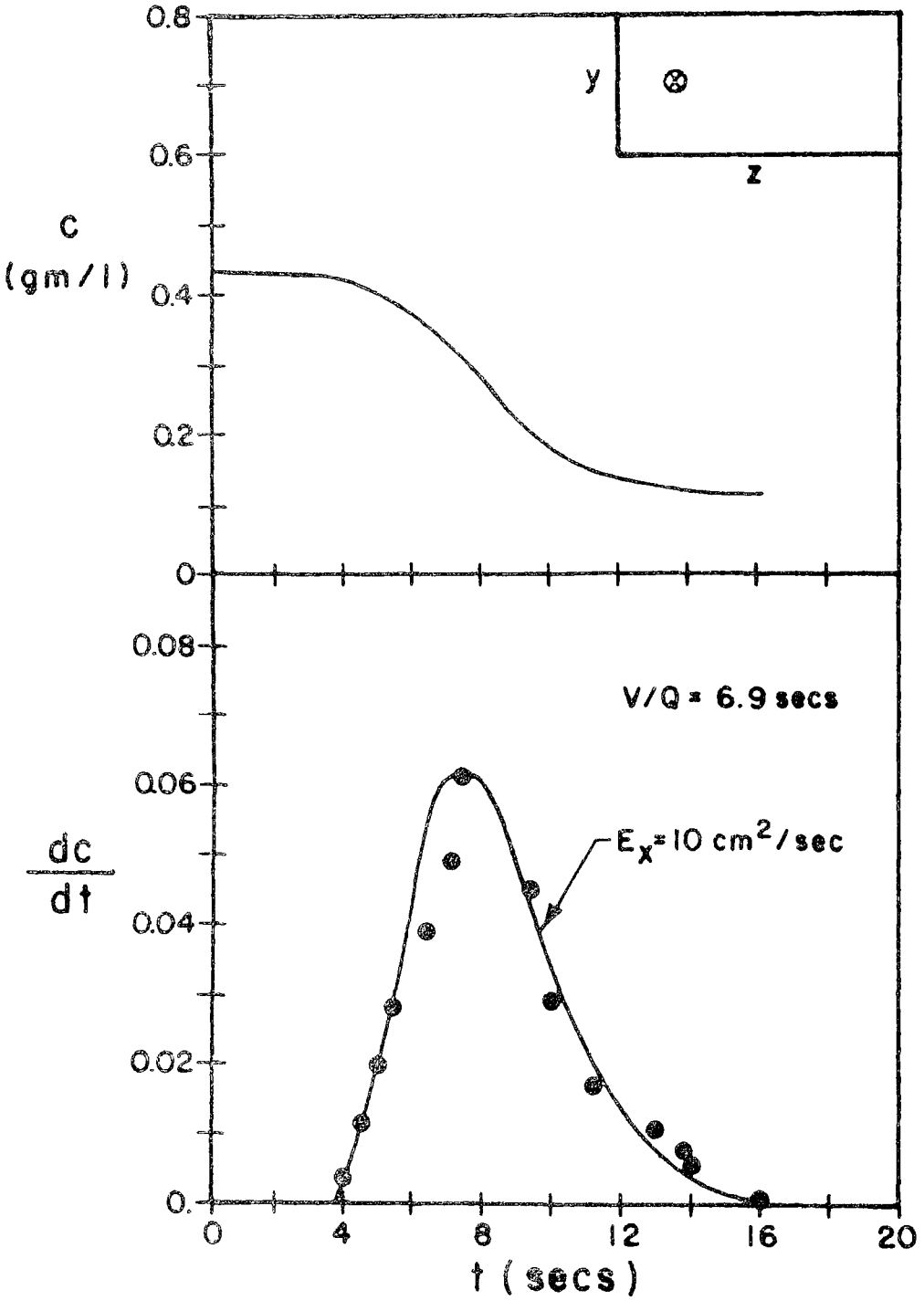

Figure 7.
Tracer Analysis

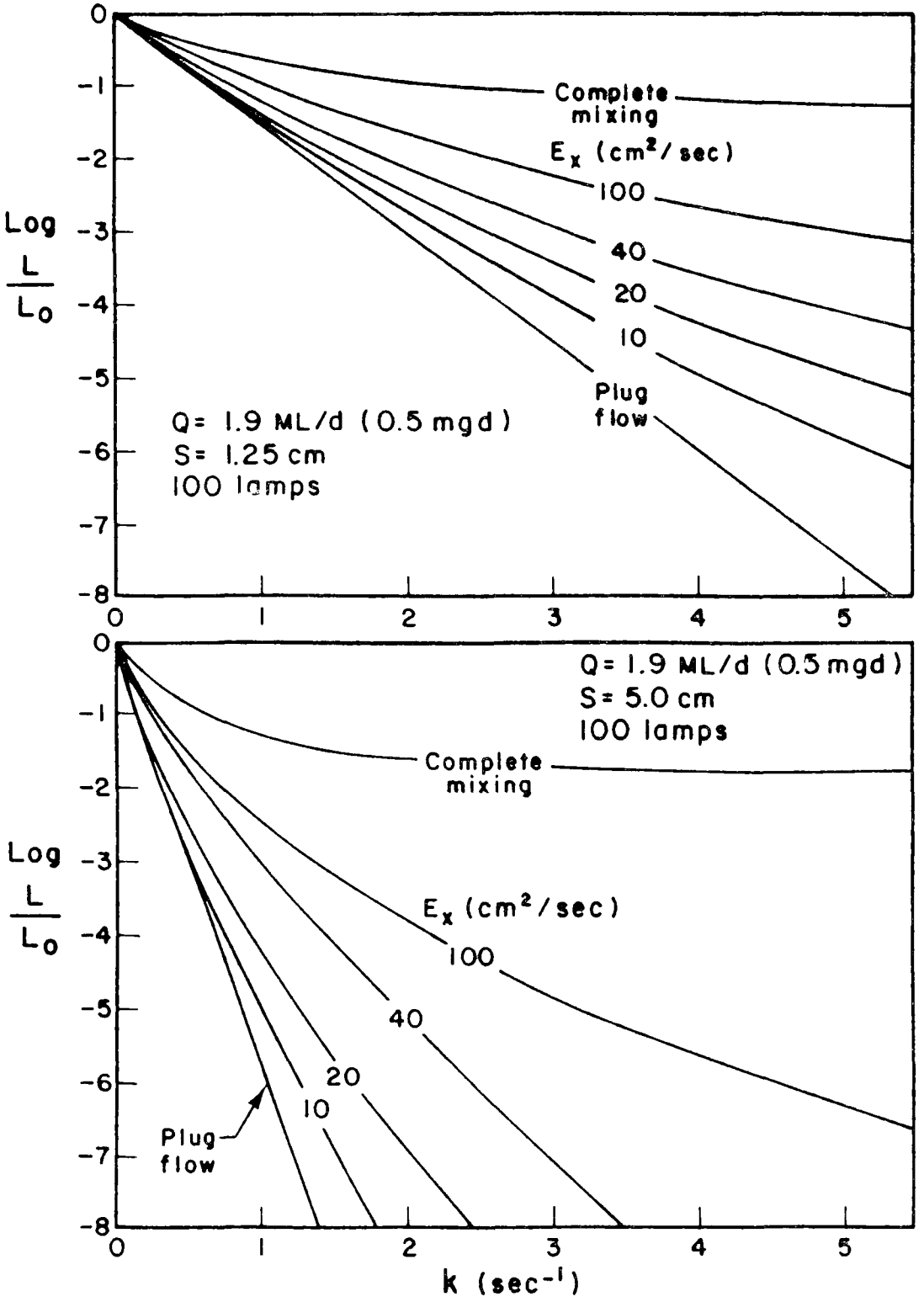

Figure 8.
Sensitivity to Dispersion Coefficient

thus there are fewer photons striking each unit of surface area. This dissipation can be calculated by surrounding an energy source by a sphere of radius R:

$$I = \frac{S}{4\pi R^2} \tag{5}$$

where I = intensity at distance R (cm) in watts/cm^2
S = output of UV energy source in watts

The second attenuation mechanism relates to the absorptive properties of the medium through which the energy is transmitted. This is described by Beer's Law:

$$I \propto I_o\, e^{-\alpha R} \tag{6}$$

where I_o = intensity at a given point (watts/cm^2)
α = absorbance coefficient (cm^{-1})
R = distance from the point of I_o (cm)

Combining equations (5) and (6) yields

$$I = \frac{S}{4\pi R^2}\, e^{-\alpha R}$$

which describes the intensity at a given distance from a point source of energy.

The tubular germicidal bulb is treated in this calculation as a series of point sources. The intensity at a specific point is then the sum of the intensities from the individual point sources:

$$I(r,z) = \sum_{n=1}^{n=N} \frac{S/N}{4\pi(r^2+z^2)} \exp[-\alpha(r^2 + z^2)^{1/2}] \tag{7}$$

where $z = L\left(\frac{n-1}{N-1}\right)$

N is the number of point sources into which the line source is divided and r and z describe the coordinates of the "receiver" at which the intensity is being computed ($R^2 = r^2 + z^2$). This is shown schematically on Figure 9. To calculate intensity at point (r,z), Equation (7) must be applied N times and all solutions summed.

An assumption inherent to Equation (7) is that the receiver located at (r,z) is spherical and infinitely small. Thus, energy emitted from any

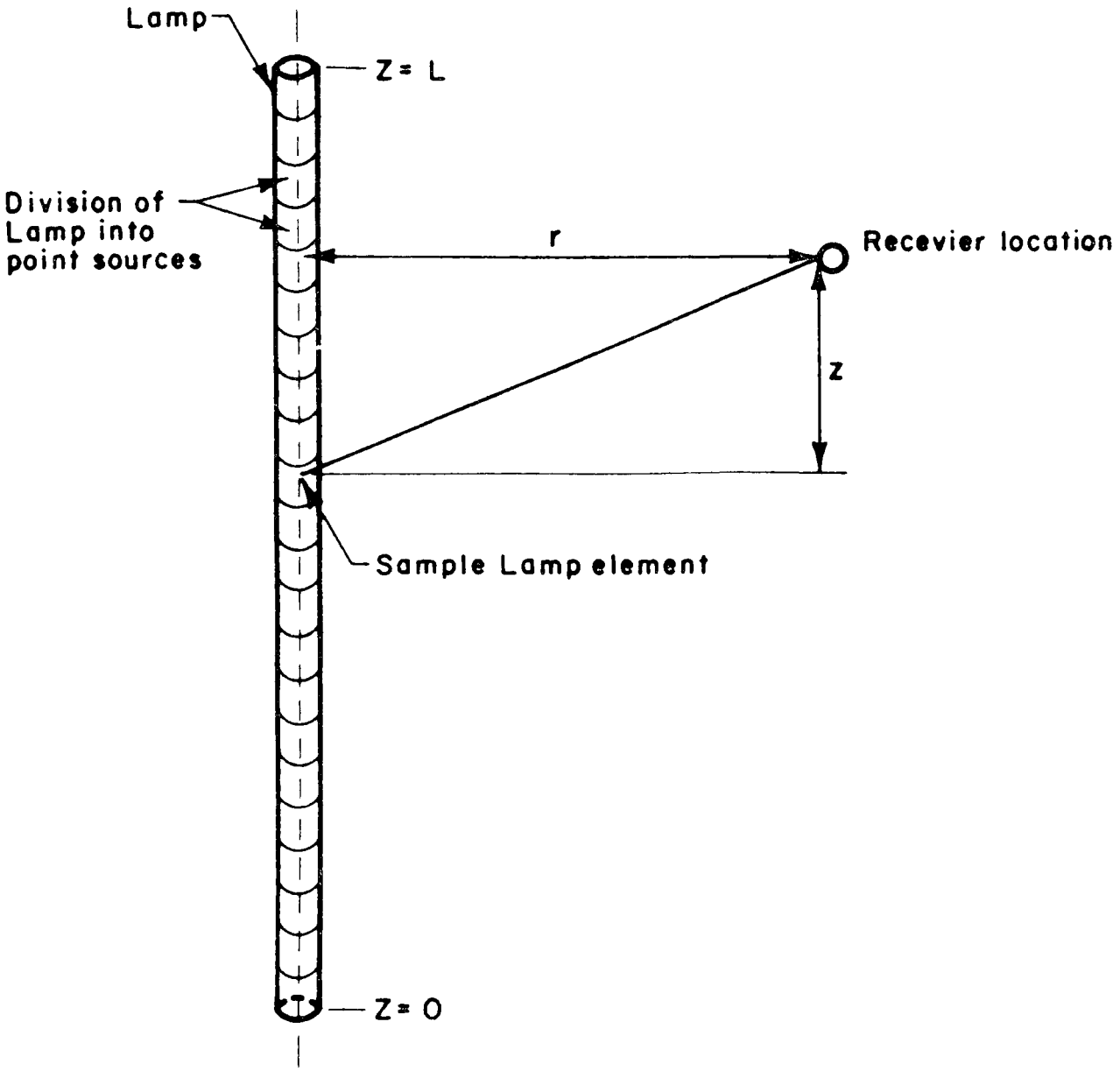

Figure 9.
Lamp geometry for point source approximation

point source element of the lamp will strike the receiver normal to its surface.

The computer model uses Equation (7) as the basic element to compute the intensity in a specific system. The model is capable of accounting for

- absorption of energy as it passes through various elements such as wastewater, quartz sleeves, teflon, air and neighboring lamps
- any system lamp configuration including assymetrical arrays, as long as the lamps are parallel to one another
- any lamp rating for UV output
- any lamp battery size (no. of lamps) and variation in output

Figure 10 displays preliminary workups for the two systems presently installed at Port Richmond. Average computed intensity is plotted for each system as a function of the UV absorbance coefficient (at 253.7 nm) of the wastewater. The lamps in this instance have a rated output of 14W each at the 253.7 nm wavelength. The reader is cautioned that these analyses are preliminary at this point. More work is anticipated to refine the calculation techniques and to experimentally confirm and/or modify the key parameters which comprise the model.

The utility of the mathematical modeling technique is that it allows an analysis of a system's sensitivity to the variables which impact its design. As an example, a key parameter in certain system designs is the spacing of the lamps. Spacing will affect the average intensity within the system, the detention time and may influence the hydraulic characteristics of a unit. The two units at Port Richmond differ in spacing by a factor of four (1.25 cm and 5 cm); as Figure 10 shows, the model indicates that the ratio of I_{avg} for Unit 2 to I_{avg} for Unit 1 increases from approximately 2.3 at an α_w of 0.2 cm^{-1} to approximately 4.1 at an α_w of 0.8 cm^{-1}. This loss of efficiency in the closely spaced unit at the lower water absorbances is attributed to the "shadowing" effect of neighboring lamps. The lower the absorptive property of the wastewater, the further the UV can penetrate. If the lamp spacing is such that this energy hits a neighboring lamp, it will be absorbed by that lamp.

Figure 11 displays the effect of this phenomenon. The percent reduction shown on the ordinate represents the loss of energy to neighboring bulbs. This is shown as a function of bulb spacing for varying wastewater absorbance coefficients. This analysis shows that the spacing of bulbs is clearly a design parameter which will be dictated by the quality of the water to be treated. Other design relationships are being developed with the model, with field verification at Port Richmond.

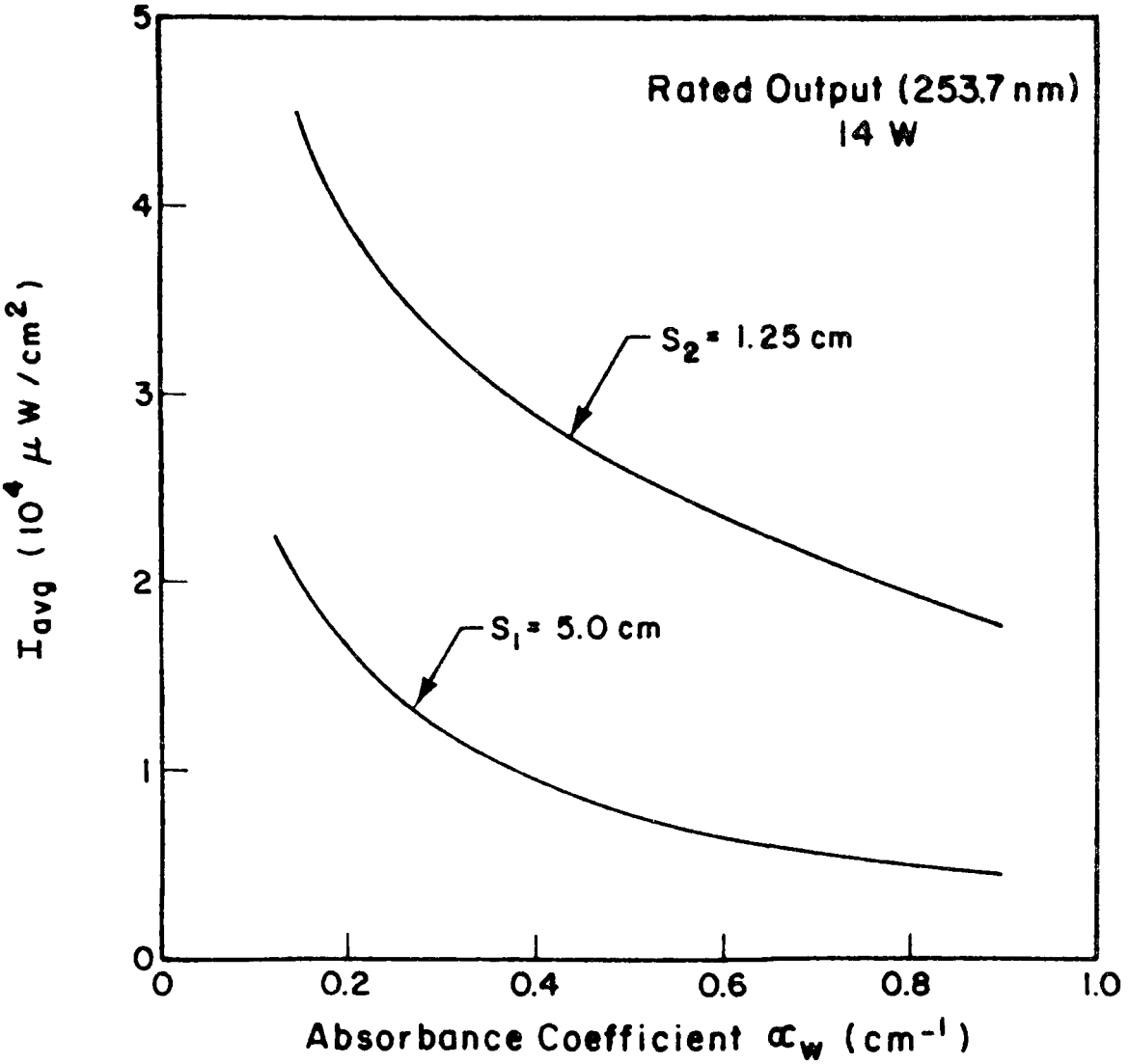

Figure 10.
Computed Intensity

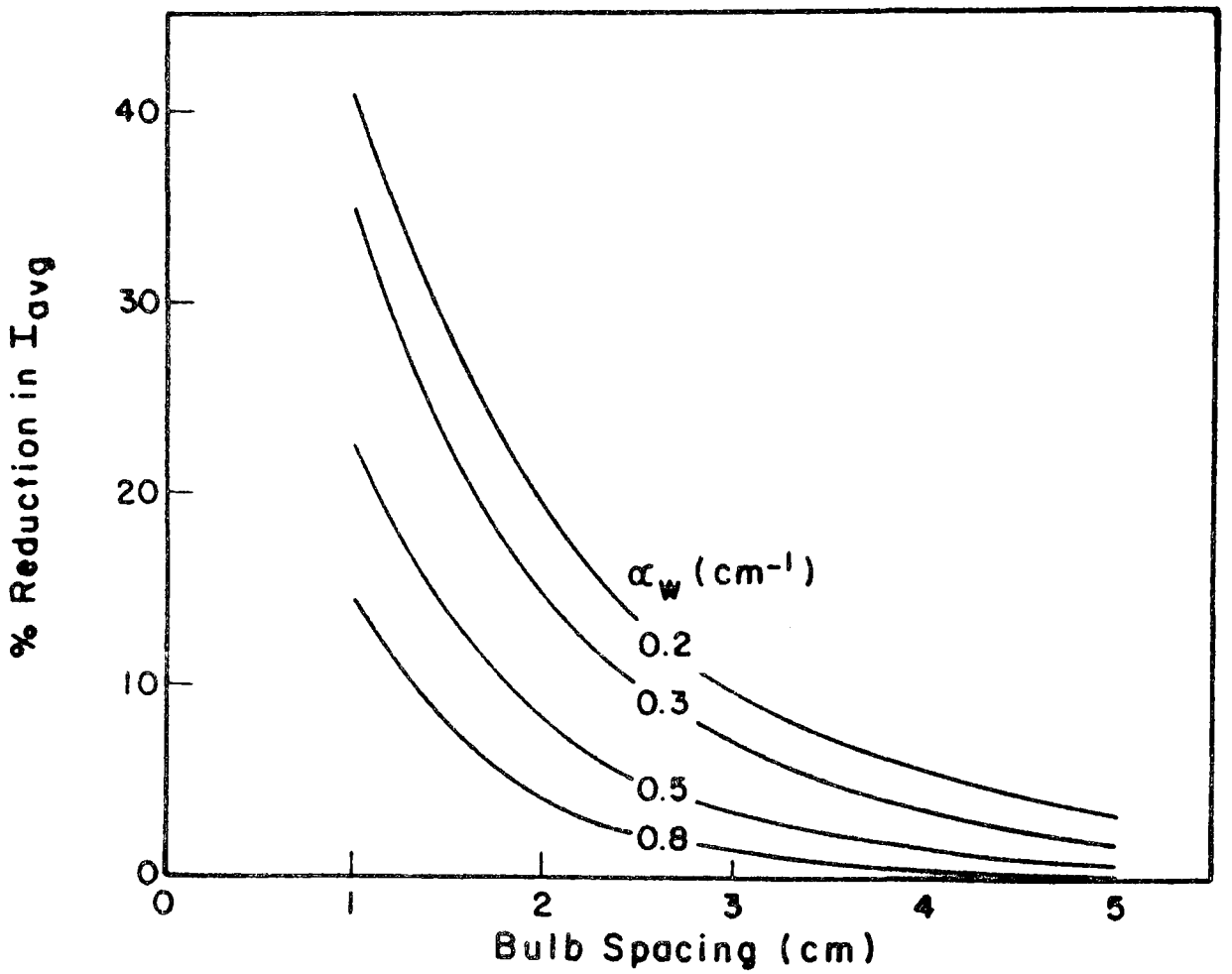

Figure 11.
Intensity Reduction vs. Bulb Spacing

WATER QUALITY

Table 2 presents a summary of analyses on the influent to the UV systems (secondary effluent). The data are limited, representing 14 days of sampling (two to four samples/day) in December, 1981, and January, 1982. The COD has been variable, with a mean of 45.4 mg/l (total). The suspended solids have averaged 20 mg/l, ranging as high as approximately 60 mg/l.

Two methods are employed to measure the absorbance (at 253.7 nm) of the wastewater. In both cases a Perkin-Elmer Model 552 Double-Beam Scanning UV/Visible Spectrophotometer is utilized. The first method is to simply measure the absorbance of a direct beam through a 1 cm cell. This is designated as the "direct" UV absorbance. The second method incorporates an integrating sphere attachment to the spectrophotometer. This accounts for light that may be scattered (and not measured by the direct beam method) and is not absorbed. Thus the "sphere" absorbance more closely corresponds to the true absorbance of the samples. The data on Table 2 indicate that the true absorbance is not significantly affected by suspended/colloidal solids. Rather the energy is scattered by these particles and remains available for disinfection purposes. Conversely there appears to be little penetration or absorbance of the energy by the particles, precluding the inactivation of organisms occluded by such material.

SCHEDULED TASKS

A major fraction of the experimental program lies ahead. Specific tasks anticipated for the project include the following:

- evaluation of the kinetics associated with disinfection
- confirm UV intensity parameters
- verify the proposed disinfection model by operation of the units under equivalent performance conditions
- install and similarly evaluate the teflon system
- evaluate the impact of photoreactivation under warm and cold temperature conditions
- monitor each system for cleaning, O&M needs, reliability

CLOSING

Ultraviolet light disinfection is a viable, cost effective process which is quickly emerging as an alternative for wastewater disinfection. The Port Richmond project will seek to develop needed information in the area of design methods and the definition of critical process performance parameters. Although we were unable to present a substantive store of data

TABLE 2

INFLUENT ANALYSIS

14 Days
2 - 4 samples/day

	mean	σ
COD_T (mg/l)	45.4	15.0
COD_F (mg/l)	34.8	16.4
TOC_T (mg/l)	16.4	6.0
TOC_F (mg/l)	14.9	6.0
SS (mg/l)	20.0	16.5
Turbidity (NTU)	5.5	4.9
U.V. abs. Direct (cm^{-1}), αw		
T	0.400	0.114
F	0.319	0.066
U.V. abs. Sphere (cm^{-1}), αw		
T	0.309	0.068
F	0.297	0.068

in this paper, the participants will make an effort to disseminate, via conference papers, the results of the study as it progresses.

ACKNOWLEDGEMENTS

The authors wish to express their thanks to Mr. Joseph McAllister, Plant Superintendent, and the Port Richmond plant operators for their help and interest in the project. We would also like to acknowledge the support and contributions being made by Mr. William Pressman, Director of R & D, New York City DEP; Mr. Gerard Cox, Engineer NYC DEP; Mr. Wilfred Dunne, Field Technician, HydroQual; and Ms. Maureen Casey, Engineer, HydroQual.

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COMPARISON OF ANALYTICAL METHODS FOR RESIDUAL OZONE

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ABSTRACT

Seven analytical methods for the determination of residual ozone in water and waste water have been compared by measuring the decomposition of ozone in water and waste water. This kinetic technique minimizes sampling errors and allows a direct comparison of methods under conditions of rapidly changing ozone concentration. Changes in the ozone-reductant reaction caused differences in ozone decay curves. Conditions which reduce ozone decay prior to the ozone-reductant reaction reduced differences among methods.

The analytical methods are compared on the ease of calibration of the reagent solutions, the stability of the reagent solutions, and the stability of the titer of the ozonated reagent solution.

INTRODUCTION

Over the last eighty years, chlorine has been widely used for disinfection of municipal and industrial waste waters. Recent concerns, however, over the toxic effects of chlorinated organic by-products produced during chlorination of potable water and waste water have renewed interest in ozone in water treatment. Ozone acts as an oxidant to remove taste, color, odor, and organic matter from water as well as serving as an effective disinfectant. The United States Environmental Protection Agency requires of any disinfectant that its residual be measured accurately and conveniently. In the case of ozone, the residual may vary from 0.05 mg/L to 30 mg/L depending on reaction time, sample contamination and dosage level. Residual ozone levels below 5 mg/L are of most interest in water treatment (15).

With a standard reduction potential of 2.07V in acid solution and 1.24V in basic solution, ozone will react with most oxidizable substances.

Ozone is usually generated by passing a stream of dry oxygen through an electrical discharge which converts 2-5% of the oxygen to ozone. Therefore, ozone, the species of interest, is only a small fraction of the gas mixture.

It would be ideal if methods for determining residual ozone could be verified by the analysis of weighed samples of pure ozone. This is impossible, however, due to the instability of pure ozone, the low solubility of ozone in water, the high volatility of ozone, and the rapid decomposition of ozone in water.

Most analytical methods for the determination of aqueous ozone take advantage of the property of ozone as a strong oxidizing agent. Some of the most popular reductants are iodide ion (6), arsenic(III) (10), and indigo

blue (2). An amperometric membrane electrode measures ozone in solution (13). The objective of this paper is to evaluate and compare these analytical methods and explain any differences.

The oxidation of iodide ion to iodine by ozone with subsequent titration of the iodine formed is the classical method for the determination of residual ozone (6). The reaction of ozone with iodide ion is described by

$$O_3 + I^- \rightarrow O_2 + IO^- \qquad \text{fast}$$

$$IO^- + H_2O \rightarrow HIO + OH^- \qquad \text{fast}$$

$$HIO + 2I^- \rightarrow I_3^- + OH^- \qquad \text{slow}$$

$$3\ HIO + 3OH^- \rightarrow IO_3^- + 2I^- + 3\ H_2O \qquad \text{very slow}$$

Upon acidification the species hypoiodite ion (IO^-), hypoiodous acid (HIO), and iodate ion (IO_3^-) are all converted to triiodide ion (I_3^-) so that the overall process is ideally

$$O_3 + 3I^- + H_2O \rightarrow I_3^- + O_2 + 2OH^-$$

and theoretically one molecule of ozone liberates one molecule of titratable iodine. Thermodynamically, the ozone oxidation of iodide ion and iodine to form iodate ion is favored at high pH.

$$3O_3 + I^- \rightarrow 3O_2 + IO_3^-$$

$$5O_3 + I_2 + H_2O \rightarrow 5O_2 + 2IO_3^- + 2H^+$$

Furthermore, at a pH above 9, iodine is unstable, readily undergoing disproportionation.

$$I_2 + 2OH^- \rightarrow IO^- + I^- + H_2O$$

$$3IO^- \rightarrow 2I^- + IO_3^-$$

Regardless of whether iodate ion is formed by direct oxidation or disproportionation of iodine, for every three moles of ozone absorbed in the pH region above 9, one mole of iodate ion should be formed.

In the standard iodometric method for analysis of ozone in water, ozone is purged into potassium iodide solution and after acidification with sulfuric acid the iodine is titrated with standard sodium thiosulfate (12). The ozone: iodine stoichiometry has been extensively studied and found to range from 0.65 to 1.5 (3,4,5,8,14). The factors affecting the stoichiometry include pH, buffer composition and concentration, iodide ion concentration, and sampling techniques. Modifications in the iodine determination include changes in endpoint detection, pH, and back titration techniques.

Theoretically, both the pH during the initial ozone-iodide ion reaction and the pH during the iodine determination can alter the ozone:iodine stoichiometry. In acid, the ozone:iodine ratio could decrease due to

$$4O_3 + 10\ HI \rightarrow 5I_2 + H_2O_2 + 4H_2O + 3O_2$$

Hydrogen peroxide could oxidize iodide ion also leading to excess iodine,

$$H_2O_2 + 2I^- + 2H^+ \rightarrow I_2 + 2H_2O$$

Air oxidation of iodide ion in acid also leads to a decrease in ozone: iodine ratio

$$O_2 + 4I^- + 4H^+ \rightarrow 2I_2 + 2H_2O$$

Any errors in the assumption that in acid iodate ion, hypoiodous acid, and hypoiodite ion are quantitatively reconverted back to iodine would lead to an increase in the ozone:iodine ratio. In base, iodate ion formation and hypoiodite ion formation lead to low iodine titers. Again, if the iodine determination is carried out under conditions where iodate ion and hypoiodite ion are not quantitatively reconverted back to iodine, the ozone:iodine ratio is high.

In summary, as the pH decreases for the ozone oxidation of iodide ion, the quantity of iodine should increase due to less iodate ion formation, hydrogen peroxide formation, and air oxidation of iodide ion. As the pH for the iodine determination decreases, the iodine titer should increase due to reconversion of iodate ion to iodine and air oxidation of iodide ion. In the Iodometric method ozone is reacted with iodide ion in buffers of pH 3.5 to 9.0. A known excess of sodium thiosulfate is added, the pH adjusted to 2 with sulfuric acid, and the excess thiosulfate ion titrated with standard iodine (12).

In the amperometric method, ozone oxidizes iodide ion at pH 4.5 in the presence of a known excess of sodium thiosulfate, phenylarsineoxide (PAO) or inorganic As(III). Without acidification these excess reagents are then titrated with standard iodine to an amperometric endpoint (12).

In the As(III) back titration method, ozone oxidizes iodide ion at pH 6.8 in the presence of a known excess of inorganic As(III). Without pH change the excess As(III) is back titrated with standard iodine (14). The DPD Method is an iodometric method carried out in phosphate buffer pH 6.4 (7). Ozone oxidizes iodide ion to iodine which then oxidizes N,N-diethyl-p-phenylenediamine cation (DPD) to a pink Wurster cation. The Wurster cation is quantitated colorimetrically. In the direct oxidation of As(III), ozone reacts with either inorganic As(III) or PAO at pH 4-7, the pH is adjusted to 6.5-7 and the excess As(III) species is back titrated with standard iodine (10). The Indigo method is performed at pH 2 (2). Ozone adds across the carbon-carbon double bond of a sulfonated indigo dye and decolorizes it.

The change in absorbance is determined spectrophotometrically. The Delta electrode (13) and the UV method (2), which measures ozone directly by its UV absorption at 259 nm, involve no reagents and no pH restrictions.

The kinetic and mechanistic description of the decomposition of aqueous ozone has been extensively investigated but no detailed mechanism is generally accepted. Results indicate that decay leads to free radicals. The half-life of dissolved ozone is readily affected by pH, UV light, concentration of ozone, and concentration of radical scavengers (1,9,11). The experimental results published prior to and during the 1950's fit either a one-term or a two-term rate law (9). Recent work of Hoigné (11) also supports a two-term rate law.

A kinetic technique was developed for producing ozone solutions of known concentration in the 24-0 mg/L range. By means of this kinetic technique, advantage is taken of the self-decomposition of residual ozone. A steady state solution of ozone is prepared and allowed to decompose. At known time intervals during the decomposition process, the ozone level of the solution is determined by two or more different analytical methods.

The resulting time-concentration profile for each analytical method is graphed and is fitted to the generalized rate law

$$\frac{-d[O_3]}{dt} = k_1[O_3][OH^-] + k_2[O_3]^2$$

using FIT80. FIT80 is a computer program based on the method of Gauss which allows the simultaneous least squares fitting of first and second order parallel reactions. This rate law is a mathematical model which describes time-concentration curves for ozone decay and does not relate directly to a specific mechanism. The calculated rate constants are apparent rate constants and not true rate constants. They are calculated and compared for each kinetic run and are used for method comparisons within each run and not between different runs. The kinetic parameters calculated for each method should be identical if each method gives the same result.

Even though a mechanism is not necessary for the application of the kinetic technique since comparisons among calculated kinetic parameters indicate discrepancies between methods, a general mechanistic scheme aids understanding of potential method differences. Based on a set of clever experiments, Hoigné (11) has recently proposed the mechanism shown below:

$$O_3 + OH^- \rightarrow O_2^{\bullet -} + HO_2^{\bullet} \qquad k_{OH^-} \sim 170\ M^{-1}sec^{-1}$$

$$O_2^{\bullet -} + O_3 \xrightarrow{+H^+} OH^{\bullet} + 2\ O_2 \qquad k_{O_2^{\bullet -}} \sim 1.6 \times 10^9\ M^{-1}sec^{-1}$$

$$OH^{\bullet} + \text{scavenger} \rightarrow \text{products}$$

$$OH^{\bullet} + O_3 \xrightarrow{-H^+} O_2 + O_2^{\bullet -} \qquad k_{OH^{\bullet}} \sim 4.7 \times 10^8\ M^{-1}sec^{-1}$$

$$O_2^{\bullet -} + O_3 \rightarrow OH^{\bullet} + 2O_2$$

$$2\ HO_2^{\bullet} \rightarrow H_2O_2 + O_2$$

$$HO_2^{\bullet} + O_2^{\bullet -} \rightarrow HO_2^- + O_2$$

$$H_2O_2 + OH^- \rightleftharpoons H_2O + HO_2^- \qquad pK_a = 11.65$$

$$HO_2^- + O_3 \rightarrow OH^{\bullet} \qquad k_{HO_2^-} \sim 5 \times 10^6\ M^{-1}sec^{-1}$$

Ozone decay is initiated by hydroxide ion attack on ozone to form the superoxide radical anion ($O_2^{\bullet -}$) and hydroperoxyl radical ($HO_2^{\bullet}$). Then in an almost diffusion controlled reaction, superoxide radical anion reacts with a second ozone molecule to form the hydroxyl radical ($OH^{\bullet}$). This hydroxyl radical either can react with any radical scavenger present or can react with an ozone molecule in an almost diffusion controlled reaction to generate another superoxide radical anion which in turn reacts with ozone to generate another hydroxyl radical.

The hydroperoxyl radical, a by-product of ozone decay, can dimerize to hydrogen peroxide or can react with superoxide radical anion to form hydroperoxyl anion. Hydrogen peroxide reacts with ozone slowly. Hydroperoxyl anion, however, can catalyze ozone decay by attack on ozone to form the hydroxyl radical in a very fast reaction. A hydrogen peroxide concentration in excess of 10^{-7} M will make the reaction of hydroperoxyl anion with ozone as important as the reaction of hydroxide ion with ozone. The concentration of hydrogen peroxide found as a reaction product increases with a decrease in pH.

According to this mechanism, the lifetime of ozone in aqueous solution depends on added solutes or impurities. The hydroxyl radical, which forms upon ozone decomposition, is a chain carrier for further ozone decomposition and any solute or impurity which scavenges this radical will retard ozone decomposition.

A simplified mechanism consistent with Hoigne's model is shown below.

$$O_3 + OH^- \rightleftharpoons [O_3{\cdot}OH^-]$$

$$[O_3{\cdot}OH^-] \rightarrow O_2^{\bullet -},\ HO_2^{\bullet},\ OH^{\bullet},\ H_2O_2 + \text{products}$$

$$[O_3{\cdot}OH^-] + O_3 \rightarrow \text{products}$$

Initially ozone complexes with hydroxide ion and this reactive intermediate can undergo either a first order electron transfer reaction or a second order reaction with ozone. The first order electron transfer reaction could lead to the formation the superoxide radical anion, the hydroperoxyl radical and the hydroxyl radical which in turn either act as chain propagators and lead to further ozone decay or react with radical scavengers to form other products. The second order reaction of ozone with the ozone-hydroxide intermediate leads directly to products without formation of discrete short-lived radical intermediates. This simple model is consistent with the two term rate law for parallel first and second order reactions used by FIT80 to describe the time-concentration curves for the kinetic technique.

RESULTS AND DISCUSSION

The use of PAO as a direct reductant for ozone is based on the assumption that PAO (As(III)) is exclusively oxidized by ozone to phenylarsonic acid (As(V)) and that oxidation of the arsenic-carbon bond and the carbon-carbon double bonds are negligible. Since ozone has been used to digest organic arsenicals and since any PAO decomposition would lower residual ozone measurements, inorganic As(III) and PAO were compared by the kinetic technique. In order to attribute any inconsistency to PAO decomposition, both reductants were ozonized in acetate buffer pH 4.5. The pH was adjusted to 7 with 0.5 M sodium bicarbonate solution and the excess reductant back titrated with iodine to an amperometric endpoint.

Examination of the decay curves (Fig. 1)(or the rate constants) calculated by the FIT80 program reveals the similarity in behavior of PAO and inorganic As(III) as reductants for ozone. Although the PAO seems to be consistently high, the deviations are small enough to be within experimental error. Therefore, it is concluded that no noticeable decomposition of PAO occurs. Fig. 1 also illustrates the recurring and worrisome observation that in a kinetic comparison, one or two data points may fall significantly off the decay curve calculated by the FIT80 program. This observation cannot be neglected.

As shown in Table I, PAO and sodium thiosulfate are equivalent when used in the amperometric method. Here both function as reductants for iodine and are not directly involved in an ozone reaction. When these amperometric method results are compared with the results from As(III) direct oxidation, however, the ozone concentration determined by As(III) direct oxidation is almost nine percent low. Thus, when the ozone reductant changes from iodide ion to arsenic(III), clearly, a difference occurs.

Table 1. Amperometric Comparison of PAO and Sodium Thiosulfate *vs* the As(III) Direct Oxidation

Amperometric		As(III) Direct
PAO	$Na_2S_2O_3$	pH 4 - 4.5
mg/L O_3	mg/L O_3	mg/L O_3
14.5 ± 0.3	14.8 ± 0.3	13.5 ± 0.1

The DPD and the arsenic(III) back titration methods differ significantly in two ways: first, in the excess reagent present when ozone oxidizes iodide ion to iodine and second, in the iodine quantification. Since the DPD calibration curve is based on standard iodine, the iodine quantification should not be responsible for differences between the methods. In the arsenic(III) back titration, ozone could be reduced by iodide ion or by arsenic(III) although reduction by arsenic(III) occurs at a much slower rate than reduction by iodide ion. However, the formation of arsenic(V) by the direct oxidation by ozone or by the indirect oxidation through iodine, still maintains the stoichiometry of one ozone per arsenic(V). In the DPD method, ozone could oxidize iodide ion and oxidize DPD or the Wurster cation to the diimine cation. Any direct attack of ozone on the indicator would lead to low results. The decay curves in purified water determined by the DPD and the arsenic(III) back titration method (Fig. 2) show considerable scatter and in fact are good examples to illustrate the importance of the kinetic technique. If conclusions had to be based on single point comparisons, then the first, second, and third data point sets would lead to three different conclusions. Similar ozone decay curves in purified water spiked with 5 mg/L hydrogen peroxide prior to ozonation and in tap water show that scatter decreases as the radical scavenger concentration increases. Our results confirm the equivalence of the DPD method and the arsenic(III) back titration method.

The DPD method is not equivalent to the Indigo method as shown by the decay curves for ozone in purified water (Fig. 3). The DPD points are scattered and the ozone concentrations are low compared to the smooth Indigo plot. The critical differences between the methods are ozone reduction by a carbon-carbon double bond at pH 2 in the Indigo method and ozone reduction by iodide ion at pH 6.4 for the DPD method.

Iodate ion formation could be responsible for the low DPD titer. Iodate ion formation is also indicated in comparisons made using the Iodometric method at pH 3.5, 5, 7, 9 and the arsenic(III) direct oxidation method in bicarbonate ion solution at pH 7. Least squares analyses of plots of the iodometric method results on the x-axis vs the arsenic(III) direct oxidation results on the y-axis are shown in Table II.

Table 2. Comparison of Iodometric and Arsenic(III) Direct Oxidation Methods.

pH	Y-intercept	slope	correlation
3.5	0.95	1.00	0.9998
5.0	0.88	0.89	0.9979
7.0	0.95	0.70	0.9947
9.0	0.74	0.81	0.9986

The positive Y-intercept shows that the arsenic(III) direct oxidation method consistently gives low results. The slope shows the trend that the iodometric method gives lower results as the pH increases. This error is

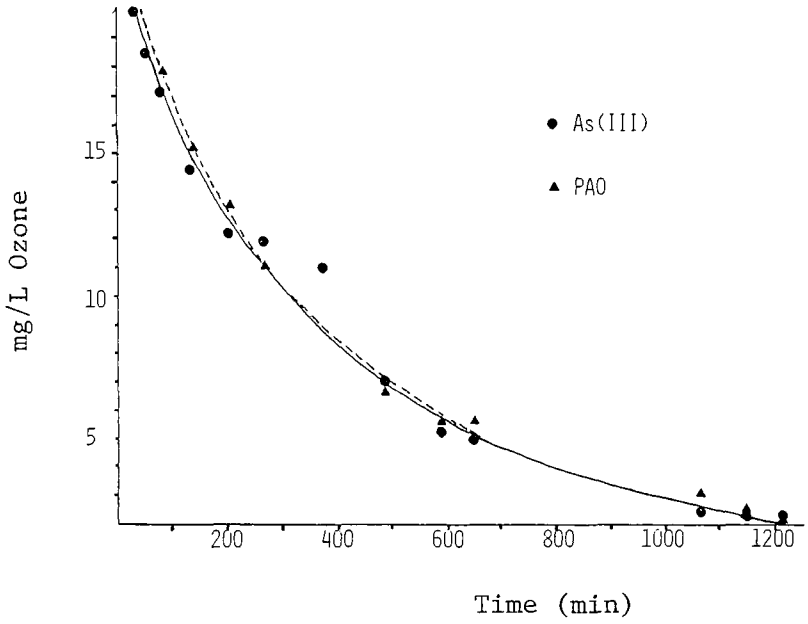

Figure 1. As(III) - PAO Comparison.

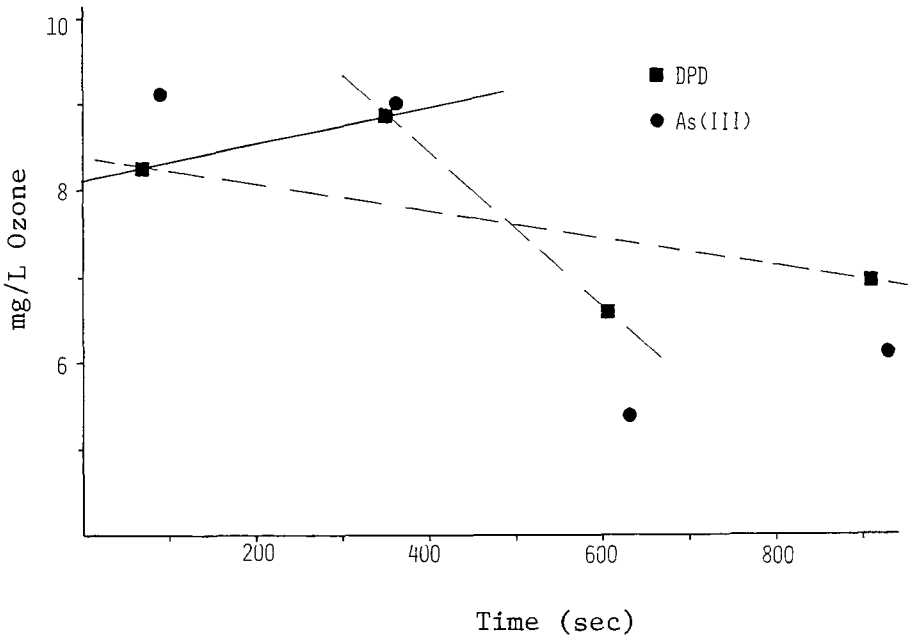

Figure 2. DPD vs. As(III) Back Titration in Purified Water.

consistent with iodate ion formation and with hydrogen peroxide formation. It would imply, however, that iodate ion is not quantitatively reconverted to iodine under the acidic titration conditions.

Iodate ion formation was measured for the ozone oxidation of 2% iodide ion solutions in 0.1 M phosphate buffers at pH 7.3, 6.8, 5.3, and 2.2. Immediately after the ozone injection, standard arsenic(III) was added and two aliquots were removed: one for iodate ion analysis and the other for ozone analysis by the back titration of excess arsenic(III). The back titration aliquots at pH 5.3 and 2.2 were brought to neutrality with sodium hydroxide. All iodate ion aliquots were immediately made strongly basic for differential pulse polarographic (DPP) analysis (16). The DPP is capable of detecting an iodate ion concentration as low as 1×10^{-8} M. This corresponds to 0.001 mg/L ozone taking into account the 3:1 ozone:iodate ion stoichiometry. As expected, iodate ion formation tends to increase with ozone concentration and with pH. The largest iodate ion concentration was found at pH 7.3 and correspond to 0.265 mg/L ozone (Table III). No iodate ion was detected at pH 2. The iodate ion concentrations are too low to explain the observed differences in the ozone decay curves traced by the arsenic(III) back titrations and by the Indigo method (Fig. 4). When the above experiment was repeated for pH 7.0 and pH 2.0 with arsenic(III) present in the iodide ion solution during the ozone addition, iodide ion concentrations corresponding to less than 0.024 mg/L ozone were found at pH 7.0 and no iodate ion was detected for pH 2.0.

Table 3. Iodate Ion Formation (in ozone equivalents) with Time(sec).

pH 2.2		pH 5.3		pH 6.8		pH 7.3	
sec	mg/L O_3	sec	mg/L O_3	sec	mg/L O_3	sec	mg/L O_3
None		90	0.176	74	0.147	26	0.173
		280	0.143	240	0.107	197	0.234
		510	None	500	0.091	420	0.100
		755	0.058	745	0.100	685	0.265
		1035	None	985	0.058	900	0.078
		1565	None	1555	None	1485	0.109

Hydrogen peroxide can be formed by the ozone-iodide ion redox reaction and by the decomposition of ozone in water. Its concentration, measured by DPP after one hour ozonation of 0.07 M phosphate buffers, varied with pH and with exposure to UV light (Ace Hanovia high pressure, mercury vapor lamp). The detection limit of 1×10^{-7} M corresponds to 0.0034 mg/L. In the absence of UV light, the hydrogen peroxide level increases with pH and in the presence of UV light the hydrogen peroxide level decreases with an increase in pH (Table IV). The hydrogen peroxide concentration varies dramatically under

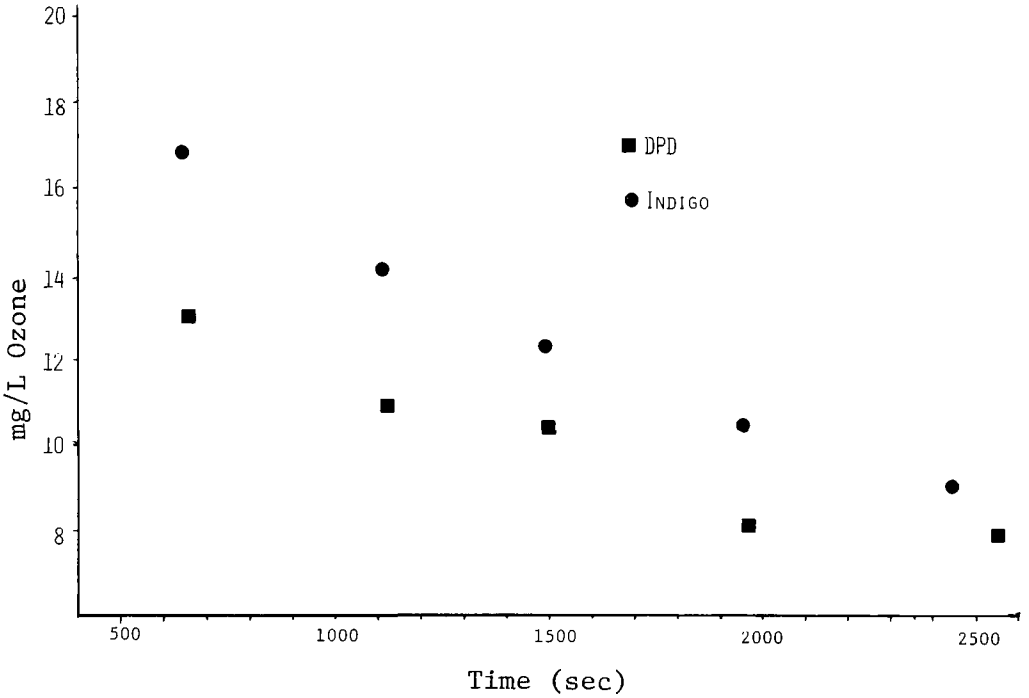

Figure 3. DPD vs. Indigo in Purified Water.

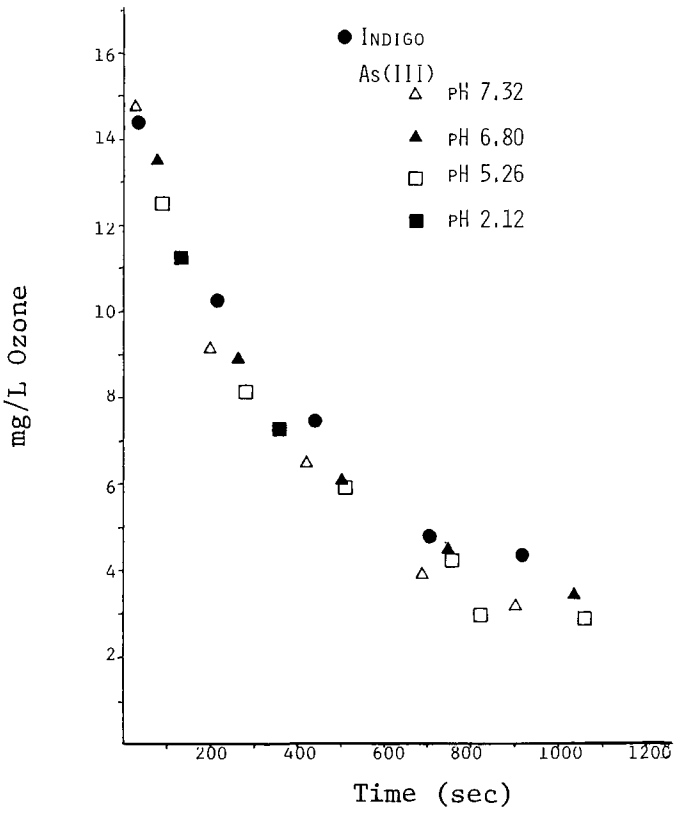

Figure 4. Indigo vs. As(III) Back Titration in Purified Water at pH 7.21.

Table 4. Hydrogen Peroxide Formation with Ozone Decay

pH	$[H_2O_2] \times 10^7$ M	$[H_2O_2] \times 10^7$ M with UV
2.1	4.8	
7.1	7.1	
12.0	10.0	
7.0	2.9	9.5
11.8	4.0	8.7
7.8		77.9
11.8		4.7

seemingly similar conditions and all the hydrogen peroxide titers are too small to account additively for method/pH differences. The hydrogen peroxide probably catalyze ozone decomposition since at 10^{-7} M hydrogen peroxide the hydroperoxyl anion is as important as hydroxide ion as a catalyst (11).

The scatter observed in kinetic comparisons of ozone decay in purified water is reduced by working with ozone in buffered purified water. Because this trend shows up with all methods, the significant difference must lie in the ozone solution itself. A comparison of the Indigo method, the arsenic(III) back titration and the arsenic(III) direct oxidation methods on ozone decay in purified water buffered to pH 6.7 with perchloric acid-phosphate mixtures shows the three methods to be alike.

When the Indigo method, the direct UV measurement, and the arsenic(III) direct oxidation method are compared on acidified ozone solutions undergoing minimal decay, they are also equivalent.

The ozone decay curves by the Indigo method, by direct UV measurement, and by arsenic(III) direct oxidation method for ozone in purified water buffered to pH 7.7 with a KH_2PO_4-NaOH mixture, however, show the Indigo method to give 10-15 percent higher residual ozone concentrations than the arsenic(III) direct oxidation method.

The kinetic technique has revealed differences among methods. These variations could be caused by:

1) the reaction of ozone or ozone decay products with the oxidized indicator (e.g. iodate ion formation).

2) the reaction of ozone decay products with the reductant or indicator.

3) the further decomposition of ozone prior to reaction with the reductant. This could be caused by the pH of the reductant solutions, solutes in the reductant solution, or by a relatively slow reaction between ozone and reductant.

The effect of hydrogen peroxide on the ozone titer was examined. Oxford tap water was ozonated, acidified for stabilization and analyzed for residual ozone by UV analysis, the Indigo method, the direct oxidation of As(III), and the back titration of As(III). Then, the residual ozone titer was determined with the addition of 3 mg/L H_2O_2 to the reductant solution immediately prior to ozone sampling. The results are given in Table V.

Table 5. Effect of Hydrogen Peroxide on Residual Ozone

	mg/L O_3 (No H_2O_2)	mg/L O_3 (H_2O_2)
UV	8.98 ± 0.05	8.79 ± 0.12
Indigo	9.52 ± 0.45	9.11 ± 0.05
As(III) direct	9.42 ± 0.23	9.32 ± 0.04
As(III) back	11.62 ± 0.20	12.22 ± 0.27

The arsenic(III) back titration method gives a residual ozone level 2.64 mg/L or 29% higher than the UV method for analyses on the acid stabilized ozone solution. Hydrogen peroxide increased this error to 3.43 mg/L or 39% for the arsenic(III) back titration method. The Indigo and arsenic(III) direct oxidation titers agreed within 6% of the UV titers.

Ozone decomposition prior to reaction with the reductant is most likely to complicate the arsenic(III) direct oxidation method due to the relatively slow reduction of ozone by arsenic(III). Iodide ion reduces ozone in a virtually diffusion controlled reaction. The Indigo reductant solution is buffered at pH 2 minimizing ozone decay prior to attack of the carbon-carbon bond. In fact, when an arsenic(III) back titration reductant solution is dosed with concentrated ozone, the amber iodine color appears immediately and then fades as the iodine reacts with the arsenic(III). This clearly demonstrates that the ozone reacts faster with iodide ion than with arsenic(III).

When a dilute ozone solution undergoing minimal decay was directly reduced by arsenic(III) in acetate buffer at pH 4.5 and in phosphate buffer at pH 6.8, both arsenic determined decay curves were scattered compared to the UV curve. The acetate curve had wide deviations from the calculated curve.

The residual ozone concentrations for three steady state solutions were also determined by these methods and compared with the concentrations determined by the direct oxidation of arsenic(III) in unbuffered solution at pH 7. (Table VI). The ozone titers do not consistently increase with a decrease in pH and the ozone titers determined in unbuffered arsenic(III) are low relative to buffered solutions and the UV method. The UV method provides a convenient and rapid reference method when working with ozone solutions free from other absorbing materials.

Table 6. Buffer Effect on Direct Arsenic(III) Method.

UV	Acetate pH 4.5	Phosphate pH 6.8	No Buffer pH 7
6.62	6.13	6.42	5.92
7.00	7.04	6.64	6.34
9.06	8.91	8.81	8.49

If ozone decay prior to reduction by arsenic(III) causes low ozone titers, then any change in the reductant medium to slow decay should increase the ozone titer. As generally accepted and as illustrated in Fig. 5, the rate of ozone decay decreases with decreasing pH. These rates were determined by direct UV measurement in 0.1 M phosphate buffers ranging in pH from 9.4 to 5.9.

Ozone decay, however, is a complex function of pH and solutes. The relative rates for ozone decay in solutions at pH 7.0 - 7.2 containing varying concentrations of phosphate and carbonate ions are listed in Table VII.

The effect of these anions is enormous. The half-life ($t_{1/2}$) for purified water containing no phosphate ion or carbonate ion and adjusted to pH 7.0 with sodium hydroxide, is less than 500 sec. The $t_{1/2}$ in 0.1 M phosphate, 0.1 M carbonate is longer than 12 hours.

Table 7. Relative Rates for Ozone Decay (pH 7.0 - 7.2)

		[carbonate]		
		0.0 M	0.01 M	0.1 M
[phosphate]	0.0 M	200	2.2	2.4
	0.1 M	38	2.6	2.0
	0.25 M	22	6.0	5.3
	0.50 M	46	10	8.4

The Delta amperometric membrane electrode measures ozone concentration *in situ* and should not be influenced by ozone instability or overoxidation. A teflon membrane selectively transports gaseous molecules like ozone to the cathode and prevents transfer of polar species and ions. The electrode operates at a potential where only very strong oxidants are reduced. Thus, the Delta electrode promises the ideal combination of chemical selectivity and the capability for continuous monitoring. To be practical, the electrode must be easily calibrated, must remain calibrated for a reasonable length of time, perhaps a minimum of one working day, and the calibration must be valid over the working range of the electrode, 0-10 mg/L. Direct UV measurement of ozone was used to calibrate the electrode.

The kinetic comparison of ozone decomposition by direct UV measurement, the Indigo method, and the Delta electrode shown in Fig. 6, illustrates two recurring problems. First, ozone decay determined by the Delta electrode followed its own rate law. Second, the electrode rarely maintained calibration on switching from one solution to another. The initial points measured by the electrode of Fig. 6 are low for this reason.

The change in rate law was traced to the lack of linear response of the electrode over a large concentration range. The electrode was calibrated by UV analyses on an acidified ozone solution. A volume of this solution was removed and replaced with an equal volume of acidified water and the electrode response and UV absorbance measured. This dilution was repeated several times to obtain stable ozone solutions of varying concentration within a 2.5 hour period.

The results in Table VIII show that as the ozone concentration decreases relative to the calibration concentration, the error increases. For electrode 1, originally purchased from Delta Scientific, a 76 percent error is observed over a concentration range of 4.8 mg/L O_3. The error drops to 34 percent for a concentration change of 3.0 mg/L O_3. Electrode 2, a later model generously supplied by Delta, measured residual ozone in the range 4.0 - 1.9 mg/L with a 2.5 percent error. The error increased to 23 percent at 0.5 mg/L.

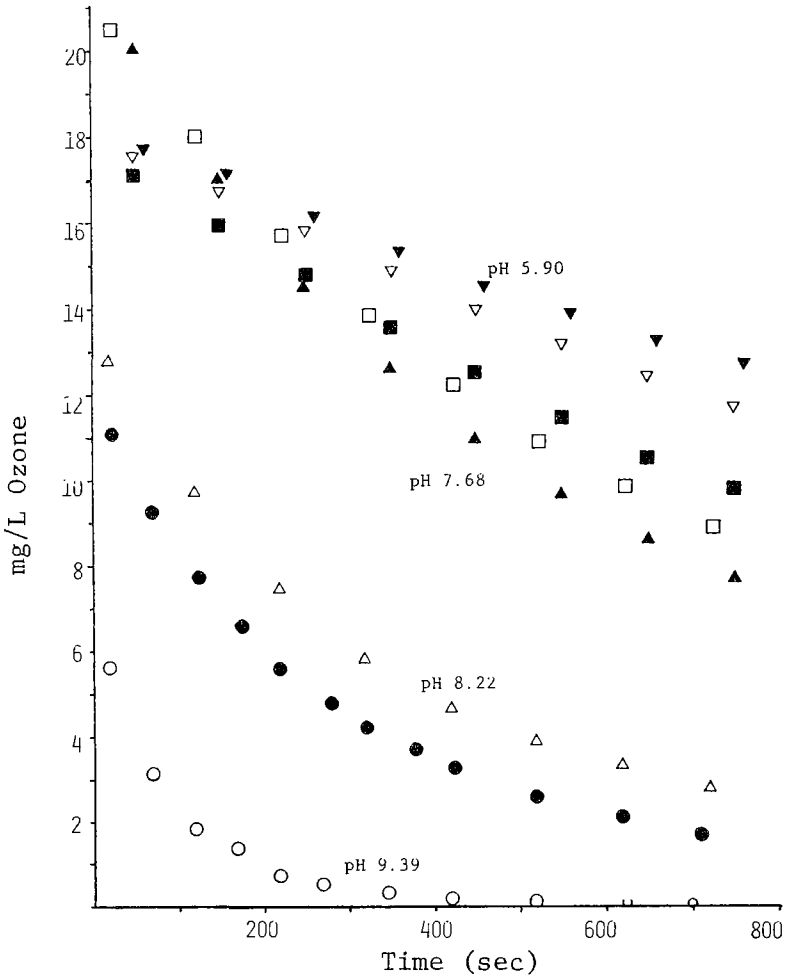

Figure 5. Effect of pH on Ozone Decay in Purified Water with Phosphate Buffer.

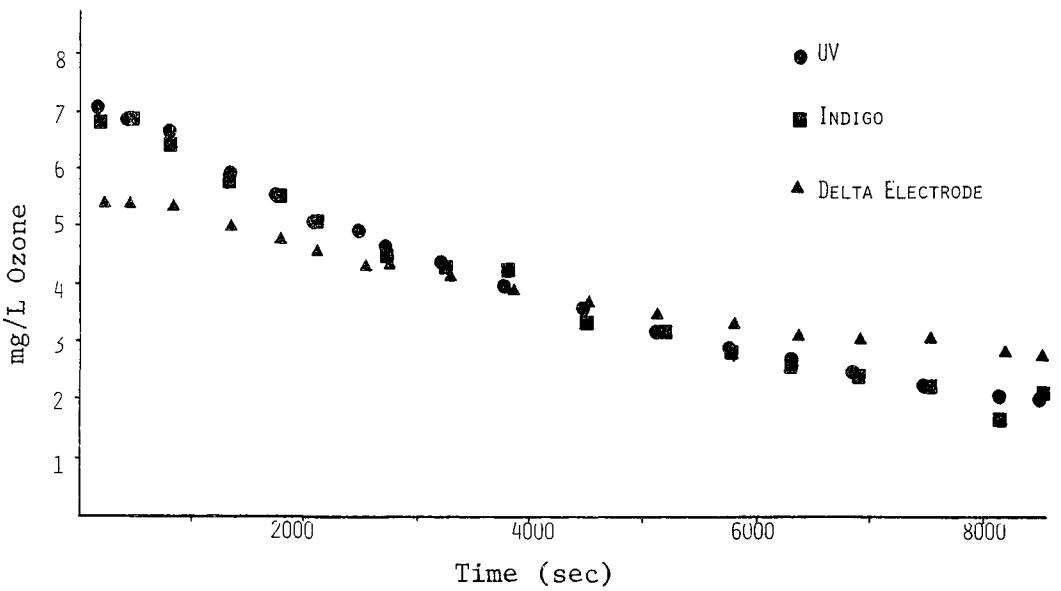

Figure 6. Comparison of Ozone Decay in Purified Water.

Table 8. Dilution Experiments for Linear Response

UV mg/L O_3	Electrode 1 mg/L O_3	% Error	UV mg/L O_3	Electrode 2 mg/L O_3	% Error
7.31	7.38		4.03	4.00	-0.7
5.77	6.60	14.4	3.80	3.75	-1.2
4.33	5.81	34.2	2.65	2.55	-3.5
3.35	5.13	53.1	1.86	1.91	2.5
2.54	4.47	76.0	1.30	1.46	12.8
			0.84	0.99	17.7
			0.54	0.67	22.7
			0.30	0.50	66.4
			0.16	0.33	107.6

In a kinetic comparison, Oxford waste water treatment plant effluent was ozonated for 10 minutes and the residual ozone concentrations were determined over the next 15 minutes by the Indigo method, arsenic(III) back titration method, arsenic(III) direct oxidation method, and the Delta electrode (electrode 2). The waste water was then reozonated for 15 minutes and the decay followed as above. Results are shown in Fig. 7. The decay curve traced by electrode 2 differs from the other methods and the electrode again appears to have lost calibration between runs. This instability and unpredictability of the Delta electrode clearly emphasize the necessity for recalibration for each run.

Notice that the second decay curve traced by the Indigo method and the arsenic(III) back titration and arsenic(III) direct oxidation methods is very similar to the first curve. This was a consistent observation in sequential ozonation experiments with Oxford Sewage Treatment Plant effluent.

For example, the rate of ozone decay in waste water was determined following an initial 30 minute ozone treatment. A two hour ozone treatment followed the next day. On the third day, a 30 minute ozone treatment was repeated and the ozone decay followed. The decay curves were superimposable. The ozone decomposition rate was also similar after each of five minute consecutive ozone treatments on Oxford Sewage Treatment Plant effluent (Fig. 8). These experiments imply that once ozone satisfies the initial ozone demand of waste water, residual ozone levels are controlled by ozone self-decomposition and not by direct reaction with impurities. Making the reasonable assumption that sufficient radical scavengers are present to quench the first order decay process, the residual ozone decay should be pH controlled.

The pH controlled decay rate can be measured by determining the ozone $t_{1/2}$ at a given pH with increasing concentrations of radical scavengers. The $t_{1/2}$ should reach a limiting value for each pH. With knowledge of the pH of the ozonated effluent and also the maximum $t_{1/2}$ at this pH, demonstration of the presence of ozone after a calculated time could be sufficient to assure disinfection.

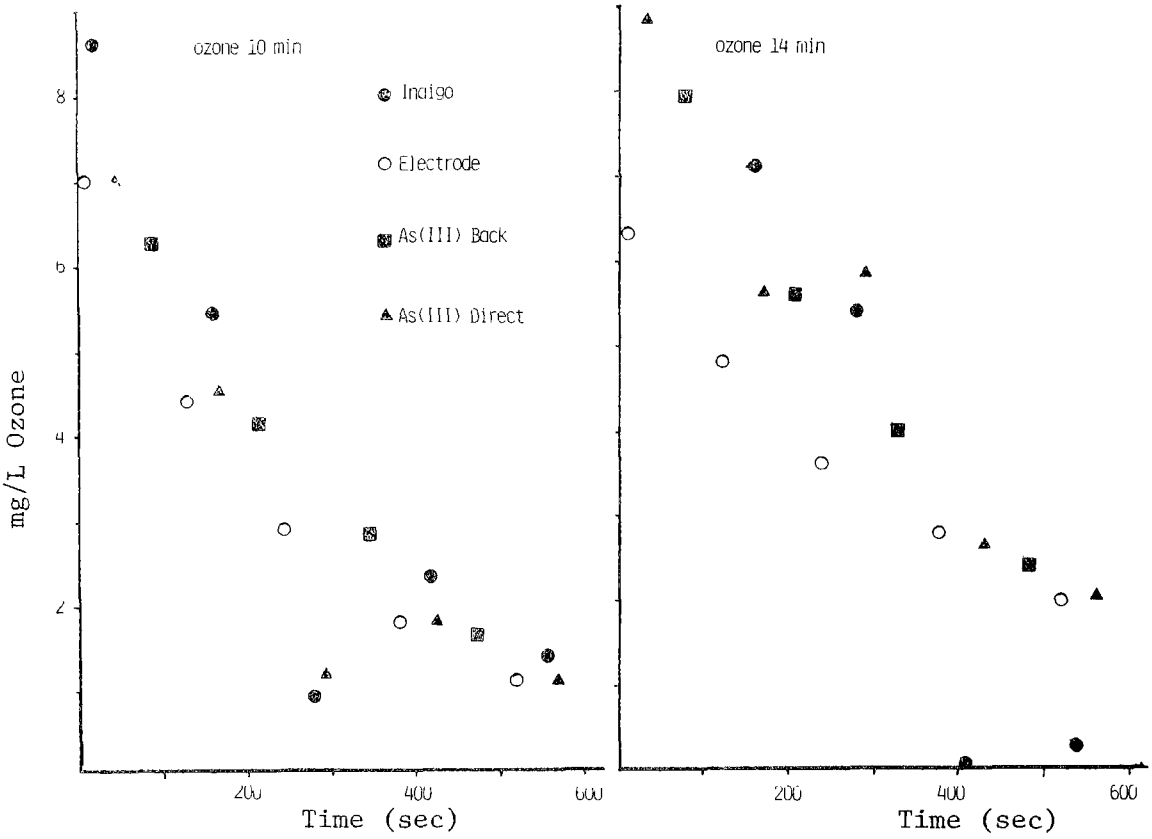

Figure 7. Ozone Decay in Waste Water.

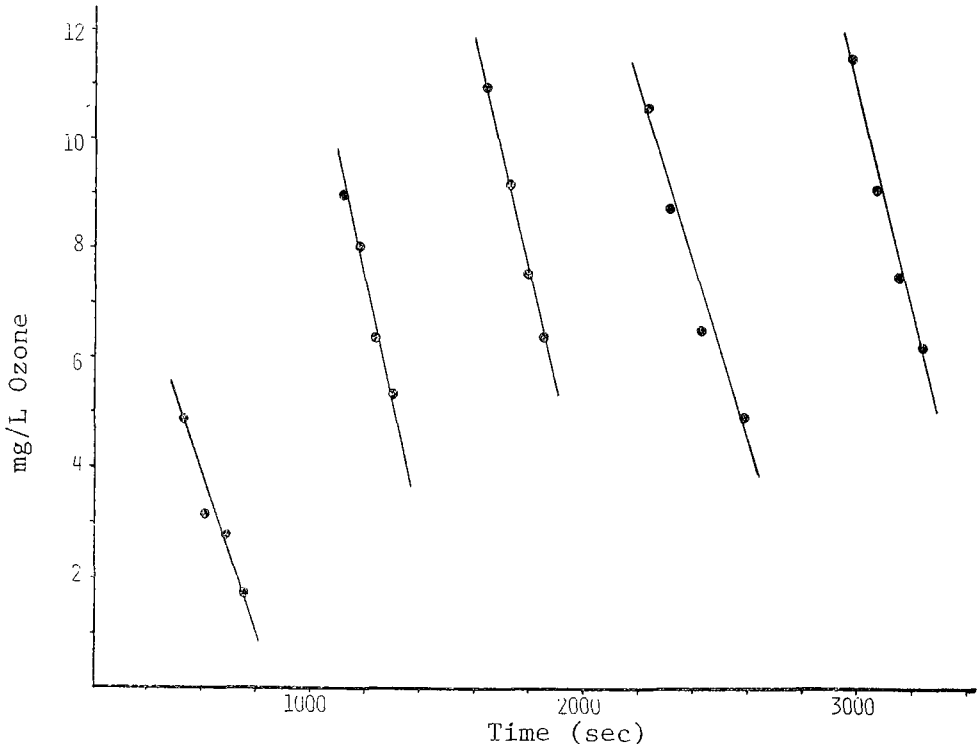

Figure 8. Ozone Decay in Waste Water Following Sequential Ozonation.

For water and waste water treatment, continuous monitoring of residual ozone is ideal. The direct measurement of the absorbance of aqueous ozone at 259 nm is the most straightforward and simplest method. However, in waste waters, many impurities absorb in this region producing a large background absorption. A membrane electrode promises continuous, specific ozone analysis. Unfortunately the technology is not yet available to provide the requisite reliability and stability.

All volumetric methods occassionally give a point 30-50 percent removed from that calculated on an otherwise smooth decay curve. This makes a single point analysis for residual ozone untrustworthy. The titer or absorbance of a solution of reductant or indicator should be sufficiently stable to allow convenient laboratory analyses and ideally to allow field collection with later laboratory analysis. With the DPD method, the ozone titer changed rapidly with time for ozone in purified water and for ozone solutions with added hydrogen peroxide. The arsenic(III) back titration titer steadily increased for ozone solutions with added hydrogen peroxide (Fig. 9). The ozone titer by the amperometric method with excess sodium thiosulfate increased 4 percent in 9 minutes with ozone in purified water. The ozone titer determined by the arsenic(III) direct oxidation method and the Indigo method varied less than 3 percent over 3 hours even with added hydrogen peroxide. The arsenic(III) solutions are stable standard solutions readily prepared by weight. Stock Indigo trisulfonate would need replacement at least every ten weeks. Calibration is time consuming. These problems could be avoided if higher purity dye were readily available and calibration could be based on weight. The arsenic(III) direct oxidation method shows variable and significant blanks.

The ozone titers differed among methods only when changes in the ozone-reductant reaction were involved. Conditions which reduce ozone decay prior to reaction with reductant, reduced the scatter observed within a single method and reduced the differences observed among the analytical methods. This is understandable since direct oxidation by the ozone molecule is selective and stoichiometric. Oxidations by ozone decay products such as the hydroxyl radical are non-selective and non-stoichiometric.

The Indigo method minimizes ozone decay by operating at pH 2. Buffers which slow ozone decay increase the ozone concentration determined by direct arsenic(III) oxidation. Multiple analyses on waste water show few differences because waste water impurities scavenge the hydroxyl radical and prevent its reaction with reductant and its catalysis of ozone decay.

The experiments reported here also clearly demonstrate that the purging technique--widely used to eliminate *in situ* interferences--is unreliable because of ozone decomposition during the purge and readsorption steps.

In conclusion, we have found the Indigo method and the arsenic(III) direct titration method to be the most reliable. Additional comparisons, along with the recommended detailed experimental techniques will be published separately.

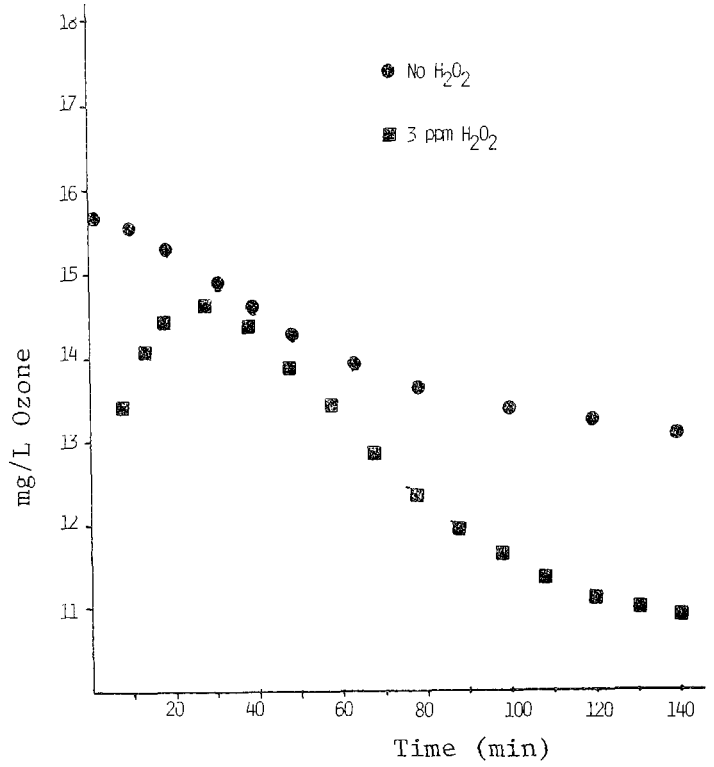

Figure 9a. Stability of DPD.

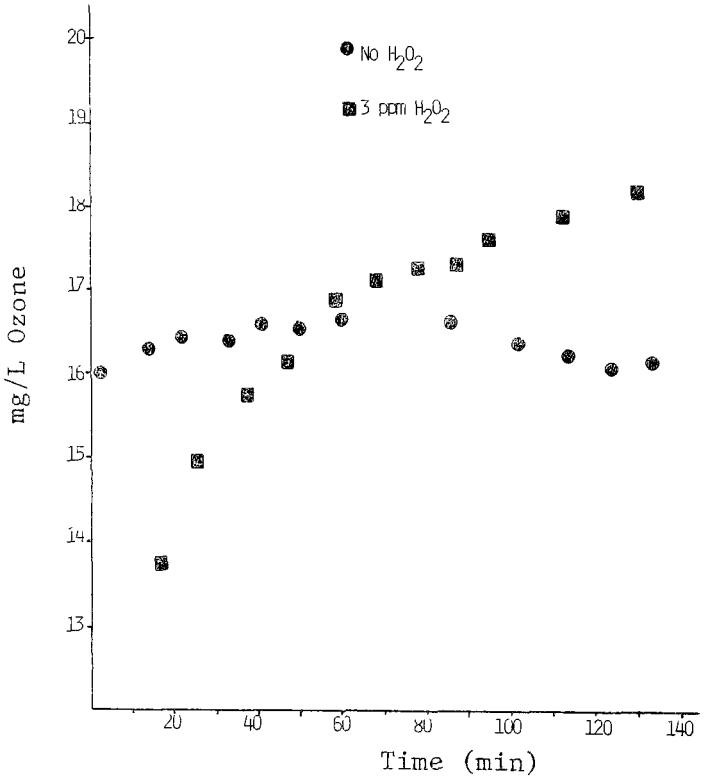

Figure 9b. Stability of As(III) back titration.

ACKNOWLEDGEMENTS

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8. CONTROL OF OZONE DISINFECTION BY EXHAUST GAS MONITORING

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ABSTRACT

The on-site manufacture of ozone is energy intensive, because it is generated from electric current. Any advance that is made to help reduce the cost of generating ozone will accelerate its acceptance as a viable alternative to chlorine.

In the field of chlorination, dose is routinely controlled by a combination of a flow proportional signal and a chlorine residual signal. No such control mechanism exists for ozone, primarily because of the difficulty in accurately measuring true ozone residual. Typically, ozone dose is controlled by turning on and off entire generators as flow increases or decreases. This is wasteful and grossly inaccurate, and consequently operating costs still remain relatively high.

This paper discusses pilot plant data gathered from six different treatment plant effluents indicating a reasonably good correlation between the concentration of ozone in the exhaust gas from the contactor and the total and fecal coliform levels in the final effluent. The advantages of this approach are: (1) true ozone is being measured (not total residual oxidant); (2) the reaction is instantaneous and extremely simple to conduct; (3) it is easily automated; (4) it is useful on a wide variety of secondary effluents; (5) it is not subject to interferences; and (6) it is not adversely affected by sudden shifts in effluent quality. There is one underlying restriction that must be observed if the method is to be successfully applied: the gas-to-liquid flow ratio must always remain constant. Thus, gas flow must be paced to liquid flow by a flow proportional signal. If this restriction is met, then dose can be controlled by monitoring ozone in the exhaust gas and automatically signalling changes in the power to the generator.

INTRODUCTION

The thrust of the Environmental Protection Agency's in-house research effort on ozone has been directed towards optimizing ozone contacting to achieve the desired bacteriological quality with the least amount of ozone applied. The reason is simply that ozone, which must be generated on-site, is energy intensive and, therefore, any effort at reducing the use of ozone will result in a substantial savings in operating costs.

Having established the bubble diffuser to be the most efficient contactor of five generic types tested (8,9,10), we deemed it necessary to determine how best to monitor and control applied dose as demand and flow fluctuated. We hypothesized that there was no reason why a compound loop control mechanism analogous to that used in the chlorination field could not be developed for ozone. Such a control mechanism would involve a flow signal, which would increase the rate of addition of ozone to the effluent, and a demand signal, which would increase the concentration of ozone applied. The problem confronting us was the lack of ability to measure true ozone residual in the process water. Conventional measurement techniques do not differentiate ozone from other oxidants, and without such differentiation a control mechanism would be impossible.

In a brief summary of the literature we found very little research has been done in this area. In an EPA report published in August 1978 (4), Miller et al. conducted a global survey of all treatment plants (mostly drinking water) using ozone for disinfection and other uses. They reported that French plants incorporate a closed loop control system by which the residual ozone level in the ozonated water is used to control the amount of ozone supplied to maintain that residual. In a symposium sponsored by the International Ozone Association in 1975 (6), several speakers discussed the need for measuring ozone in both the gas phase and the liquid phase. Nebel and Forde (5), in discussing the principles of industrial and municipal odor control with ozone, demonstrated that, by installing an ozone meter in the exhaust gas stack coming off the contactor, it was possible to detect small concentrations of ozone in the exhaust gas stream and then feed signals to the generator to vary the ozone input to the contactor. Trussell (7) expressed a desire to be able to monitor ozone in the exhaust gas from the contactor to determine the efficiency of consumption and the amount of ozone needed to be destroyed before discharge to the ambient atmosphere. His primary interest, however, was measuring ozone in water for dose control purposes.

From the foregoing it is clear that there are two variables that can be measured for the purpose of establishing an automatic, real time monitoring tool. They are: ozone residual in the liquid stream and ozone concentration in the exhaust gas. Both of these variables represent unused ozone, the former being that amount of ozone transferred to the liquid but not reacted or decomposed, the latter being that amount of ozone not transferred to the liquid. The magnitude of both will depend upon the demand of the liquid, the ozone transfer efficiency of the contactor, the concentration of ozone in the inlet gas, and the gas flow rate relative to the liquid flow rate. This paper will discuss measurement of ozone residual and exhaust gas relative to coliform destruction and attempt to demonstrate the superiority and reliability of exhaust gas measurements as a dose monitoring and control technique.

MATERIALS AND METHODS

Sources of Secondary Effluent

Secondary effluent was obtained from six different sources: (1) the

Fairfield Wastewater Treatment Plant, Fairfield, Ohio; (2) the Indian Creek Wastewater Treatment Plant, Cincinnati, Ohio; (3) the Loveland Wastewater Treatment Plant, Loveland, Ohio; (4) the Mill Creek Wastewater Treatment Plant, Cincinnati, Ohio; (5) the Muddy Creek Wastewater Treatment Plant, Cincinnati, Ohio; and (6) the Sycamore Wastewater Treatment Plant, Cincinnati, Ohio. Fairfield, Mill Creek, and Muddy Creek are conventional activated sludge treatment plants. Loveland and Sycamore are contact stabilization plants and Indian Creek uses rotating biological contactors to treat municipal wastewater. Of the six treatment plants, five receive raw wastewater of municipal origin. The 6th, Mill Creek, receives wastewater with a high concentration of industrial wastes (about 50 percent of the organic loading by weight).

Approximately 20 m^3 of a given effluent was collected in a tank truck on the day of an experiment or the day before and transported to the U.S. EPA Test and Evaluation Facility, located adjacent to the Mill Creek treatment plant. When an effluent was collected the day before an experiment, it was recirculated in the tank truck for a minimum of one hour before initiation of the experiment.

Ozone Generation

Ozone was generated from oxygen in a plate type corona discharge generator (Computerized Pollution Abatement Corporation Model OZ-180G). Oxygen flow to the ozonator was maintained at a constant 33 L/min. Liquid flow was a constant 75 L/min, resulting in a gas-to-liquid flow ratio of 0.44. Changes in the applied dose were accomplished by varying the concentration of ozone in the gas flow (i.e., by increasing or decreasing the power applied to the generator).

Ozone Contactor

The ozone contactor used was a bubble diffuser with 3 columns connected in series. Its design and operating characteristics are fully described elsewhere (9).

Sampling

All effluent samples were grab samples and were analyzed for total and soluble chemical oxygen demand (TCOD and SCOD), total organic carbon (TOC), total suspended solids (TSS), and turbidity by Standard Methods (1). Total Kjeldahl nitrogen (TKN), ammonium nitrogen (NH_4^+-N), and nitrite-nitrogen (NO_2^--N) were measured according to Methods for Chemical Analysis of Wastes (3) and nitrate-nitrogen (NO_3^--N) according to Kamphake, Hannah, and Cohen (2). Ozone concentration in the inlet and exhaust gases was periodically determined iodometrically (1) and continuously by ultraviolet adsorption analyzers (Dasibi Environmental Corporation, Glendale, California). Ozone residual (as total residual oxidant) was measured by the reverse titration standard iodometric method for chlorine residual (1) using the amperometric end point.

Samples collected for bacteriological analysis were assayed for total and fecal coliforms by the standard Membrane Filtration (MF) method (1), using 0.45 μm GN-6 membrane filters (Gelman Instrument Company). All chemical and bacteriological samples were collected from a sample tap located at the bottom of the contactor's third column.

Procedure

The approach chosen was based on the hypothesis that an empirical relationship found previously predicts, with a reasonable degree of accuracy, the total coliform density of a municipal wastewater effluent following treatment with ozone. The relationship is:

$$\log_{10} TC \quad 4.38 - 4.58 \ (\log_{10} T) + 0.040 \ TCOD \qquad (a)$$

where TC total coliforms/100 ml after ozonation, and

T = ozone transferred, mg/L

If one knows the TCOD of an effluent, one may be able to predict the final coliform density at various levels of absorbed ozone. To test this hypothesis we chose six local treatment plants and grouped them according to the mean TCOD concentrations in their effluents. Before the ozonation experiments were conducted, nine effluent samples from each of the treatment plants were collected over a 3-week period and measured for TCOD. The mean TCOD levels were then compared and like plants were grouped accordingly. Three groupings resulted: (i) a low TCOD group (three treatment plants), (ii) a medium TCOD group (two treatment plants), and (iii) a high TCOD group (one treatment plant).

By rearranging equation (a) and solving for T at 5 different total coliform levels, we computed five absorbed ozone levels for each of the three groupings. The applied dose levels needed to achieve the five absorbed ozone levels were calculated by assuming an average transfer efficiency of 90 percent (previous data had indicated that 90 percent transfer efficiencies were possible in the bubble diffuser contactor, with oxygen as the feed gas, at gas-to-liquid flow ratios of ≤ 0.5). The resulting five relative dose levels for each of the treatment plant groupings are presented in Table 1. The five applied doses (labeled A through E) are relative in the sense that each one theoretically yields equivalent coliform densities consistent with effluent quality as long as dose is varied by changing the ozone concentration in the inlet gas stream and maintaining a constant gas-to-liquid flow ratio. If the empirical model were a good predictor, dose A would yield the same total coliform density in all effluents, dose B would yield a lower number in all effluents, and so on.

By grouping the plant effluents in the above fashion, it would facilitate further analysis of factors affecting ozone disinfection, should there be a significant difference between effluent sources with respect to post-ozonation

coliform densities. To minimize any trend in wastewater effluent quality in a given day, the design was balanced so that each dose level occurred the same number of times at each time of day. The five sets of observations were taken over a period of approximately four hours. Plant effluents were collected five times from each of the six plants according to a randomized collection schedule.

Table 1. Relative Dose Scheme for the Three Treatment Plant Groupings Used in the Study

Relative dose	Calculated Resulting Log_{10} Total Coliform Density	Actual dose, mg/L group i[a]	group ii[b]	group iii[c]
A	5.0	1.3	2.3	5.1
B	4.0	3.3	5.1	10.0
C	3.0	5.0	7.6	15.0
D	2.0	6.7	9.8	20.5
E	1.0	8.6	12.4	25.1

[a]Effluent Sources Indian Creek Plant, Loveland Plant, and Muddy Creek Plant

[b]Effluent Sources Fairfield Plant and Sycamore Plant

[c]Effluent Source Mill Creek Plant

RESULTS

Effluent Quality

Table 2 summarizes the physical-chemical and bacteriological characteristics of the effluent sources prior to ozonation. The Mill Creek effluent contained substantially higher amounts of TCOD and TOC than any of the other effluents. This was due to the high proportion of industrial components present in the raw wastewater entering the plant.

Effect of Absorbed Ozone on Coliform Numbers

We have shown previously (9,10) that total and fecal coliform levels in a given effluent can be predicted if the demand properties of the effluent and the absorbed ozone dose are known. Figure 1 is a graph of the log total and fecal coliform numbers in the six effluents as a function of the amount of ozone transferred to the effluents. The data were averaged over the five replicate runs. Clearly, dose responses in five of the six effluents were

similar. The coliform decline in the sixth effluent, the Mill Creek Treatment Plant, deviated significantly from the others, reflecting the substantially higher demand characteristics of that effluent and indicating that such responses are not universally predictable or applicable. Thus, attempts to monitor disinfection efficiency by measuring ozone transfer may lead to erroneous results.

Table 2. Secondary Effluent Characterization of the Six Treatment Plants Prior to Ozonation

Parameter	Effluent Source					
	Fairfield mean (range)	Indian Creek mean (range)	Loveland mean (range)	Muddy Creek mean (range)	Mill Creek mean (range)	Sycamore mean (range)
TCOD, mg/l	39 (63-22)	26 (48-16)	39 (57-30)	29 (45-16)	74 (103-53)	38 (56-26)
TOC, mg/l	5.6 (11.1-1.3)	5.2 (9.0-2.2)	6.8 (13.8-3.7)	5.0 (9.0-1.2)	19.6 (29.9-13.9)	8.3 (12.7-4.6)
TSS, mg/l	4.1 (13.2-1.4)	9.2 (36.8-1.8)	8.3 (34.0-3.7)	3.8 (9.2-0.4)	11.5 (25.0-2.8)	8.6 (18.8-3.0)
TKN, mg/l	5.3 (12.3-1.9)	1.7 (7.2-0.5)	13.8 (17.8-9.8)	1.5 (4.5-0.6)	18.7 (31.0-8.1)	7.5 (17.6-4.4)
NH_4^+-N, mg/l	4.3 (12.4-0.1)	0.3 (0.7-<.1)	11.7 (18.6-6.4)	0.8 (3.6-<.1)	19.3 (31.8-7.3)	5.6 (7.3-4.5)
NO_2^--N, mg/l	0.6 (1.8-<.1)	0.2 (1.1-<.1)	0.5 (1.3-0.1)	0-2 (0.9-<.1)	0.6 (2.6-<.1)	0.5 (1.1-<.1)
NO_3^--N, mg/l	7.5 (13.8-4.3)	7.5 (14.1-3.2)	0.1 (0.2-<.1)	6.0 (8.8-3.0)	0.2 (0.7-<.1)	2.5 (5.8-<.1)
Turbidity, JTU	2.5 (5.3-1.0)	1.7 (7.2-0.5)	13.8 (17.8-9.8)	1.5 (4.5-0.6)	18.7 (31.0-8.1)	7.5 (17.6-4.4)
pH	(8.3-7.3)	(8.4-7.9)	(8.2-7.4)	(8.1-7.3)	(7.9-7.4)	(7.9-7.2)
$\log_{10}$TC/100 ml	5.41 (6.51-4.28)	5.59 (6.11-5.08)	6.56 (7.27-6.16)	5.41 (6.33-4.65)	5.72 (6.84-4.68)	6.43 (6.86-6.08)
$\log_{10}$FC/100 ml	4.69 (5.60-3.30)	4.43 (5.00-3.76)	5.80 (6.36-5.15)	4.59 (5.21-3.79)	4.75 (6.18-3.85)	5.67 (5.94-5.46)

Effect of Ozone Residual on Coliform Numbers

Figure 2 is a plot of total and fecal coliform numbers in the six effluents as a function of ozone residual in the liquid. Again, the data were averaged over the 5 replicate runs. Response patterns are similar to those shown in Figure 1, although the deviations in the Mill Creek effluent are not as great. Use of ozone residual in the liquid as a real time monitoring

tool may be appropriate if it is expected that fluctuations in wastewater quality are relatively minor. However, as will be shown below, even if the quality has not changed significantly, the presence of compounds or substances which interfere with the measuring technique may argue against use of ozone residual as the primary control technology.

Effect of Exhaust Gas Ozone on Coliform Numbers

Results of plotting log coliform numbers as a function of ozone concentration in the exhaust gas are presented in Figure 3. Clearly, the data from all six treatment plants fit the indicated curve quite well, suggesting strongly that measurement of ozone in the exhaust gas from the contactor may be an excellent control strategy. There is a very important restriction, however, that must be incorporated when using this strategy: the gas-to-liquid flow ratio must be held constant at all times. The reason is that the mass transfer efficiency of the contactor decreases markedly as the gas flow increases (9). Thus, an increase in gas flow relative to liquid flow may result in a higher exhaust gas ozone concentration without any corresponding increase in mass transfer or coliform reduction. In contrast, by maintaining a constant gas-to-liquid ratio and varying the power (or frequency) to the generator, the increase in ozone transferred will be almost in direct proportion to the higher ozone concentration in the inlet gas, up to the limit defined by Henry's Law. A higher exhaust gas level will occur also, but only after more ozone has been transferred to the water. Thus, an increase in coliform reduction will necessarily take place.

Interferences in Residual Measurement

To demonstrate further the inferiority of monitoring ozone residual in the liquid to control disinfection, all 125 data points from five of the six treatment plant effluents (the Mill Creek effluent was excluded) were used to plot log total coliforms in the effluent as a function of ozone residual. Results are shown in Figure 4. Although a clear trend is noted, 4 data points (represented by open squares) stand out as significant outlyers. Upon studying carefully the computer printout of the raw data, we discovered that these high coliform and ozone residual values all occurred in the same run with the same effluent (i.e., the Muddy Creek effluent). The only unusual feature of this effluent on that date was a high concentration of manganese, approximately 0.8 mg/L. This amount exceeded the normal levels measured on all other days by more than 25-fold. According to the 15th edition of Standard Methods (1), oxidized forms of manganese give positive interferences in all methods for total available chlorine (the method we were using for ozone), including amperometric titration. Thus, if an operator is measuring ozone residual by standard, state-of-the-art techniques, he could be misled on days when positive interferences are present unexpectedly in the effluent. On such days coliform discharge limitations could be exceeded.

We made a similar plot of log total coliform numbers versus exhaust gas ozone (Figure 5). The Mill Creek effluent is included in this plot. Clearly, no outlyers are observable on the graph because true ozone is being measured

in the gas phase. This again confirms the usefulness and reliability of exhaust gas monitoring for control of ozone disinfection at a secondary treatment plant.

DISCUSSION

In this paper we have demonstrated empirically that disinfection with ozone can be controlled by monitoring the exhaust gas ozone concentration exiting the contactor. This method is more reliable than measuring dissolved ozone because of the inherent difficulties and inadequacies of state-of-the-art dissolved residual techniques. The advantages of measuring exhaust gas ozone are summarized as follows: (1) true ozone is being measured, free of interferences; (2) ozone demand of the effluent and transfer efficiency of the contactor are automatically accounted for in one measurement; (3) the method is easily automated; (4) instruments are already available on the market for measuring ozone in the gas phase with accuracy, precision, and low level sensitivity; and (5) ozone is more stable in the gaseous phase than in the liquid phase, and consequently the operator does not have to concern himself with dissipation of the ozone from the time it leaves the contactor to the time it arrives at the analyzer.

It must be emphasized that exhaust gas monitoring is only applicable if the gas-to-liquid flow ratio is held constant. The control loop is then envisioned as follows: (1) a flow proportional measurement signals a change in the gas flow from the ozone generator to the contactor as liquid flow changes, thereby keeping the ratio constant; (2) as ozone concentration in the exhaust gas changes either as a result of a change in demand of the effluent or a change in flow conditions, a signal is sent to the ozone generator to change the power or frequency input accordingly. Thus, disinfection is controlled easily, reliably, and with confidence. The effect this control strategy has on the cost of ozone production has yet to be evaluated. The data presented in this paper were obtained using a plug flow bubble diffuser contactor. There is no reason to believe, however, that the control strategy would not be applicable to other types of ozone contactors as well.

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We thank Messrs. Harold P. Clark and Harld L. Sparks for enumeration of coliforms in all samples. Ms. Rebecca McCutcheon and Mr. John Rogers assisted in sampling, performance of ozone analyses, and operation of the ozone disinfection equipment. Chemical analyses were conducted by the Waste Identification and Analysis Section, Wastewater Research Division, Municipal Environmental Research Laboratory, U.S. EPA, Cincinnati, Ohio.

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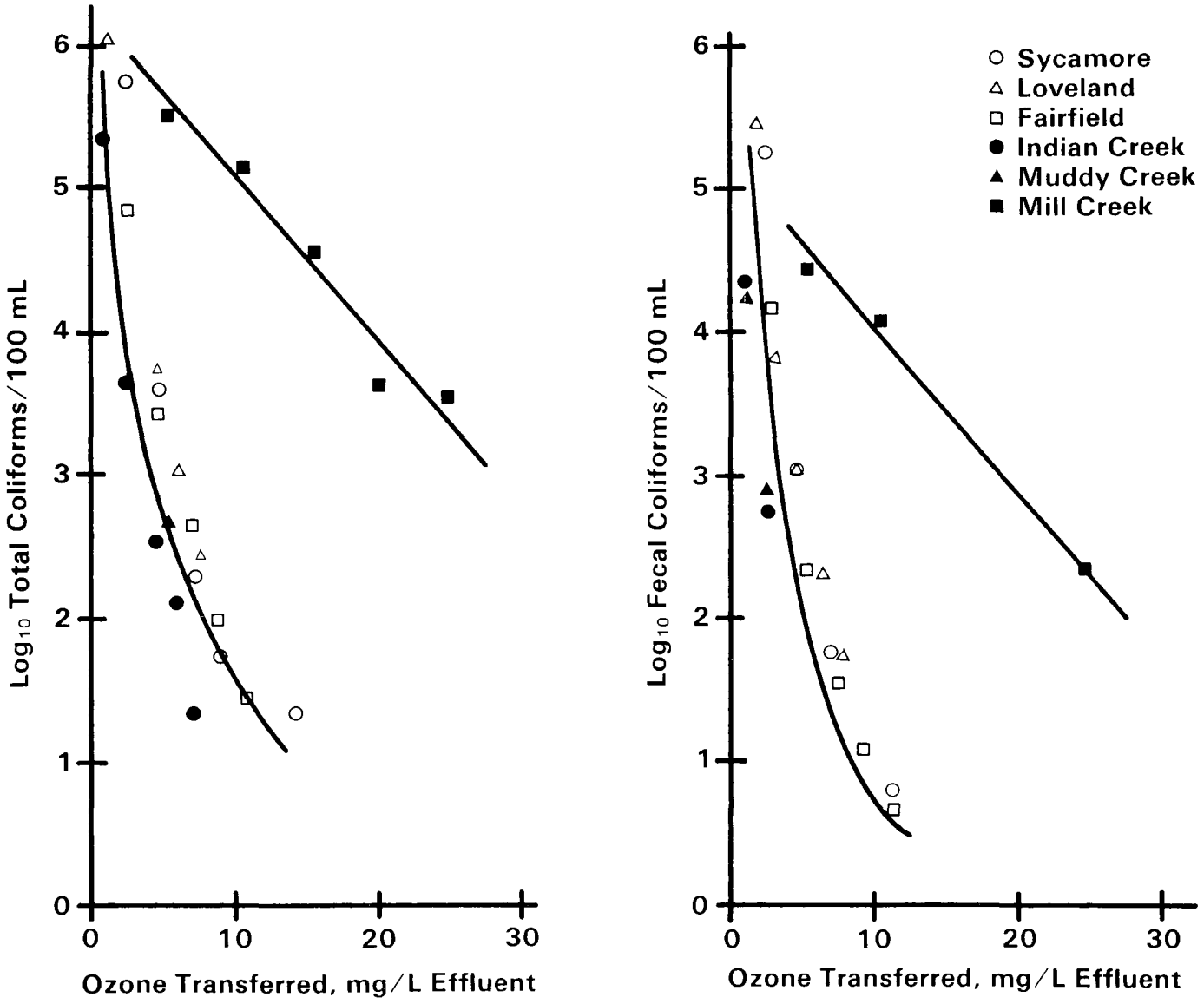

Figure 1. Effect of Ozone Transferred on Coliform Densities in Secondary Effluents. Each Point is Average of 5 Data Points.

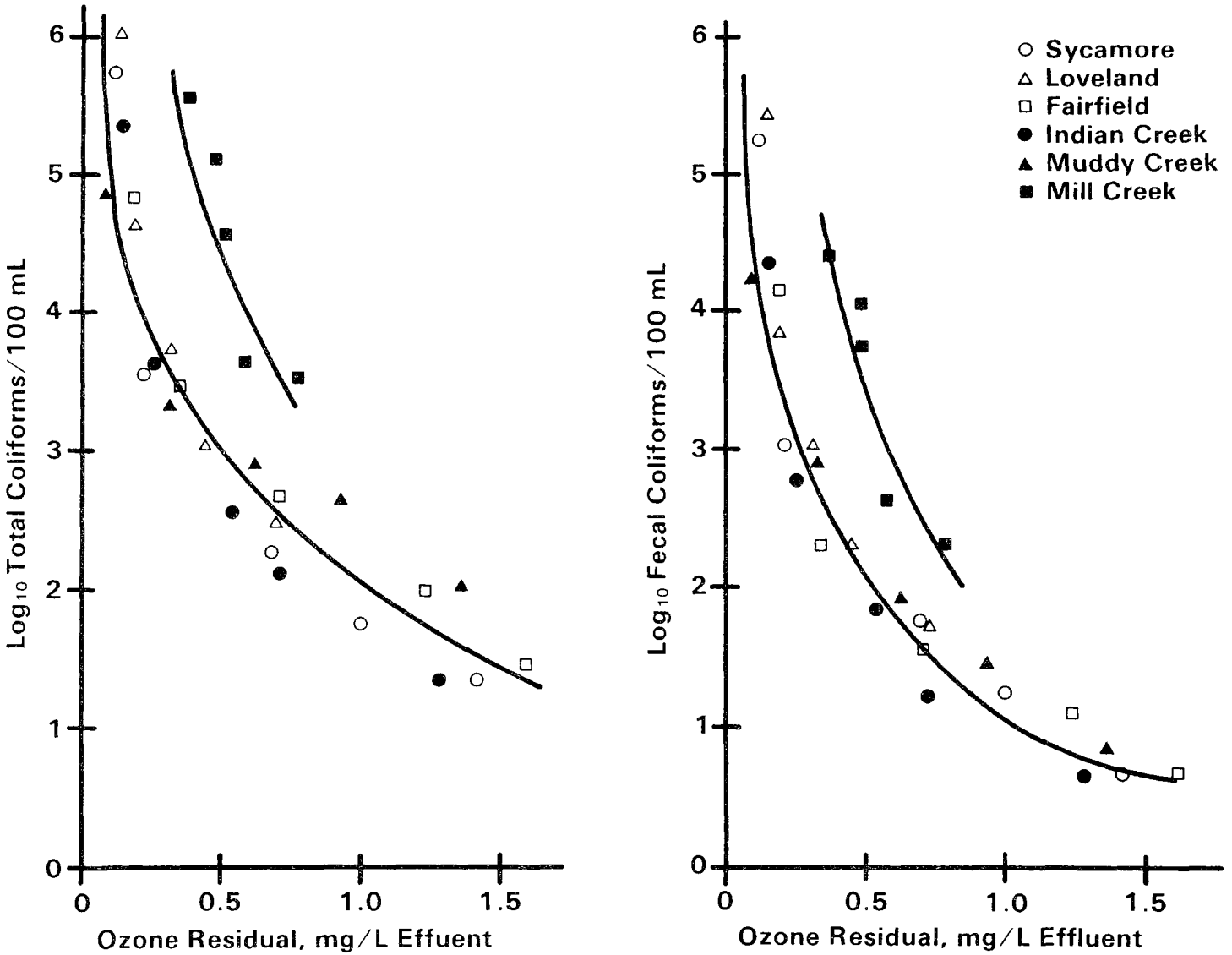

Figure 2. Effect of Ozone Residual on Coliform Densities in Secondary Effluents. Each Point is Average of 5 Data Points.

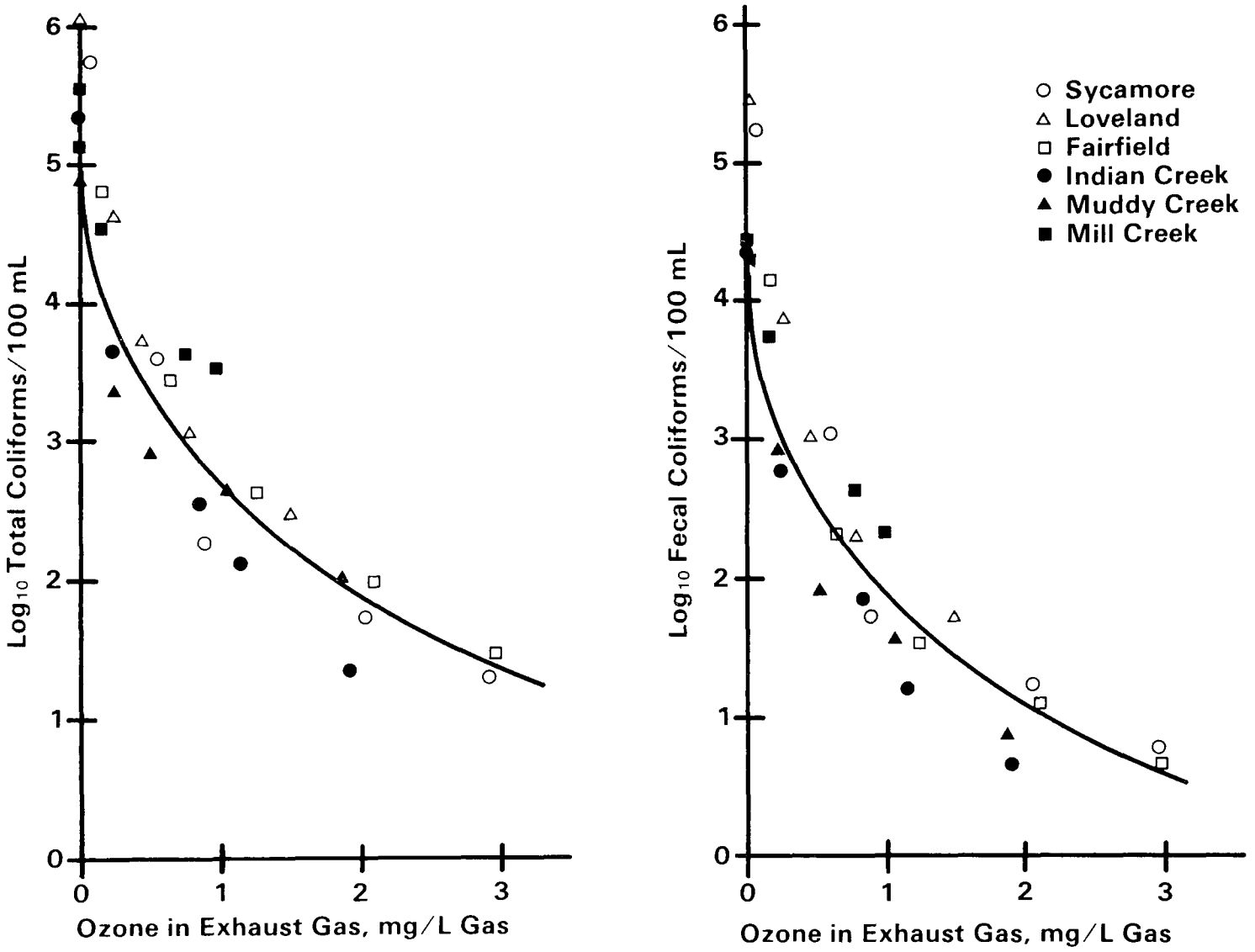

Figure 3. Effect of Ozone Concentration in Exhaust Gas on Coliform Densities in Secondary Effluents. Each Point is Average of 5 Data Points.

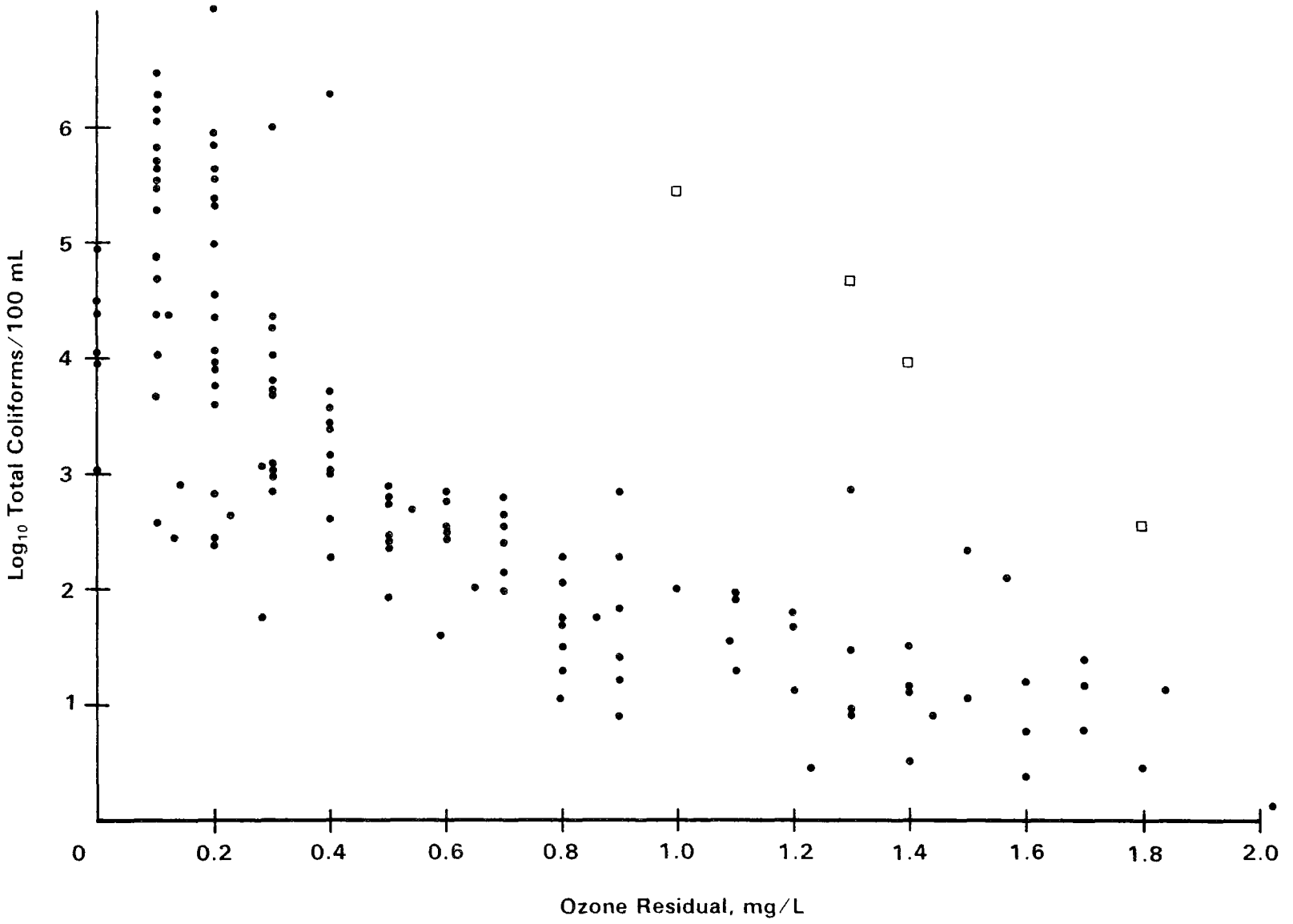

Figure 4. Effect of Ozone Residual on Total Coliforms in 5 of the 6 Effluents (All Data).

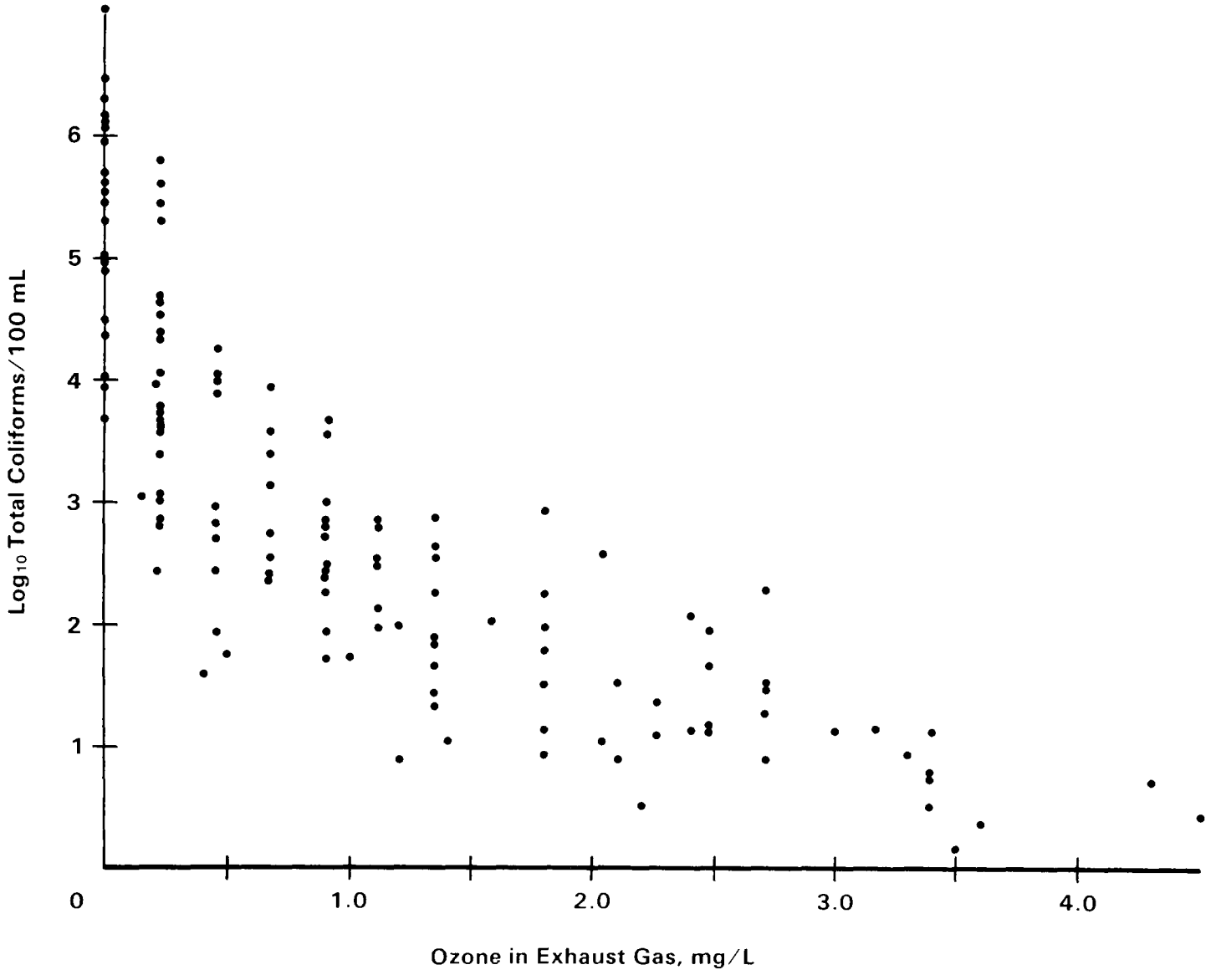

Figure 5. Effect of Ozone Concentration in Exhaust Gas on Total Coliforms in All 6 Effluents (All Data Points).

9. OPTIMIZING OPERATIONAL CONTROL OF OZONE DISINFECTION

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ABSTRACT

Ozone is being developed and employed for disinfection of municipal wastewater effluents in the United States as an effective alternative to chlorine. The factors affecting ozone system performance, water quality, transfer efficiency, and absorbed ozone concentration, are key issues to be addressed in both design and operation of ozone systems for municipal wastewater disinfection. The ozone contacting system cannot be optimized independently of the ozone generating equipment, because transfer efficiency in the contactors and power requirements for ozone generation are both related to ozone concentration in the carrier gas and the carrier gas flow rate.

Optimum operation of this equipment is required to minimize electric power consumption and thus operating costs. However, cost optimization must also consider achievement of the disinfection objectives. Therefore, the disinfection requirements in conjunction with both ozone production and ozone transfer efficiency become the key factors in optimization of ozone disinfection systems. A relationship must be developed between the disinfection requirements and the ozone system operating conditions to maximize disinfection and minimize power consumption. Upon development of these relationships the optimum operating conditions can be achieved.

Proper monitoring of these relationships such as ozone gas monitoring and residual liquid ozone monitoring is required to maintain adequate disinfection in the most efficient manner possible. This paper presents one approach for optimizing operation of ozone disinfection systems, including definition of disinfection requirements, ozone contacting and ozone generating equipment, as well as instrumentation requirements for monitoring and control.

INTRODUCTION

Municipal wastewater disinfection by ozonation is a relatively new and rapidly developing concept in the United States today, and is perceived by many to be the most attractive alternative to chlorination. There is very little information available describing design factors and operation criteria

for ozone disinfection facilities. The primary objectives that must be considered include the design and operation of an effective, reliable, economic and safe ozone disinfection system with minimal power consumption and maintenance requirements. Design and operation of such a system requires an understanding of the ozone generation equipment, ozone contacting equipment, factors affecting performance of this equipment, and ozone system instrumentation, monitoring and controls.

Water quality, ozone transfer efficiency and absorbed ozone requirements are key factors affecting ozone system performance that must be addressed for efficient design and operation of ozone systems for municipal wastewater disinfection. Since water quality influences both the ozone dose requirements and the ozone transfer efficiency, the ozone contacting system capabilities must be defined. The ozone contacting system cannot be optimized independently of the ozone generating equipment, because transfer efficiency in the contactors and power requirements for ozone generation are both related to ozone concentration in the carrier gas and the carrier gas flow rate.

An evaluation of ozone production efficiencies over the expected operating conditions must be considered for proper equipment selection and optimum operation. This can be accomplished by monitoring or mapping the power consumption versus ozone production over the available carrier gas flow range and available applied voltage range. This type of information can then be used to determine the proper size and number of ozone generators required to achieve the most economical design and optimal operating conditions. The ozone contacting equipment can also be evaluated to define the optimized operating conditions by monitoring the ozone transferred into the wastewater at various operating conditions. This concept for design of ozone disinfection systems has been previously described (2,3). The purpose of this paper is to combine these concepts of ozone equipment definition with disinfection requirements, instrumentation and monitoring equipment and controls for optimizing operational control of ozone disinfection.

OPTIMIZING POWER CONSUMPTION

In order to optimize or minimize power consumption for ozone production, it is necessary to evaluate the ozone generating equipment to define the economics of disinfection with ozone produced from the appropriate carrier gas (air or oxygen). This can be accomplished by mapping the ozone generator by monitoring the power consumption versus ozone production, as shown in Figures 1 and 2. In Figure 1 the ozone output is shown as a function of power consumption for the production of ozone from air, while in Figure 2 the ozone output is shown as a function of power consumption for the production of ozone from oxygen by the same ozone generator. The differences in generator power requirements for air versus oxygen generation of ozone are significant, as can be observed in these figures. The economics of oxygen supply at a given site would have to be considered as a function of carrier gas preparation to evaluate the actual total difference in economics for oxygen versus air operation. The total system power requirements (economics) for carrier

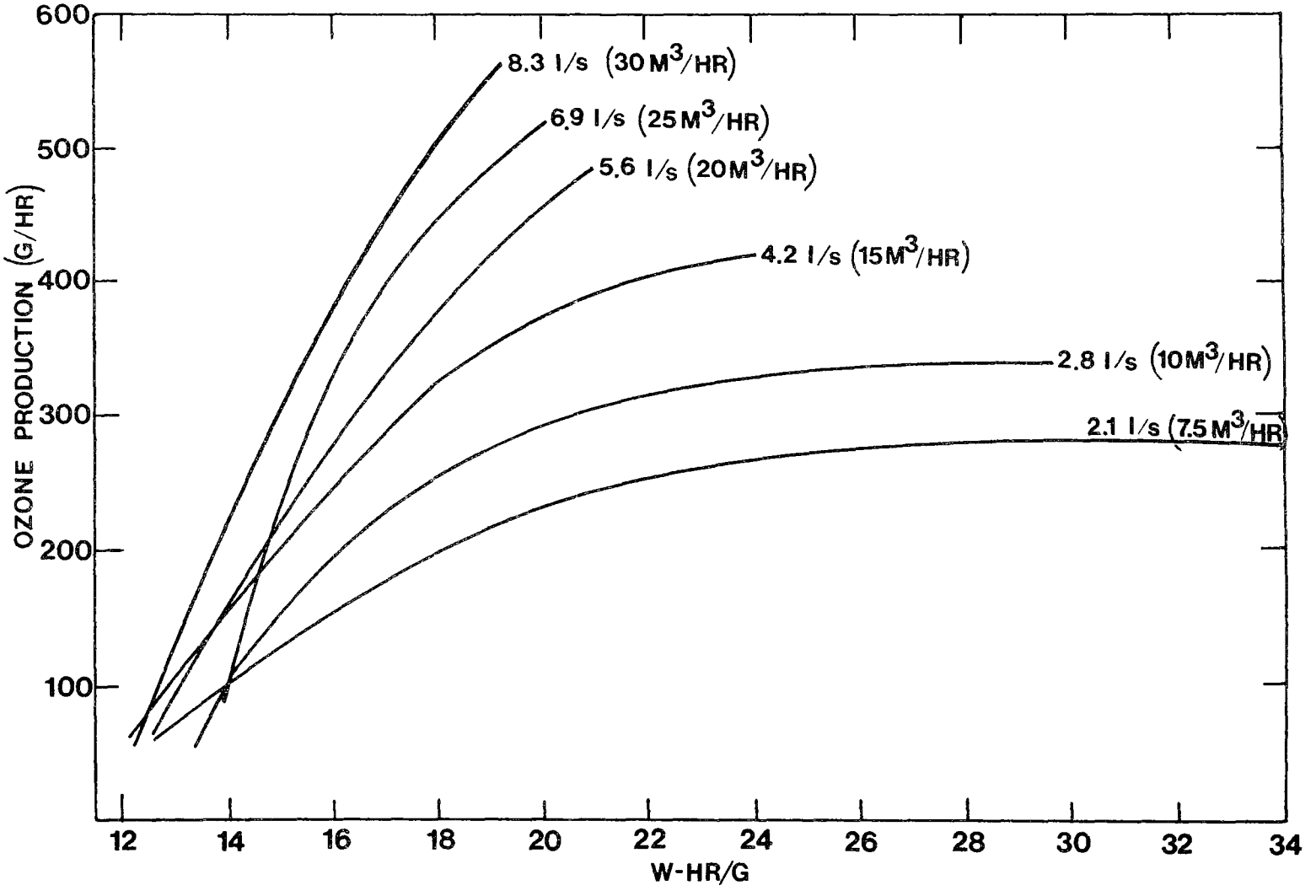

Figure 1. Ozone Generator Performance - Ozone Production Versus Power Consumption at Various Gas Flow Rates Using Air Carrier Gas.

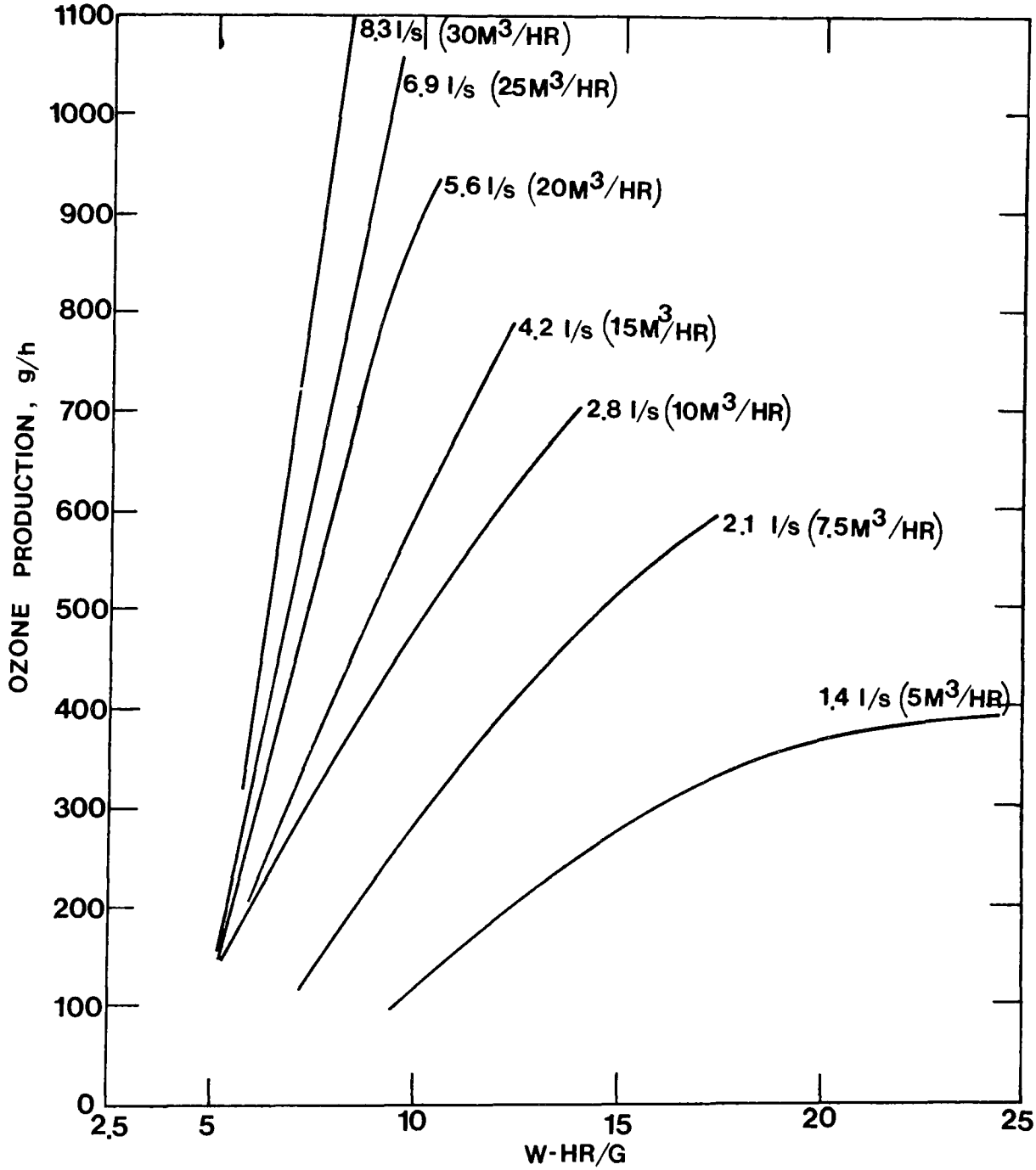

Figure 2. Generator Performance - Ozone Production Versus Power Consumption at Various Gas Flow Rates Using Oxygen Carrier Gas.

gas preparation and handling, ozone production, ozone contacting and ozone destruction can then be evaluated for comparison of air versus oxygen operation.

Economics of ozone disinfection dictate that ozone be utilized very efficiently due to the relatively expensive methods of ozone production available. Thus, the ozone contacting system must be designed for optimal ozone transfer or utilization by employing established principles of mass transfer and reaction kinetics. The ozone contacting system capabilities must be defined for optimization of the total ozone system (generation and contacting). The optimum obtainable ozone transfer efficiency compatible with economic ozone production required to achieve the disinfection objectives can then be determined. The ozone contacting equipment can be evaluated to define the optimized operating conditions by monitoring the percent ozone transferred into the wastewater versus the carrier gas ozone concentration at constant applied ozone doses, as indicated in Figure 3 for filtered secondary effluent. The shaded region represents the ozone gas concentration range where oxygen carrier gas operation starts becoming necessary to achieve the higher ozone concentrations.

As the applied ozone dose and subsequently the gas to liquid ratio increases at a constant carrier gas concentration, the percent ozone transfer into the effluent decreases even though the absorbed ozone concentration or quantity of ozone added to the effluent increases. The percent ozone transfer at a given applied ozone dose increases with increasing carrier gas concentration and corresponding decreasing gas to liquid ratio. Ozone contactor transfer efficiencies are higher during oxygen operation due to the higher ozone concentrations and lower gas to liquid ratios available to achieve the same applied ozone dose requirements compared to air operation. Under identical operating conditions of applied ozone dose, carrier gas ozone concentration, gas to liquid ratio and hydraulic flow rate, the percent ozone transferred into the effluent is independent of the type of carrier gas (air or oxygen), as shown in Figure 3.

As observed in Figure 3, several different operating conditions and applied ozone doses can be employed with a given effluent quality to achieve the same absorbed ozone concentration. In combining the contactor evaluation with the generator evaluation the ozone generating-contacting system can be optimized to achieve the desired absorbed ozone concentration. The absorbed ozone concentration(s) must then be correlated to the required disinfection objective(s), and proper instrumentation, monitoring and control used to maintain the disinfection objective(s) while ensuring optimized operation of the ozone generation and contacting equipment.

INSTRUMENTATION AND MONITORING REQUIREMENTS

Correlation of disinfection requirements with absorbed ozone concentrations provides the opportunity to achieve the disinfection objectives while maintaining optimal operation of both the ozone contactor and generator by

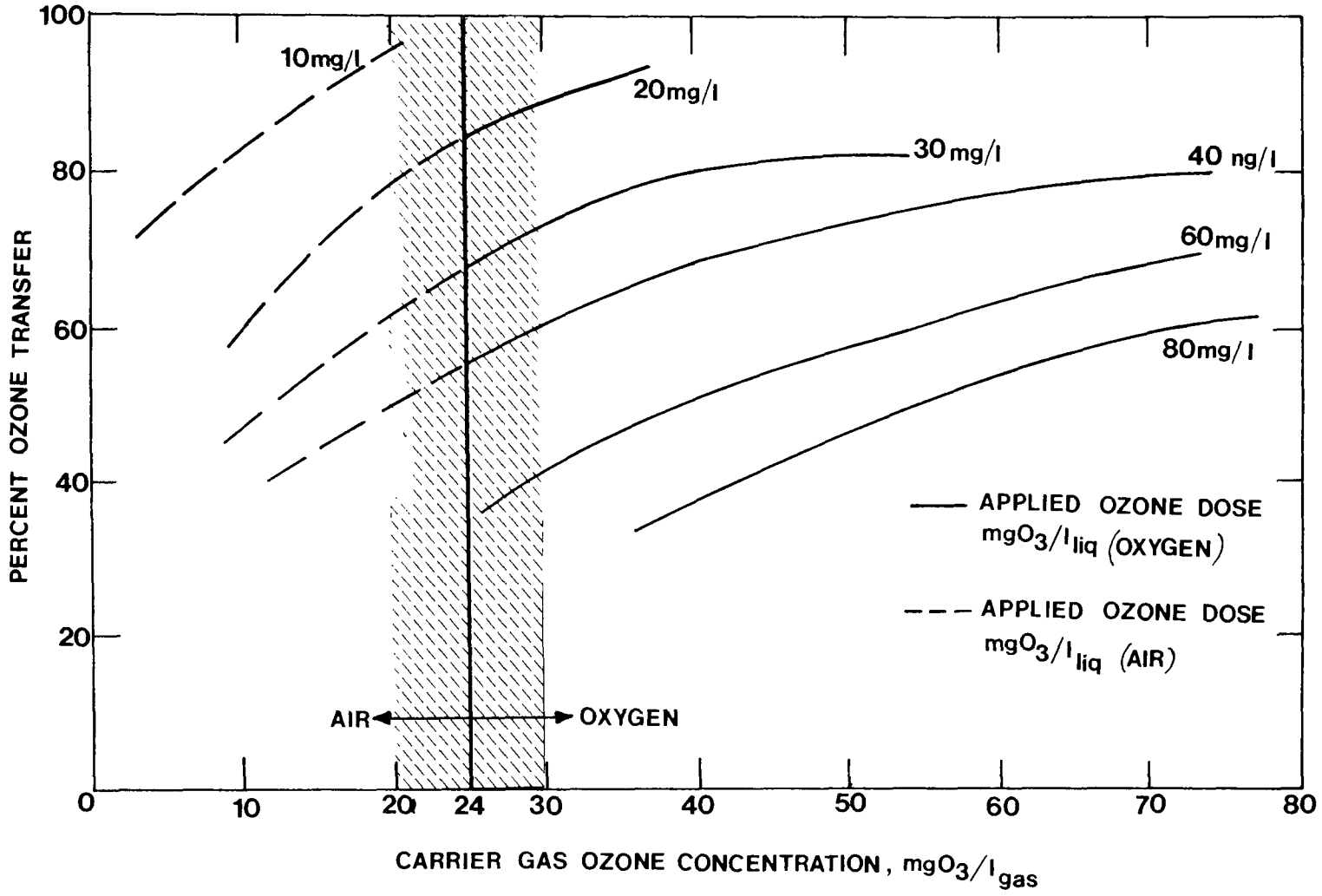

Figure 3. Ozone Transfer Efficiency at Various Applied Doses for Filtered Secondary Effluent at Average Effluent Flow Rate (2.5 l/s, 9 M^3/HR) Using Both Air and Oxygen Carrier Gas.

the methods established in the previous section. The results of such an analysis are presented in Figure 4 for the filtered secondary effluent quality shown in Table 1. This analysis was conducted from the data collected over a two year time period. Over the wide range of operating conditions evaluated during this time period, good correlation was always observed between total and fecal coliform reduction and absorbed ozone concentration.

Table 1. Summary of Filtered Secondary Effluent Characteristics

Parameter	Mean	Standard Deviation	Minimum	Maximum
TSS, mg/ℓ	4.8	3.8	1.6	16.4
Turbidity, NTU	4.2	2.2	1.4	12.0
COD, mg/ℓ	40	6.5	21	52
pH	--	--	6.9	7.9
Temperature, ^{o}C	--	--	6	21
Log_{10} total coliforms/100 mℓ	5.4	0.5	4.5	6.4
TOC, mg/ℓ	20	--	10	40
TKN, mg/ℓ	34	--	21	52
NH_3-N, mg/ℓ	14	--	12	16
NO_2-N, mg/ℓ	0.2	--	--	--
NO_3-N, mg/ℓ	0.1	--	--	--
Color, Pt-Co	50	--	45	100

Determination of the absorbed ozone concentration requires monitoring of the carrier gas ozone concentration and the contactor off-gas ozone concentration. From these gas measurements the percent ozone transfer efficiency can be determined and multiplied by the applied ozone dose to yield the absorbed ozone concentration (4). Ozone concentration in the carrier and off-gasses can be determined iodometrically by the method of Birdsall, Jenkins and Spadinger (1). This procedure is a manual method requiring collection of a gas sample in a gas washing bottle containing potassium iodide solution and measurement of the gas volume sampled by a wet test meter. The amount of ozone reacted with the potassium iodide is then determined by titration. This procedure can be used to accurately determine the ozone gas concentration; however, it is too cumbersome and time consuming to be used as a monitoring tool for ozone production and requires manual feedback to the generator for control of ozone production.

Instrumentation, such as the Dasibi Environmental Corporation Ozone Analyzer (Model 1003-HC) used in this study, are also available for monitoring ozone concentration in the gas streams. These instruments when properly maintained and recalibrated on a daily basis by the previously described iodometric procedure can provide reliable monitoring of system performance. In order to provide system control by absorbed ozone concentration, both the carrier gas and off-gas ozone concentrations would have to be monitored by two analyzers or one analyzer with alternating gas streams. Next the

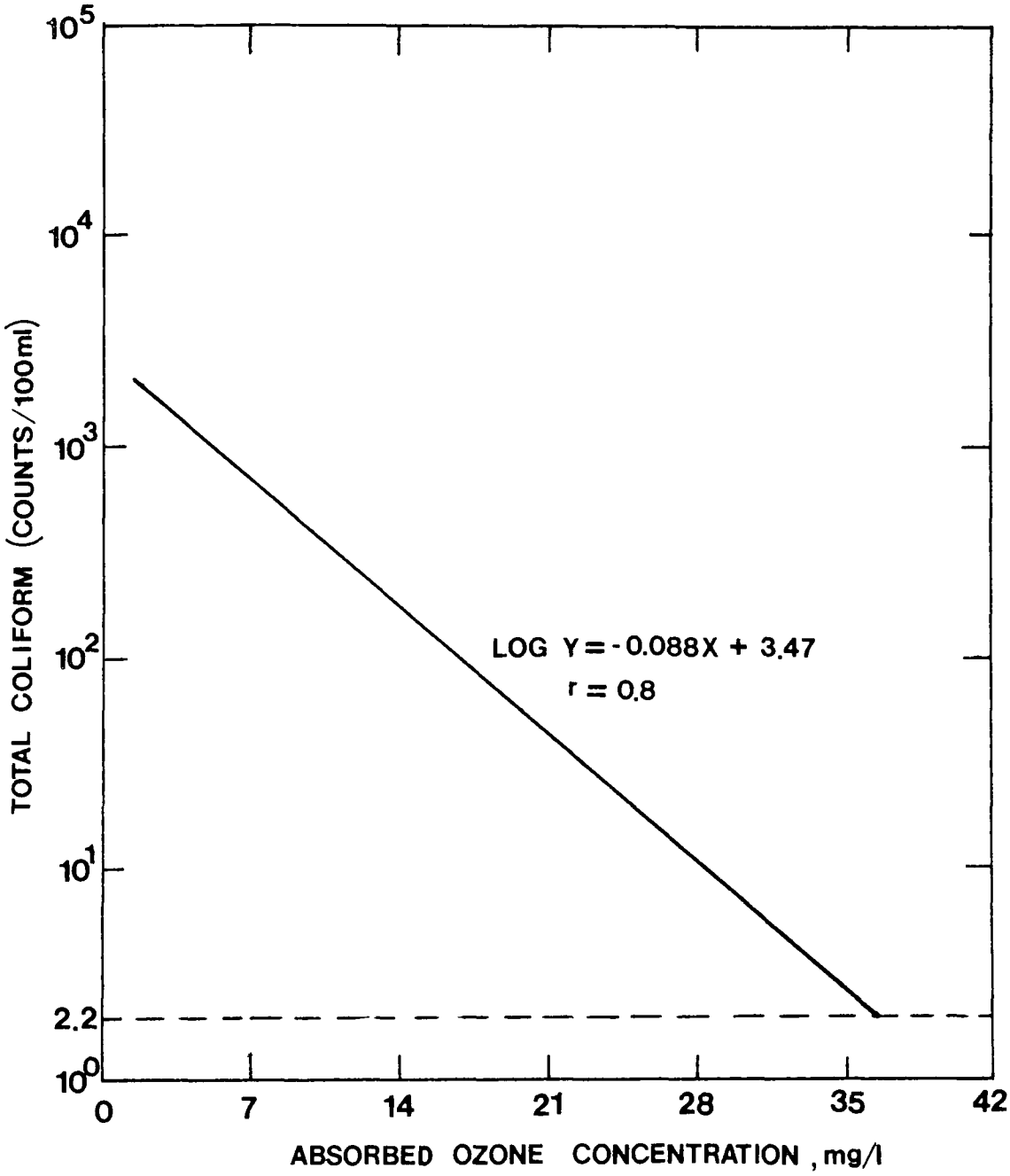

Figure 4. Effluent Total Coliform Value Versus Absorbed Ozone Concentration.

absorbed concentration would have to be determined and a signal relayed to the ozone generator to control the ozone output. The ozone output must be determined in terms of the applied ozone dose to the contactor and the absorbed ozone concentration evaluated as a function of both generator production and contactor transfer efficiency, as previously explained. Even with reliable instrumentation this is a very complicated procedure which still requires manual input to maintain optimized operating conditions and disinfection.

Excellent correlations of total and fecal coliform reduction were also observed with effluent total residual oxidants or total residual ozone, as shown in Figure 5 for total coliforms. A modification of the amperometric titration method for total residual chlorine was employed throughout the two year study period for determination of total residual oxidants and establishment of the solid line relationship shown in Figure 5 (4). This test method measures total residual oxidants, such as ozone, peroxides, etc., that may be produced during the ozonation process. A Delta Scientific Continuous Automatic Ozone Monitor Controller (Model 8340) was also used during the latter stages of the project to determine dissolved ozone levels in the ozonated effluent. Monitoring was performed continuously during this stage of the project with the immersed Delta Scientific probe that is claimed to be specific for dissolved ozone. This Delta Scientific residual ozone monitor also provided reliable instrumentation capabilities when properly maintained and calibrated on a daily basis.

This instrument provided residual ozone readings that correlated well with the total residual oxidant levels determined amperometrically, as shown in Figure 6. The residual ozone levels were typically around 60 percent of the total residual oxidant values. Total residual oxidants and residual ozone both correlated well with the absorbed ozone concentration as shown in Figure 7. Since these parameters correlate well with absorbed ozone concentration, they both provide excellent potential as a process control parameter for ozone disinfection.

Determination of total residual oxidants is a manual procedure, and thus, presents the same disadvantages as manual determination of absorbed ozone concentration for use as a process control parameter. However, instrumental determination of residual ozone provides an excellent opportunity for process control by providing a direct signal to the ozone generator. A simple feedback control loop from the residual analyzer to the ozone generator could be used to provide process control by monitoring of a single parameter. The excellent correlation of residual ozone to total residual oxidants (Figure 6) allowed the development of the dashed line in Figure 5 which presents effluent total coliforms as a function of effluent residual ozone concentrations.

DISINFECTION SYSTEM OPTIMIZATION APPROACH

The filtered secondary effluent quality of this study required residual ozone concentrations of around 0.5 mg/ℓ, 2.5 mg/ℓ and 5.0 mg/ℓ to achieve

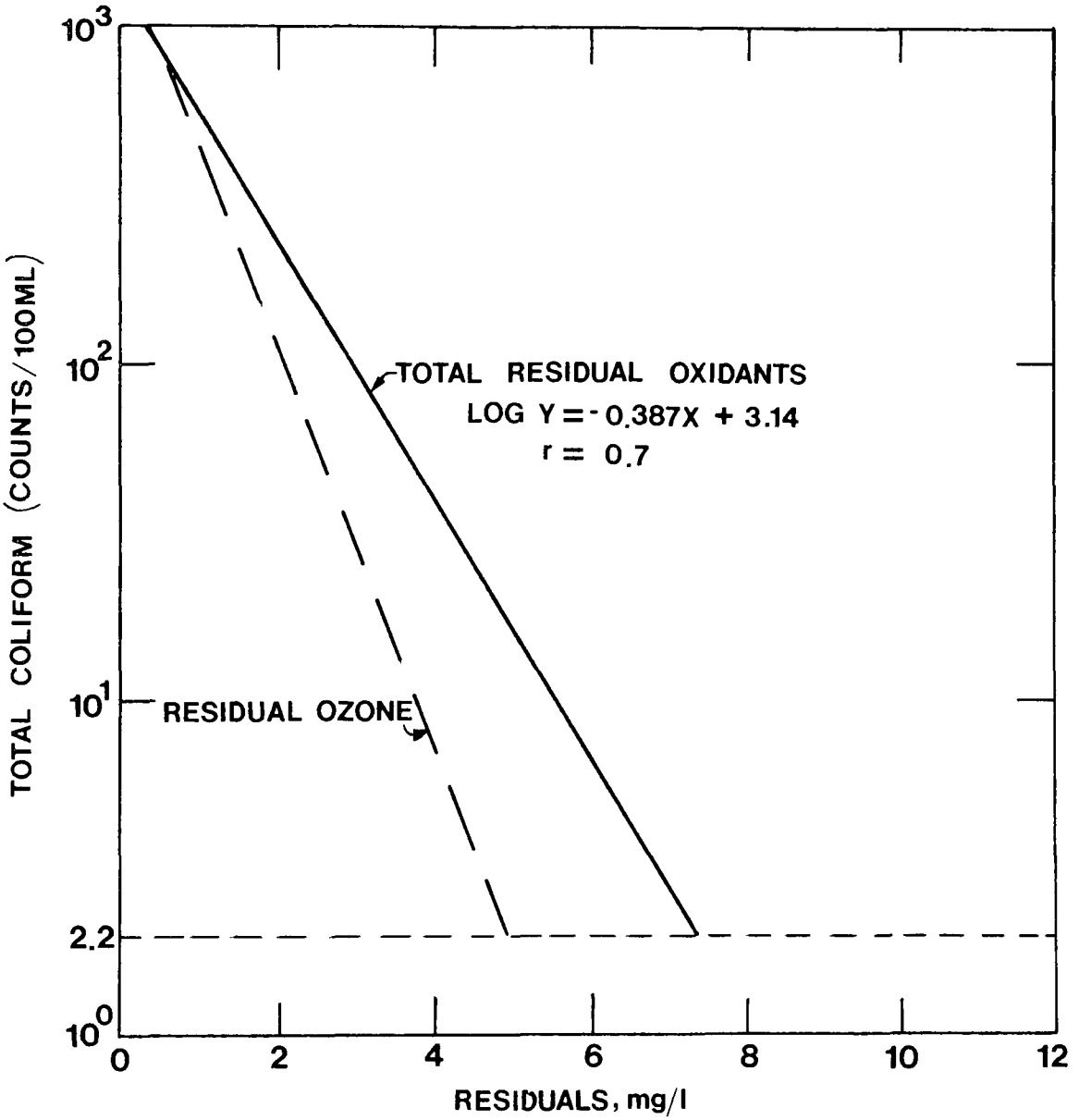

Figure 5. Effluent Total Coliform Value Versus Total Residual Oxidants and Residual Ozone.

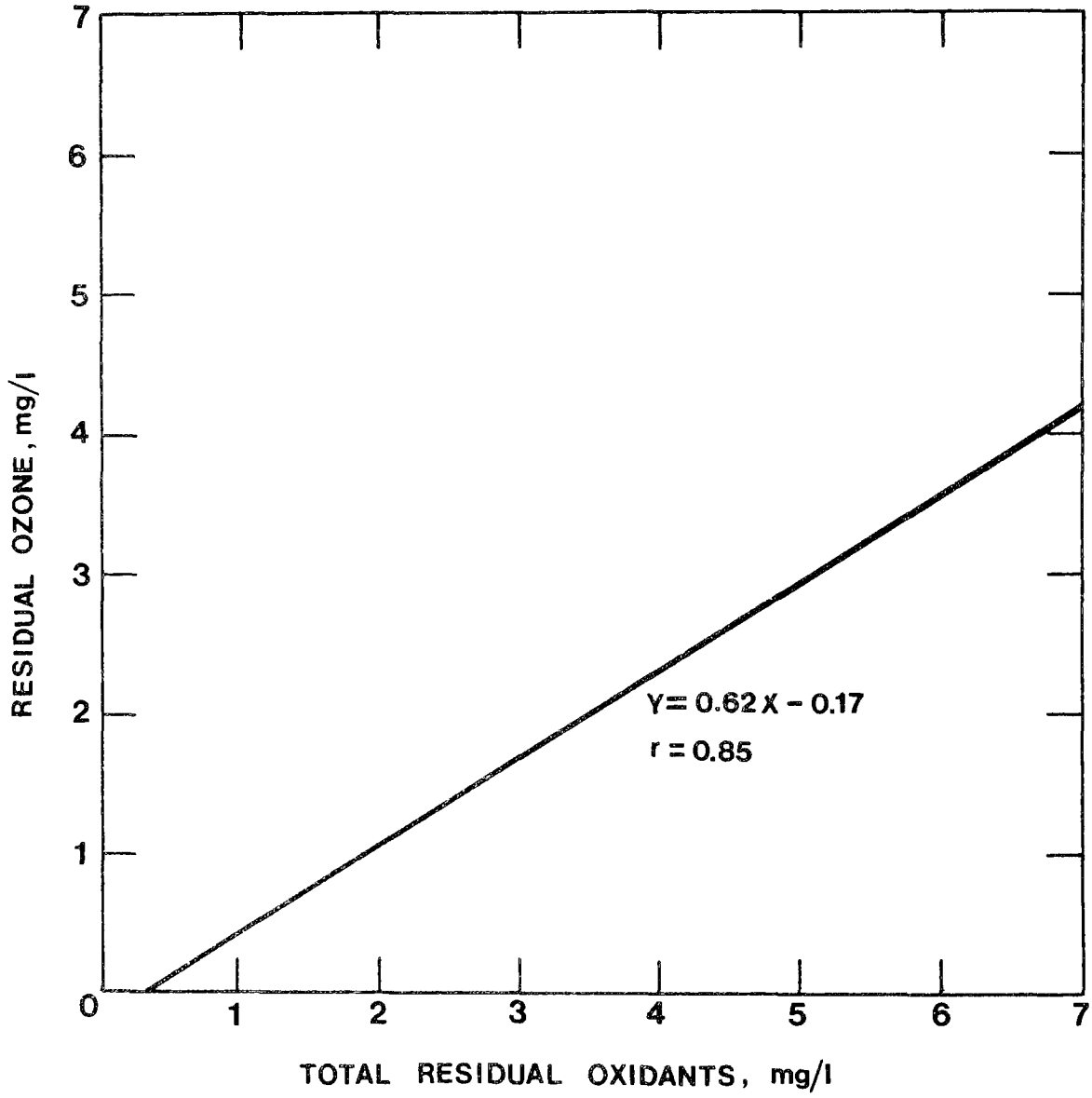

Figure 6. Residual Ozone Versus Total Residual Oxidants.

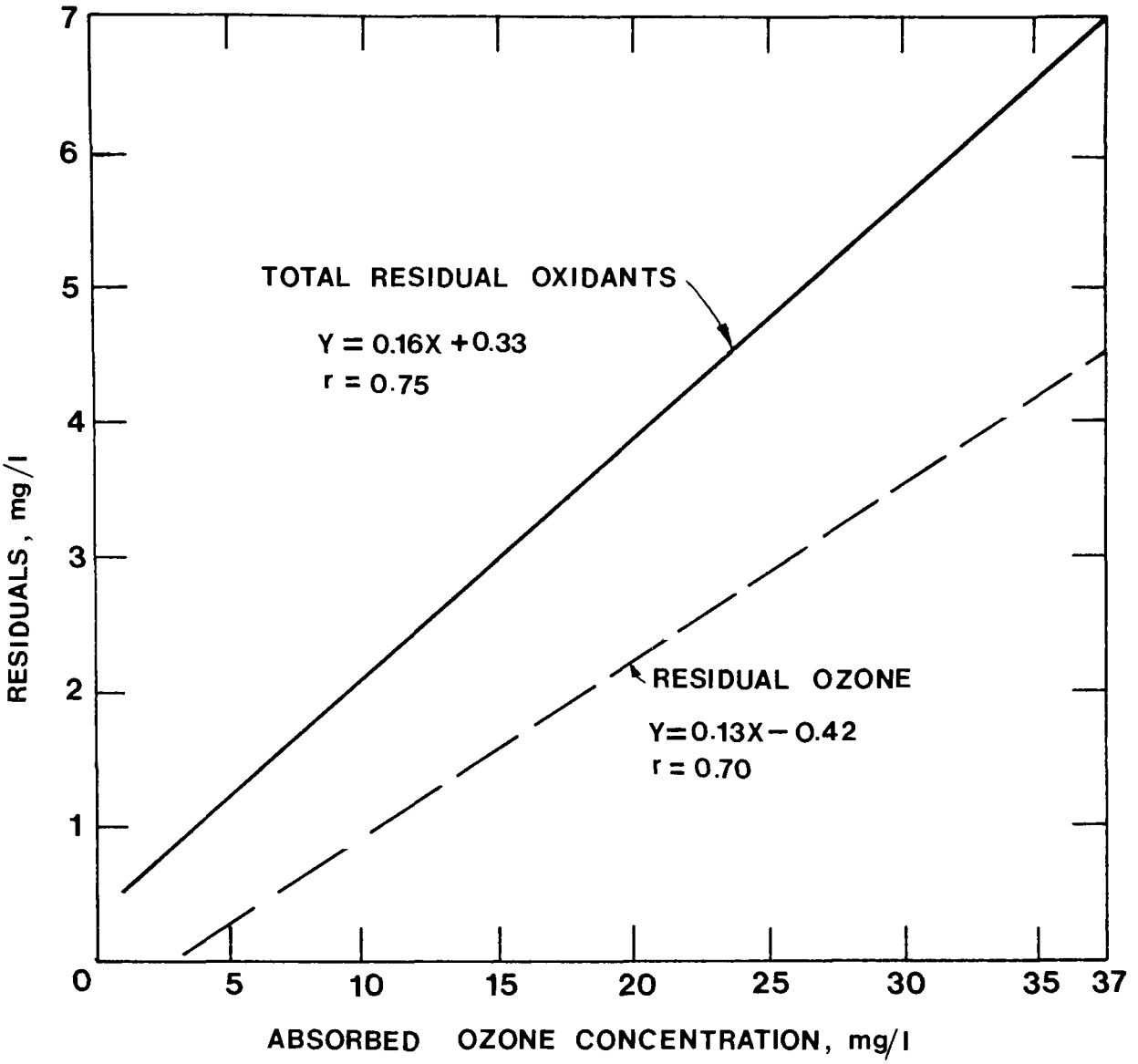

Figure 7. Total Residual Oxidants and Residual Ozone Versus Absorbed Ozone Concentration.

effluent total coliform values of 1000, 70 and less than 2.2 counts per 100 milliliters, respectively, (Figure 4). These residual ozone values can then be correlated with the required absorbed ozone concentrations to provide the required levels of disinfection, as shown in Table 2. These required absorbed ozone concentrations can then be evaluated in conjunction with the contactor and generator mapping curves to determine the optimized operating conditions for both ozone production and ozone contacting to achieve the desired absorbed ozone concentrations. This economically optimized absorbed ozone concentration thus provides the required residual ozone concentration and the required disinfection objectives.

Table 2. Effluent Residuals and Absorbed Ozone Requirements to Obtain Disinfection Objectives

Effluent total coliforms/100 mℓ	Total Residual Oxidants, mg/ℓ	Residual Ozone mg/ℓ	Absorbed Ozone mg/ℓ
1000	0.5	0.5	5
70	3.5	2.5	20
<2.2	7.5	5.0	38

An evaluation of this ozonation system operating for achievement of the disinfection objective of 70 total coliforms per 100 milliliters is shown in Table 3. The numbers generated in Table 3 can be developed from the information shown in Figures 1,2 and 3. As can be seen in Figure 3, this system providing a 20 mg/ℓ absorbed ozone concentration is operating in the range of ozone production requirements where oxygen operation starts becoming feasible due to the high ozone gas concentrations required. Operation with oxygen carrier gas would be more efficient than air operation because of the lower power requirements, less wasted power and higher ozone transfer efficiencies possible. As the ozone gas concentration increases, the gas flow rate requirement decreases, the ozone output decreases, and the total power requirement for ozone production decreases. Of course the economics of air versus oxygen preparation and handling would also have to be considered. The only difference in total economics for this system analysis presented here during air versus oxygen operation would be in gas handling and preparation costs.

The optimized operating condition during air carrier gas operation was at an applied ozone dose of 25 mg/ℓ with an ozone transfer efficiency of 80 percent and generator power requirement of 4275 watt power draw. This operating condition corresponded to both optimum ozone production and maximum ozone transfer efficiency. The optimized operating condition during oxygen carrier gas operation was at an applied ozone dose of 30 mg/ℓ with an ozone transfer efficiency of 67 percent and generator power requirement of 1897 watt power draw. This operating condition corresponded to optimum ozone production (minimal power requirement) but not maximum ozone transfer efficiency. Even though 89 grams per hour of ozone was wasted, the total generator power

Table 3. Combined Ozone Generating-Contacting Evaluation to Achieve 70 Total Coliforms Per 100 Milliliters (20 MG/L Absorbed Ozone Concentration Required).

Applied Ozone Dose mg/ℓ	Percent Ozone Transfer Required	Carrier Gas Ozone Concentration mg/ℓ(g/m^3) (Figure 3)	Carrier Gas* Flow Rate ℓ/s (m^3/hr)	Required** Generator Output (g/hr)	Generator Power Requirements W-hr/g	Watts	Wasted Ozone Output Percent	g/hr
			Operation with Air Carrier Gas (Figure 1)					
25	80	30 (30)	2.1 (7.5)	225	19.0	4275	20	45
30	67	24 (24)	3.1 (11.3)	271	18.5	5013	33	89
35	57	22 (22)	4.0 (14.3)	315	18.0	5670	43	135
40	50	20 (20)	5.0 (18.0)	360	18.5	6660	50	180
50	40	18 (18)	6.9 (25.0)	450	18.0	8100	60	270
			Operation with Oxygen Carrier Gas (Figure 2)					
20	100	45 (45)	1.1 (4.0)	180	13.0	2340	0	0
25	80	30 (30)	2.1 (7.5)	225	9.0	2025	20	45
30	67	24 (24)	3.1 (11.3)	271	7.0	1897	33	89
35	57	22 (22)	4.0 (14.3)	315	7.0	2205	43	135
40	50	20 (20)	5.0 (18.0)	360	7.0	2520	50	180
50	40	18 (18)	6.9 (25.0)	450	6.5	2925	60	270

* Applied Ozone Dose = Gas Ozone Concentration $X\left(\frac{\text{Gas Flow Rate}}{\text{Effluent Flow Rate}}\right)$

** Generator Output = Carrier Gas Ozone Concentration X(Carrier Gas Flow Rate).

Effluent Flow Rate = 2.5 ℓ/s (9.0 M^3/HR).

consumption was lower when compared to lower applied ozone doses and corresponding higher ozone transfer efficiencies. This evaluation shows the importance of combining the ozone generating equipment with ozone contacting to optimize the overall disinfection process to minimum power consumption. The optimum operating condition with oxygen carrier gas required only 44 percent of the generator power requirement compared to the optimum operating condition with air carrier gas.

During this two year test period, continuous operating periods were conducted to include night time and weekend testing for evaluation of the ozonation system disinfection reliability, including process control, instrumentation and equipment reliability, at changing water qualities due to diurnal variations. One such operating period was conducted to evaluate disinfection of the filtered secondary effluent to 70 total coliforms per 100 milliliters during operation with air carrier gas. During this operating period the generator output was paced to maintain a constant applied ozone dose of 30 mg/ℓ. The maximum to minimum effluent flow rates varied by a four to one ratio. The gas flow rate was varied manually to simulate an automatic gas flow regulation since the research facility did not have automatic gas flow rate controllers. During this test period the effluent quality varied as shown in Table 4. With the applied ozone dose of 30 mg/ℓ a mean absorbed ozone concentration of 20 mg/ℓ and effluent geometric mean total coliform count per 100 milliliters of 22 was achieved. Successful disinfection to the 70 total coliform level was achieved in greater than 80 percent of the test observations during this test period.

Table 4. Variability in Filtered Secondary Effluent Characteristics During Continuous Operating Period (Disinfection Objective of 70 Total Coliforms per 100 ML.)

Parameter	Mean	Standard Deviation	Minimum	Maximum
TSS, mg/ℓ	4	1.6	2	11
Turbidity, NTU	5.4	1.8	2.9	9.3
COD, mg/ℓ	31	6.1	12	53
pH	--	--	6.4	7.9
Temperature, ^{o}C	15.0	--	14.5	15.5
Log_{10} total coliforms/100 mℓ	5.0	0.4	4.0	5.9

The total power draw in watts of the complete ozone disinfection system was monitored over this continuous operating period. The ozone thermal destruct unit required a constant 4200 watt power draw, and the two submerged turbine type ozone contactors averaged 2600 watt power draw each. However, each ozone contactor was oversized to allow the total research facility hydraulic flow to be disinfected in each contactor, as has been previously described (5). This design was developed for the facility to provide the operational flexibility required for evaluation of the feasibility of high

level ozone disinfection. The power draw of the ozone generator and air preparation equipment was variable during this test period. The average power requirement for ozone generation during this test period was 22 W-hr/g, while the average power requirement for both ozone generation and air handling and preparation was 24 W-hr/g. This power requirement was beyond the optimum operating range for ozone production by this generator with air carrier gas, due to lack of optimized operational control during this test period and the changing hydraulic flow rates and ozone demands (the primary objective at the time of this test period was to demonstrate reliable disinfection to 70 total coliforms per 100 milliliters). The changing hydraulic flow rates and corresponding relationships of ozone transfer and ozone production must be evaluated for complete system optimization, as has been previously presented (2,5).

DISCUSSION AND CONCLUSIONS

The importance of optimizing operational control of ozone disinfection has been demonstrated during the continuous monitoring period described. The disinfection objective was achieved; however, the overall system power requirements were not optimized. This lack of ozone generator optimization was due partially to the required operating conditions which approach oxygen carrier gas ozone concentrations. Lower power requirements for ozone production could be realized with a larger ozone generator or by operating the existing generator with oxygen carrier gas. However, the optimization approach presented here can be used both in design or optimization of operations of an existing system.

Monitoring ozone residual with the proper instrumentation and providing a direct feed back signal to the ozone generator allows the generator operating conditions to change in response to changing water quality or changing ozone demand. The ozone output, ozone concentration and carrier gas flow rate can be changed to maintain the proper conditions for both optimized ozone production and ozone transfer. This requires mapping or monitoring both the ozone generator power requirements and ozone contactor transfer capabilities to combine this information into the most economical operating condition.

Another possible optimization approach suggested by the results of the continuous testing period would be monitoring of the contactor off-gas concentration as an indicator of the absorbed ozone concentration. During this test period the applied ozone dose was maintained constant at 30 mg/ℓ, while the generator voltage and gas flow rate were varied with the changing hydraulic flow rate to maintain a constant gas to liquid ratio in the contactor. With this operating strategy the mean absorbed ozone concentration was maintained at around 20 mg/ℓ. Since the gas flow rate varied with the hydraulic flow rate, the gas ozone concentration was maintained constant. Thus, both the gas ozone concentration and the contactor transfer efficiency were approximately constant during this test period. With both a constant carrier gas ozone concentration and transfer efficiency, the contactor off-gas

concentration would also be constant. Under this operating strategy the off-gas ozone concentration could be monitored with the proper instrumentation and a direct feedback signal employed to control the generator output by maintaining the constant off-gas concentration. This approach has also been suggested by Venosa (6).

Both approaches presented here appear applicable for optimizing operational control of ozone disinfection. The off-gas monitoring approach requires control of both the ozone generator gas concentration and flow rate to maintain a constant gas to liquid ratio. The residual monitoring approach only requires control of the ozone generator gas concentration to maintain the desired residual ozone concentration. However, the data generated during this research study indicates that optimized ozone disinfection, considering both generation and contacting, appears to be obtained at a constant gas to liquid ratio. Therefore, liquid residual monitoring would also require maintenance of a constant gas to liquid ratio in the contactor, not to accomplish the disinfection objective but to accomplish economic optimization

ACKNOWLEDGEMENTS

The data used in this analysis was developed at the Marlborough, Massachusetts ozone disinfection research project sponsored by the U.S. Environmental Protection Agency, the Commonwealth of Massachusetts and the City of Marlborough, Massachusetts. At the time of this study the author was Director of Research and Development at Metcalf & Eddy, Inc. of Boston, Massachusetts.

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10. PILOT STUDIES OF OZONE DISINFECTION AND TRANSFER IN WASTEWATER

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ABSTRACT

Studies of ozone disinfection and transfer in wastewater, using counter-current flow contactors, were undertaken in Whitehorse, Yukon. The studies focussed on the effectiveness of ozone in the reduction of indicator organisms and relevant factors influencing this reduction in screened, dilute wastewater (BOD and suspended solids approximately 60 to 120 mg/L). The primary factor influencing bacterial survival was the amount of ozone utilized in the contactors. A log-log relationship was evident between bacterial survival and ozone utilization. Other variables which demonstrated an apparent effect on bacterial survival included wastewater strength, temperature, and ozone residual. At fecal coliform reductions of 99.9%, approximately 20 mg/L of ozone utilization was required. Using a high quality secondary effluent, only 4 mg/L of ozone was required to achieve the equivalent reduction. Other benefits of the ozonation system included substantial wastewater strength reductions and high dissolved oxygen levels in the effluent. Factors exhibiting an apparent effect on ozone transfer efficiency included the ozone-oxygen gas flowrate, amount of ozone applied, wastewater strength, and ozone residual.

INTRODUCTION

Ozonation of screened, dilute, cold, wastewater was studied at pilot plant scale in the City of Whitehorse, Yukon Territory, Canada. The study objectives were to evaluate ozone effectiveness in disinfection of the wastewater and to assess relevant performance factors. Secondary benefits of ozonation also were to be assessed.

Cold, dilute wastewater often results from the practice of discharging cold tap water to sewers to prevent water pipe freezing. It can also result from high in-flow and/or infiltration.

Review of this wastewater management problem in northern regions of Canada indicated that the most important treatment requirement may be proper disinfection. With this treatment requirement in mind, the screening-ozonation pilot scale study was developed as one of a number of alternative

treatment techniques for cold, dilute wastewater. These treatment techniques have been analyzed by Smith and Given (3).

DESCRIPTION OF PILOT PLANT

The pilot plant consisted of a rotating screen, with slotted openings (both 0.76 and 0.25 mm slot sizes tested), followed by two counter-current flow ozone contact columns (5.2 m high and 150 mm diameter). Raw wastewater was supplied to the rotating screen by a submersible pump from a wet well in the main wastewater lift station for Whitehorse. The effluent from the screen was collected in a mixed holding tank from which it was pumped to the ozone columns at uniform rates.

Another holding tank was used to collect effluent from a rotating biological contactor (RBC) during the first year of the study. The same holding tank was used for lagoon effluent and high strength wastewater at the end of the second study year. These wastes were tested separately with the ozone disinfection system.

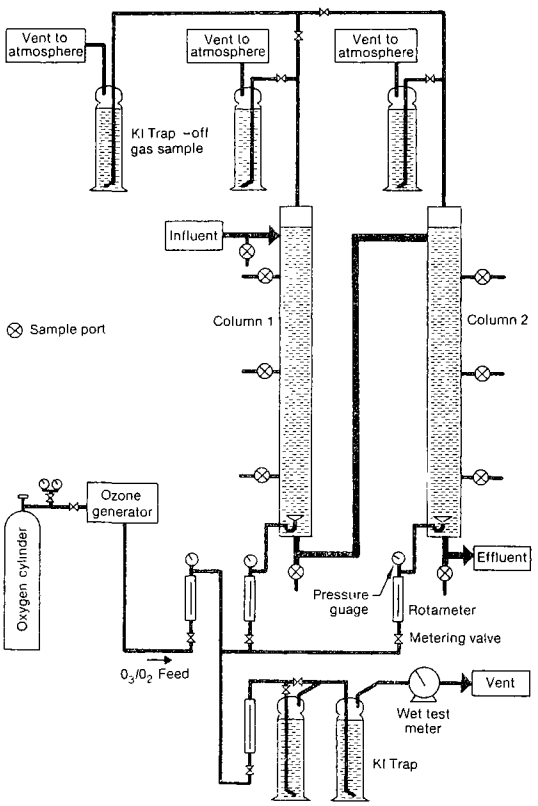


Figure 1. Schematic of Ozone Pilot Plant

Ozone was produced by passing oxygen through a 10 g/h, air-cooled ozone generator. The resulting mixture of oxygen and ozone was distributed near the bottoms of the columns through 90 mm diameter porous glass diffusers. The distribution of gas to the columns was controlled by metering valves and measured using calibrated rotameters with attached pressure gauges. Figure 1 presents a schematic of the pilot plant system.

METHODS

Pilot Plant Operation

A total of 300 runs of the ozone system were conducted over a two year period in 1977 and 1978. In the first year, a 3 x 3 x 3 operating matrix (three O_2/O_3 gas flow rates, three gas distribution ratios to the contact columns, and three wastewater flow rates) was set up. The operating matrix is presented in Table 1.

Table 1. Summary of Operation Matrix for Ozone System*

(1)	(2)	(3)
O_2/O_3 Gas Flowrate L/min @ STP	Ozone Concentration (% by wt.)	Gas Distribution % Column 1/% Column 2	Wastewater Flowrate L/min
1.6	(4.9)	50/50	3.8
3.0	(3.3)	60/40	11.4
6.0	(1.9)	70/30	30.3

* Average values presented.

In the second year of the study, a 1 x 1 x 3 matrix was used predominantly, with gas flowrate at 3.0 L/min, gas distribution at 60% to the first column and 40% to the second, and only wastewater flowrate was varied.

Normally a set of two to three runs of the ozone system was performed during a given day and as many as five sets during a given week. Most of the runs were performed during the January to April period of each year with wastewater temperatures ranging from 6 to 8 ^{o}C. Some runs were also performed in the summer when the wastewater was as warm as 14 ^{o}C.

Before sampling, the ozonation system was operated at the desired wastewater and O_2/O_3 gas flowrates for a minimum of six volume changes of the columns. Uniform wastewater flowrates to the columns were set from 3.8 to 30.3 L/min, resulting in total ozone contact times from 48 to 6 minutes, respectively. The wastewater flowrate variation was the primary method of varying the ozone dosage (mg O_3 per litre of wastewater). Ozone dosage was also varied to a limited extent by changing the oxygen flowrate through the ozone generator during different system tests. Gas flowrates were controlled by calibrated rotameters. Ozone applied and ozone in off-gases were deter-

mined by the iodometric procedure in Standard Methods (Procedure 423A) (1). Ozone residuals in the liquid were determined with a Wallace and Tiernan amperometric titrator.

Analytical Procedures

Biochemical oxygen demand over five days (BOD), chemical oxygen demand (COD), suspended solids (SS) and volatile suspended solids (VSS) were determined according to Standard Methods (1), Procedures 507, 508, 208D, 208G, respectively. Wastewater turbidity was measured with a Hach turbidimeter.

Total coliforms, fecal coliforms, fecal streptococci and 35 ^{o}C standard plate counts were enumerated by membrane filtration procedures according to Standard Methods Procedures 909A, 909C, 910B, 907. Results of Salmonella and virus determinations were reported elsewhere (2).

Data Analysis Methods

The disinfection data for screened wastewater were examined statistically to evaluate the possible effects of operating procedure, year of sampling, screen opening size, number of ozone contact columns, and wastewater characteristics. BOD and suspended solids reductions were also examined.

Comparison of disinfection efficiency of ozone using RBC effluent, lagoon effluent and a strong wastewater was made.

Because of the importance of ozone utilization on bacteria reduction, additional analyses were focussed on factors affecting ozone transfer efficiency for the contact columns.

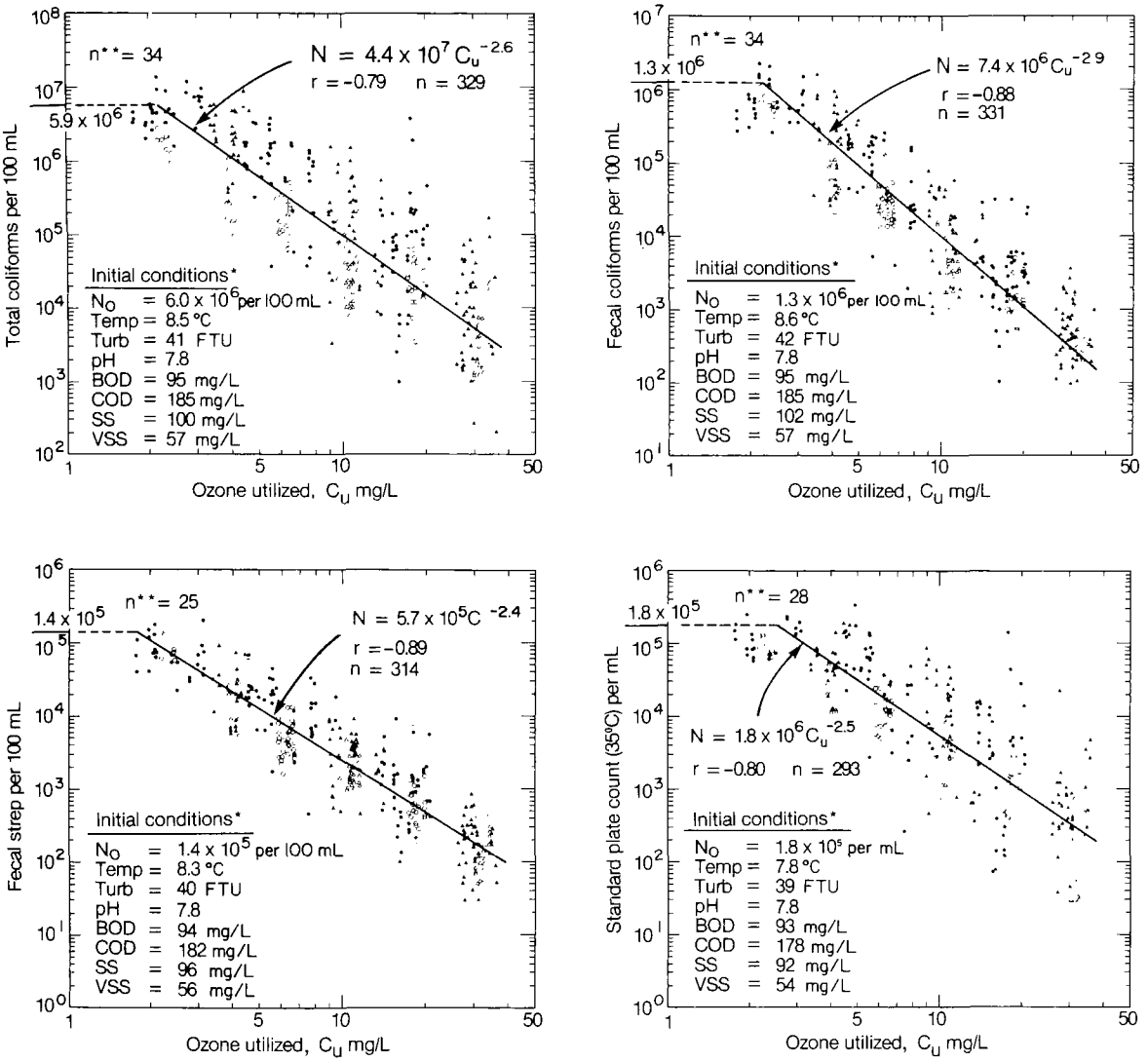
ANALYSIS OF SYSTEM PERFORMANCE

Bacterial Numbers and Survival

The disinfection results for screened wastewater are shown for total coliforms (TC), fecal coliforms (FC), fecal streptococci (FS), and standard plant count at 35 ^{o}C (SPC) in Figures 2 and 3. The figures show actual numbers and survival ratios respectively, plotted against ozone utilized (C_u). Linear regression lines and equations indicate a log-log relationship for each set of data points.

Of particular note with the indicator organism survival curves are the intercepts of the regression lines with the abscissa (C_u axis). These intercepts, which range from 1.8 to 2.5 mg/L of ozone utilized, may be thought of as the initial amount of ozone which must be utilized in the wastewater before significant bacterial reductions occur. The intercepts can be used conveniently along with the slopes of the regression lines to express bacterial survival in terms of a simple equation:

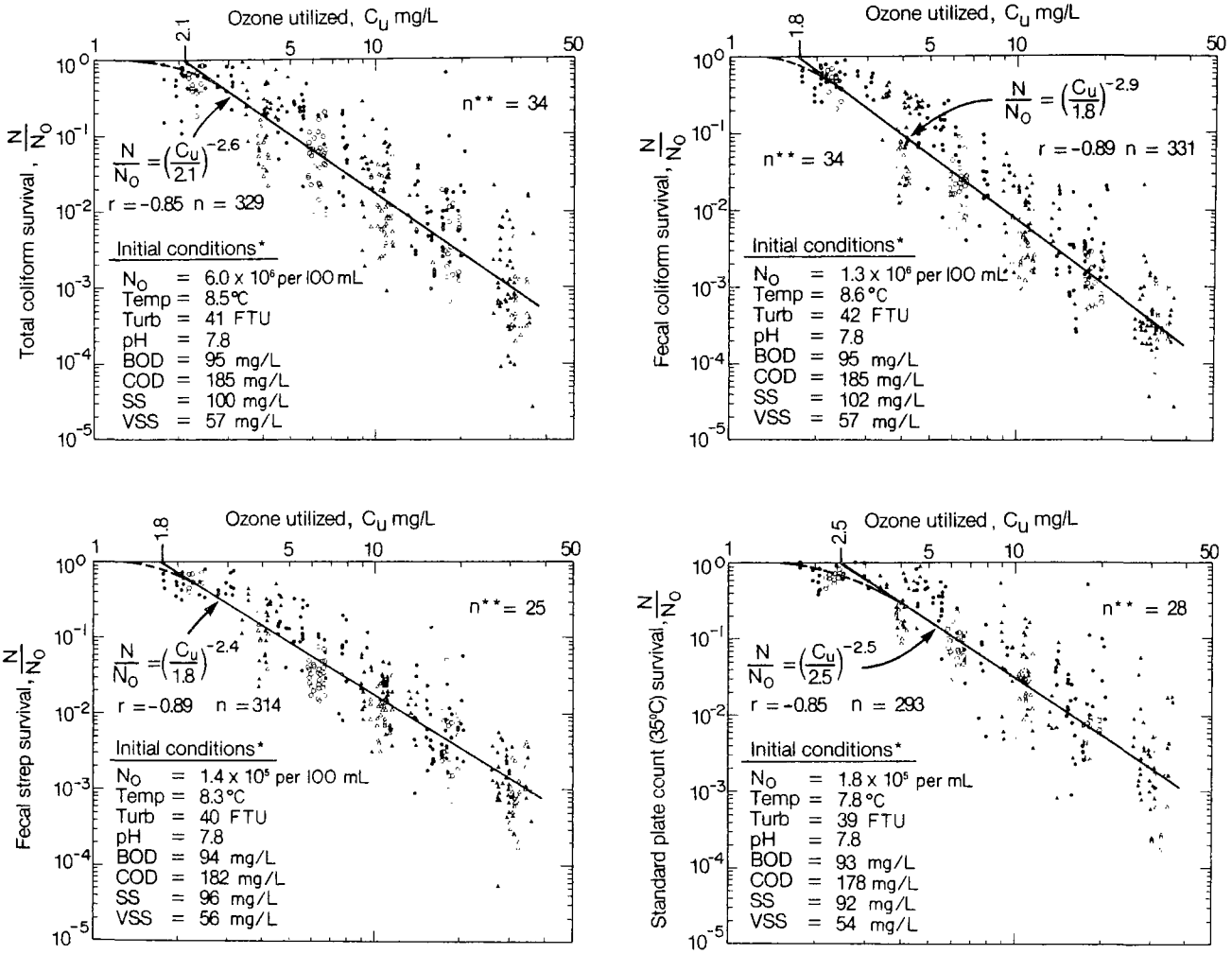
$$N/N_o = (C_u/C_{uo})b$$



LEGEND

● 1977 data, column 1 ○ 1978 data, column 1
▲ 1977 data, column 2 △ 1978 data, column 2

Figure 2. Total Coliform, Fecal Coliform, Fecal Streptococcus and Standard Plate Count (35 °C) Numbers Versus Ozone Utilized



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● 1977 data, column 1
▲ 1977 data, column 2
○ 1978 data, column 1
△ 1978 data, column 2

Figure 3. Total Coliform, Fecal Coliform, Fecal Streptococcus and Standard Plate Count (35 °C) Survival Versus Ozone Utilized

where N_o = initial number of indicator organisms
N = number of indicator organisms after ozonation
N/N_o = bacterial survival ratio
C_u = the concentration of ozone utilized (mg/L)
C_{uo} = the initial concentration of ozone utilized (mg/L) or intercept with C_u axis
b = slope of regression line (negative number)

It should be noted that data points for which ozone utilized values were less than 2.5 mg/L were not included in the regression analyses because the statistical residuals (observed values minus values predicted by the regression lines) were generally negative. This indicated that the data showed transition from the "lag phase" to the "rapid kill phase" when ozone utilized increased beyond 2.5 mg/L, for the screened wastewater.

Additional data analyses of microorganism survival indicated that there did not appear to be any significant effects of operating procedures, year of operation, screen slot size, and number of contact columns. Consequently, all of the screened wastewater data were pooled in the calculations for the regression lines.

Predictive Relationships for Bacterial Survival

Stepwise multiple linear regression techniques were used to determine the statistical significance between bacterial survival as the dependent variable, and several possible independent variables. Combinations of log (base 10) transformed and non-transformed variables were investigated to determine best predictive relationships for bacterial survival.

The log transformations for ozone utilized (log C_u) gave the best initial correlation with log (N/N_o) in all cases (Step 1). The next most significant variable generally proved to be log BOD (Step 2). However, both wastewater temperature (log T) and effluent ozone residual (log C_{re}) were more significant for some of the indicator organisms. This is illustrated in Table 2 which presents the complete summary of the analyses for each of the indicator organisms.

The tabulated values of r^2 (square of the multiple correlation coefficient) show that it is of marginal benefit to include more than two independent variables in the regression equations. The r^2 value indicates the fraction of the total variance of log (N/N_o) which is contributed by its regression on the independent variables.

The apparent effects of BOD and temperature as well as ozone utilized on the survival of fecal coliforms for the screened wastewater are shown in Figure 4. It was observed that increases in both initial BOD and temperature resulted in higher initial ozone demand. This indicates that, for a given level of ozone utilization, higher survival ratios would occur if either of these parameters are increased. It should be noted that this is an apparent effect as the project testing program did not allow absolute control of all independent variables.

Table 2. Summary of Regression Analyses for Bacterial Survival in Screened Wastewater*

INDEPENDENT VARIABLE	NUMBER OF COLUMNS	STEP	REGRESSION COEFFICIENTS FOR					r^2	n
			b_o	log C_u	log BOD	log T	log C_{re}		
log(FC/FC_o)	1	1	0.74	-2.8				0.750	109
		2	-3.2	-2.6	1.9			0.814	
		3	-3.3	-2.3	1.7		-0.16	0.833	
		4	-3.4	-2.4	1.5	0.62	-0.13	0.838	
	2	1	0.61	-2.8				0.805	137
		2	-4.2	-2.7	2.4			0.872	
		3	-4.6	-2.8	2.1	1.1		0.885	
		4	-4.2	-2.5	1.7	0.95	-0.19	0.893	
	1 & 2	1	0.72	-2.8				0.793	246
		2	-3.7	-2.7	2.2			0.854	
		3	-3.5	-2.4	1.8		-0.19	0.867	
		4	-3.8	-2.5	1.7	0.86	-0.15	0.875	
log(TC/TC_o)	1	1	0.63	-2.2				0.616	110
		2	-2.5	-2.1	1.5			0.667	
		3	-2.6	-2.2	1.3	0.79		0.679	
		4							
	2	1	0.73	-2.5				0.713	138
		2	-2.8	-2.5	1.8			0.751	
		3	-3.2	-2.6	1.5	1.1		0.766	
		4							
	1 & 2	1	0.78	-2.5				0.690	248
		2	-2.5	-2.4	1.6			0.728	
		3	-2.8	-2.5	1.3	1.1		0.742	
		4							
log(FS/FS_o)	1	1	0.48	-2.2				0.682	107
		2	-0.37	-1.6			-0.24	0.748	
		3	-2.1	-1.6	0.89		-0.21	0.767	
		4							
	2	1	0.49	-2.3				0.820	132
		2	-2.6	-2.3	1.6			0.863	
		3	-2.4	-2.0	1.2		-0.15	0.871	
		4							
	1 & 2	1	0.54	-2.3				0.780	239
		2	-0.26	-1.8			-0.25	0.819	
		3	-2.1	-1.9	1.0		-0.18	0.837	
		4							
log(SPC/SPC_o)	1	1	0.76	-2.1				0.645	98
		2	-0.70	-2.2		1.7		0.703	
		3	-2.2	-2.1	-0.80	1.5		0.717	
		4							
	2	1	1.0	-2.5				0.745	123
		2	-2.9	-2.5	1.9			0.792	
		3	-3.8	-2.6	1.7	1.6		0.815	
		4	-3.5	-2.2	1.3	1.5	-0.19	0.824	
	1 & 2	1	0.98	-2.4				0.723	221
		2	-0.61	-2.5		1.9		0.763	
		3	-2.9	-2.4	1.3	1.6		0.786	
		4	-2.8	-2.2	1.1	1.4	-0.16	0.791	

*Regression equation (with one to four independent variables):

$\log (N/N_o) = b_o + b_1 \log x_1 + b_2 \log x_2 + b_3 \log x_3 + b_4 \log x_4$, or

$N/N_o = 10^{b_o}.x_1^{b1}.x_2^{b2}.x_3^{b3}.x_4^{b4}$

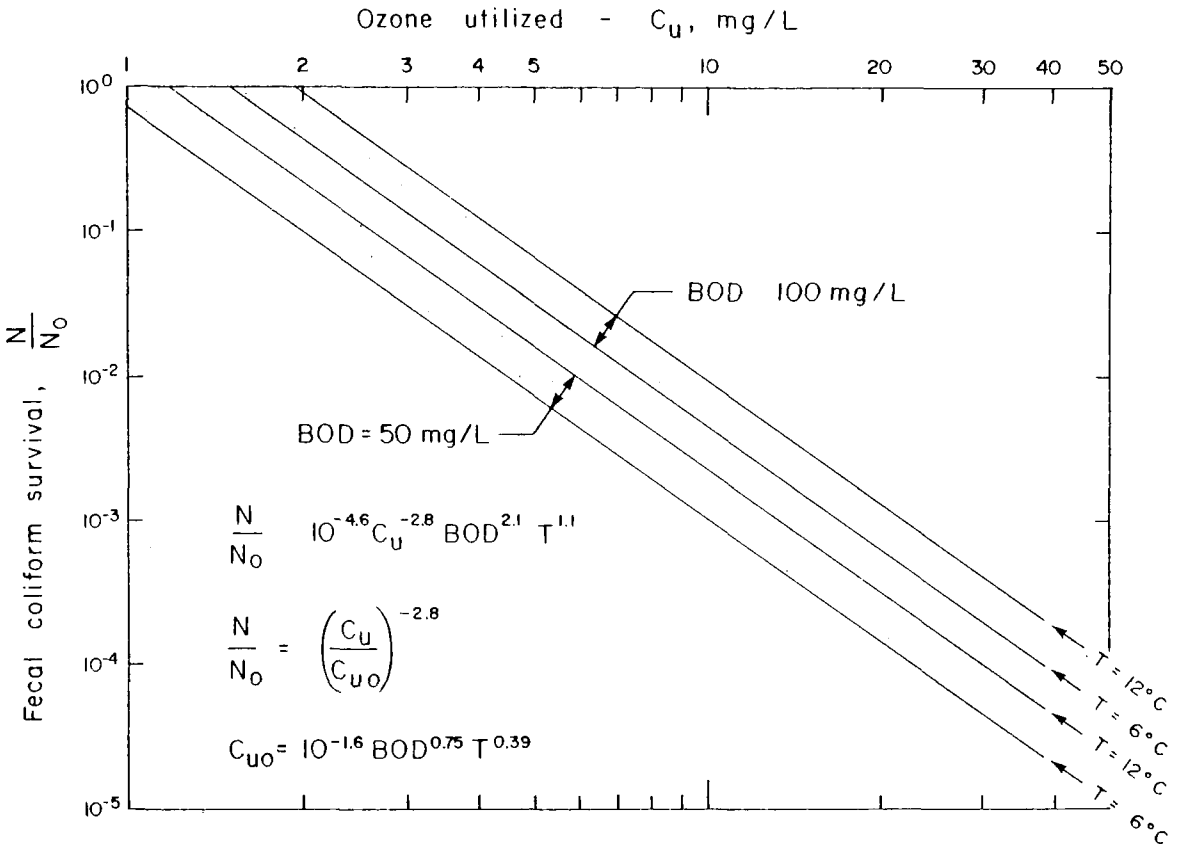


Figure 4. Apparent Effect of Ozone Utilized, BOD, and Temperature on Fecal Coliform Survival in Screened Wastewater

Comparisons of Fecal Coliform Survival Ratios for Other Types of Wastes

Data on fecal coliform survival for RBC effluent, anaerobic lagoon effluent and strong wastewater are shown together in Figure 5. The differences between the individual regression lines indicate the effect of the differences in wastewater characteristics.

The primary influence of wastewater characteristics on the FC survival curves appears to be on the initial ozone demand of the wastewaters, varying from 0.7 mg/L for the RBC effluent to 12.5 mg/L for the strong wastewater. In all cases, the initial ozone demands, C_{uo}, were significantly different at the 5% level.

The slopes of the regression lines varied from -2.9:1 for screened wastewater to -4.6:1 for lagoon effluent. This difference was highly significant (at the 0.1% level), although wastewater characteristics for screened wastewater and lagoon effluent were similar with respect to BOD and suspended solids. In comparing the screened wastewater with the RBC effluent and with the high strength wastewater, no significant differences in the slopes of the regression lines were observed. However, it should

be noted that only 13 data points were obtained for the RBC effluent and 16 points for the high strength wastewater. With additional data, significant differences between all of the slopes may have been found.

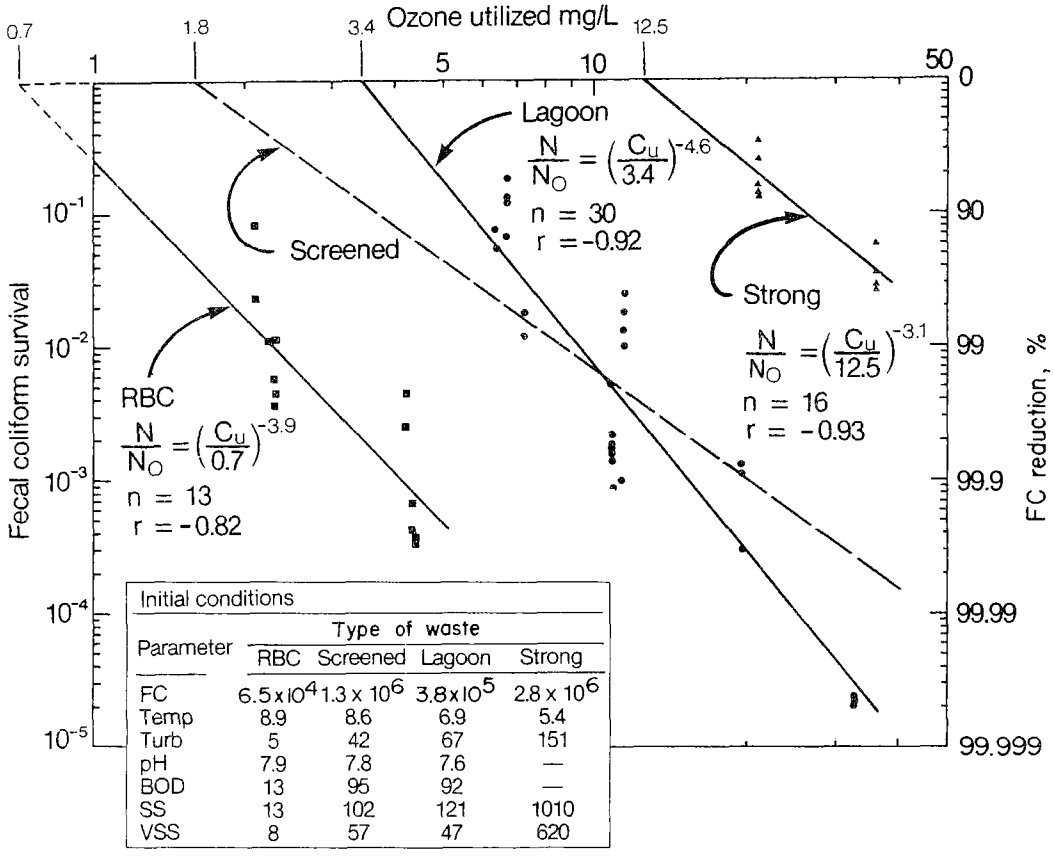


Figure 5. Fecal Coliform Survival for RBC Effluent, Anaerobic Lagoon Effluent, and Strong Wastewater (Screened Wastewater Shown for Comparison)

On-Line Operating Variables

The previously developed regression equations provide insight into the effects of various factors on disinfection; however, they have little practical significance for on-line operation of a system. The reason for this is that a number of the relevant independent variables cannot be monitored instantly. Therefore, the operator could not provide accurate on-line control over the ozone disinfection process in response to changing wastewater characteristics.

To examine this problem from the operator's point of view, data analyses were undertaken using only those variables which could be monitored on-line. Log effluent fecal coliform number (log FC) was selected as the dependent variable. Log transformations of ozone utilized, effluent ozone residual (log C_{re}), wastewater turbidity (log TURB) and wastewater temperature (log T) were selected as the independent variables. The results of these data analyses are summarized in Table 3.

Table 3. Summary of Regression Analyses for log FC in Screened Wastewater*

CASE	STEP	REGRESSION COEFFICIENTS FOR					r^2	n
		b_o	log C_u	log T	log (TURB)	log C_{re}		
1 Column	1	7.0	-2.8				0.757	85
	2	5.1	-2.9	2.0			0.816	
	3	4.1	-2.9	1.7	0.84		0.833	
	4	3.6	-2.3	1.4	0.91	-0.15	0.847	
2 Columns	1	7.0	-2.9				0.799	105
	2	5.3	-2.0			-0.46	0.863	
	3	3.7	-2.2		1.3	-0.35	0.892	
	4	2.9	-2.4	1.5	1.1	-0.26	0.908	
1&2 Columns	1	7.0	-2.9				0.791	190
	2	4.8	-3.0	2.3			0.848	
	3	3.4	-3.0	1.9	1.2		0.873	
	4	3.2	-2.5	1.5	1.1	-0.20	0.888	

* Regression equation (with one to four independent variables):

$\log FC = b_o + b_1 \log x_1 + b_2 \log x_2 + b_3 \log x_3 + b_4 \log x_4$, or

$FC = 10^{b_o} \cdot x_1^{b1} \cdot x_2^{b2} \cdot x_3^{b3} \cdot x_4^{b4}$

All four of the independent variables proved to be significant in the equations; however, use of only two of these variables, C_u and C_{re}, would result in reasonably good accuracy, indicated as follows:

$$FC \text{ (per 100 mL)} = 10^{5.3}\ C_u^{-2.0}\ C_{re}^{-0.46} \qquad (\text{with } r^2 = 0.86)$$

It should be noted that this predictive equation is only applicable to ozonation of screened wastewater in this study. The regression coefficients would likely be different for other wastewater types.

BOD and Suspended Solids Reductions

BOD and suspended solids reductions were achieved by ozonation of the screened wastewater. The results for BOD reduction are shown in Figure 6. Suspended solids reductions were similar but had more scatter in the results. BOD and suspended solids were reduced approximately 25% at an ozone utilization of approximately 30 mg/L.

In a separate evaluation, screening resulted in negligible BOD reduction and about 10% suspended solids reduction. Thus, overall BOD and suspended solids reductions, achievable by screening followed by ozonation, would be in the order of 25% and 30%, respectively, at an ozone utilization of approximately 30 mg/L.

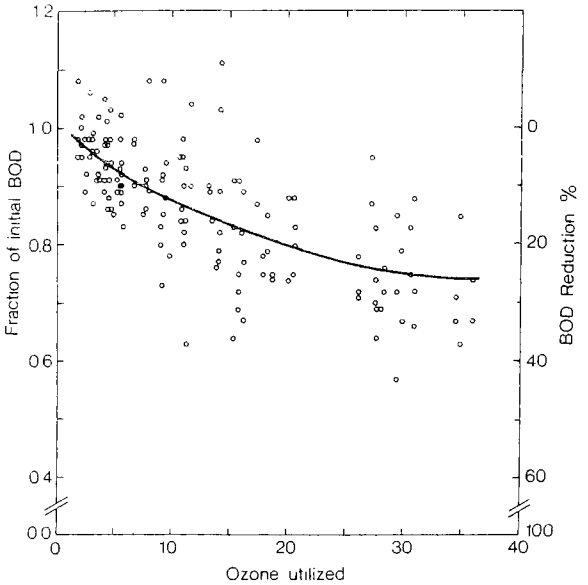


Figure 6. BOD Reduction for Ozonated, Screened Wastewater

Analysis of Ozone Transfer Efficiency

Ozone transfer efficiency may be expressed as a ratio of ozone utilized to ozone applied (C_u/C_a), or as a percent ($100\ C_u/C_a$). Because of the demonstrated effect of ozone utilization on bacterial reduction, ozone transfer efficiency for a contactor is very important. Therefore, analyses of relevant factors associated with ozone transfer efficiency (or utilization efficiency) were undertaken for the 5 m high ozone contact columns.

Stepwise multiple linear regression analyses for ozone utilization efficiency are summarized in Table 4. For the first contact column and the over-all system, factors appearing to affect ozone utilization efficiency, in order of importance, were the ozone-oxygen gas flowrate, Q_g (or ozone-oxygen gas loading rate or flowrate per unit contacting volume, Q_g/V); ozone applied, C_a; and wastewater strength, BOD. However, the variables and order of importance changed for the second contact column due to the ozone residual entering that column from the first column. The factors in order of importance were then influent ozone residual, C_{ri}; gas loading rate, Q_g/V; and wastewater strength, BOD. Certainly, other factors could also have an effect on ozone transfer efficiency, particularly where conditions differ from those examined in this study.

Figure 7 shows decreasing transfer efficiency with increasing gas loading rate (data for one and both columns together). Considerable scatter in the data points is apparent. Some of this scatter can be accounted for by considering other factors affecting efficiency, as illustrated in Figure 8. A similar type of plot is shown for the second contact column in Figure 9. Analysis of the dissolved oxygen concentration in the effluent from the system indicated high values as expected when oxygen is used as the feed gas for the ozonator.

Table 4. Summary of Regression Analyses for Ozone Utilization Efficiency of Ozone Contact Columns with Screened Wastewater

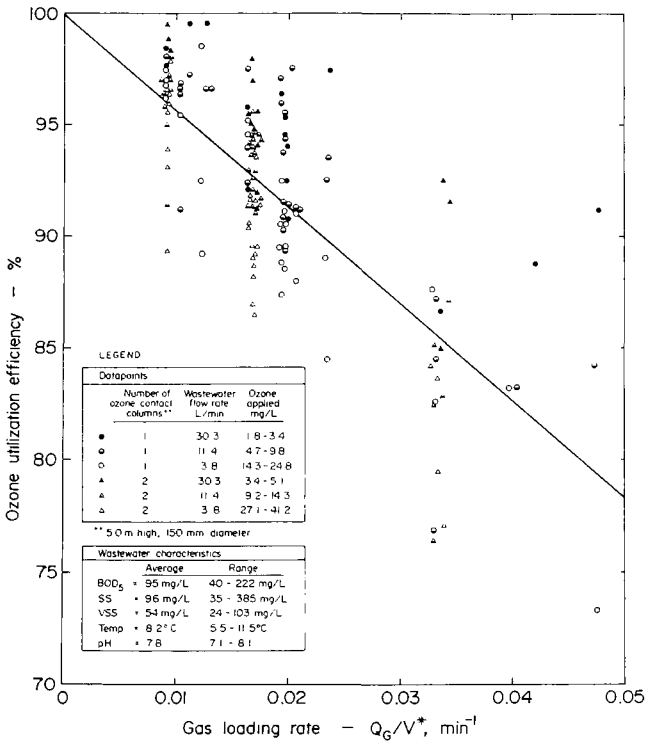
$$C_u/C_a = 1.0 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4$$

CASE	STEP	REGRESSION COEFFICIENTS FOR				r^2	n
		Q_g/V	C_a	BOD	C_{ri} (1)		
1 Column	1	-4.0				0.554	152
	2	-3.2	-0.0018			0.666	152
	3	-4.5	-0.0023	0.00039		0.724	152
2 Columns	1	-4.7				0.603	152
	2	-3.7	-0.0011			0.741	152
	3	-3.3	-0.0014	0.00039		0.798	152
1 & 2 Columns	1	-4.3				0.554	304
	2	-3.5	-0.0013			0.692	304
	3	-4.8	-0.0017	0.00038		0.749	304
Column 2 (2)	1	-7.1				0.121 (3)	152
	2	-4.8	-0.0053			0.188 (3)	152
	3	---	---	---		---	---
Column 2	1	---	---	---	-0.35	0.543	146
	2	-5.0	---	---	-0.26	0.759	146
	3	-6.3	---	0.00030	-0.27	0.775	146

(1) C_{ri}, ozone residual of column influent, only significant for Column 2
C_{re}, ozone residual of column effluent, not significant for any case

(2) C_{ri} not included in analysis

(3) Poor correlation



* Q_G = O_3-O_2 gas flow rate at STP (25°C and 760 mm Hg)
V = volume of column(s) (90 L for one column; 180 L for two columns)

Figure 7. Ozone Utilization in Screened Wastewater

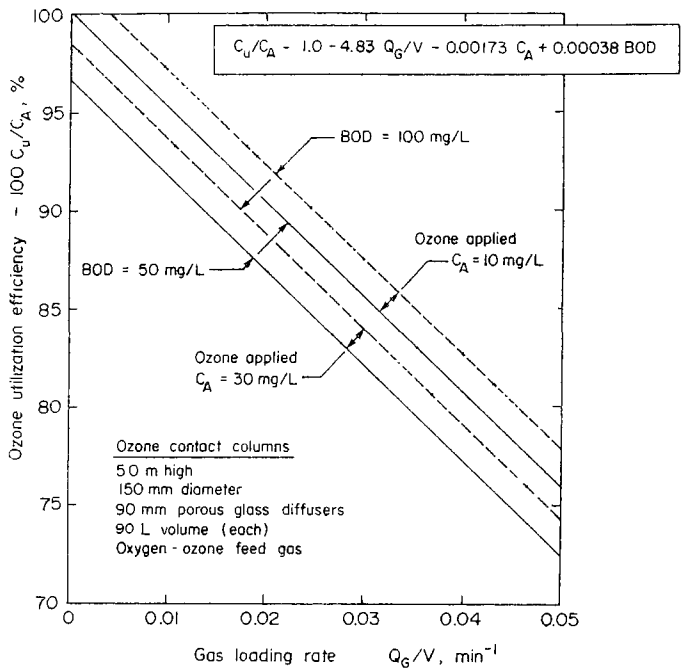


Figure 8. Factors Affecting Ozone Utilization with One or Two Contact Columns

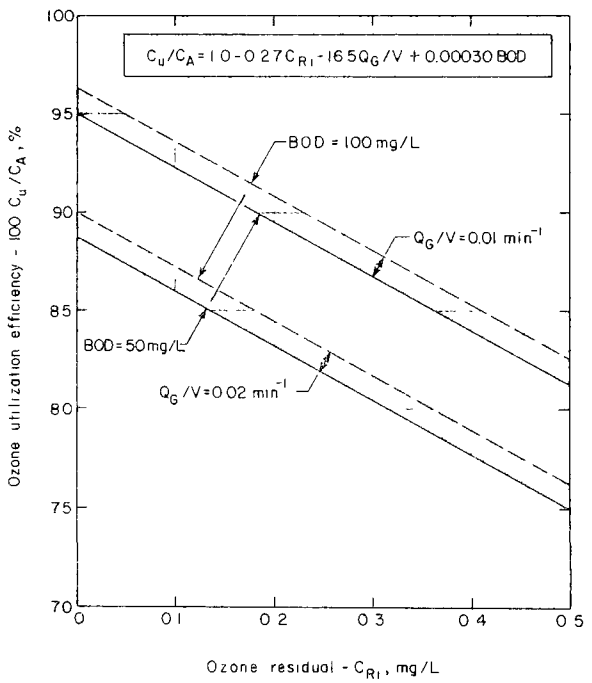


Figure 9. Factors Affecting Ozone Utilization with the Second Contact Column

CONCLUSIONS

Based on the ozone pilot plant studies, the following conclusions are applicable for screened, dilute wastewater, unless otherwise noted:

1. The principal factor influencing indicator organism survival was the amount of ozone utilized in the wastewater.

2. A log-log relationship existed between bacterial survival and ozone utilized. After transformation from the log to the power form of the equation, the relationship was expressed as:

$$N/N_o = (C_u/C_{uo})^b$$

3. Different indicator organisms demonstrated different degrees of sensitivity to ozone. To achieve a three-log reduction (10^{-3} survival ratio) of the indicator organisms, the following effluent numbers were reached at the noted ozone utilization levels:

FC = 1,200 per 100 mL at C_u = 20 mg/L
TC = 5,800 per 100 mL at C_u = 31 mg/L
FS = 130 per 100 mL at C_u = 33 mg/L
SPC = 180 per mL at C_u = 40 mg/L

4. Adverse wastewater characteristics (high BOD, turbidity, etc.) adversely influenced the effectiveness of the ozone disinfection system.

5. Other factors sometimes influencing bacterial survival in conjunction with ozone disinfection appeared to be wastewater temperature and ozone residual. Disinfection efficiency usually improved with decreasing wastewater temperature and increasing ozone residual.

6. The predictive equations that were developed for ozone disinfection of screened wastewater may not be directly applicable to other types of wastewater. Nevertheless, it is hypothesized that the general approach to data analysis presented in this paper would be applicable to other systems with different wastewater characteristics. This could lead to improved understanding of relevant factors influencing ozone disinfection and to improved ozone system operations.

7. It was demonstrated that effluent fecal coliform numbers can be predicted quite reliably with the assistance of on-line monitoring of certain variables, for example:

$$FC \text{ (per 100 mL)} = 10^{5.3}\ C_u^{-2.0}\ C_{re}^{-0.46} \qquad (r^2 = 0.86)$$

8. Ozonation of RBC secondary effluent was much more effective than ozonation of screened wastewater. The 0.7 mg/L initial ozone utilized (approximate value from the extrapolated curve) was less than half that for screened wastewater. Also, a three-log reduction of FC

(from 65,000 to 65 per 100 mL) was achieved at approximately 4 mg/L of ozone utilized. This amount of ozone was 20% of that for the screened wastewater at the same three-log reduction. The fact that the RBC effluent initially was of superior bacterial quality (lower FC) further served to accentuate the difference in effluent FC numbers after ozone disinfection.

9. BOD and suspended solids reductions of approximately 25% were achieved at an ozone utilization of 30 mg/L.

10. The ozone transfer or utilization efficiency with one contact column and with the overall system appeared to be influenced by the ozone-oxygen gas flowrate (Q_g or Q_g/V), the amount of ozone applied (C_a), and the wastewater strength (BOD).

11. The ozone transfer efficiency of the second contactor in series appeared to be influenced by the ozone residual in the wastewater from the first column (C_{ri}), the gas flowrate (Q_g or Q_g/V), and the wastewater strength (BOD).

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11. OZONE-MASS TRANSFER COEFFICIENTS FOR BUBBLE DIFFUSER

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ABSTRACT

A pilot plant project was conducted at the US EPA Test & Evaluation Facility (T&E) to evaluate ozone as a disinfectant for wastewater treatment. Various contacting devices were compared and the bubble diffuser proved to be the most cost effective contactor (4). The data generated in conducting the investigation on the effectiveness of the various contactors were used to calculate the overall gas mass transfer coefficient for ozone. The calculation consisted of dividing the overall ozone mass transfer rate by the ozone gas concentration driving force across the bubble contactor. This paper details the method that was used to calculate the overall ozone mass transfer coefficient and the effect that gas flow rates had on the ozone mass transfer coefficient. The paper also presents results that show the enhancement of the mass transfer coefficient by the impurities in wastewater

MATERIALS AND METHODS

Secondary Effluent

Secondary effluents were trucked into the pilot plant from six secondary treatment facilities located within a 32 km radius of the pilot plant. The influent flow to the diffuser was normally controlled at 75 ℓ/min, although experimental runs were made at various liquid flow rates to determine the effect of liquid flow on gas mass transfer coefficients.

Ozone Generation

The ozone generator had a maximum capacity of 10 kg/d using oxygen as the feed gas. The concentration of ozone was controlled by power and flow to obtain the dose specified for the experiments. The oxygen pressure was reduced to 60 kPa before entry into the ozonator where two air cooled units, that contained six plates per unit, were used to convert the oxygen to ozone. The power was varied between 200 and 1500 watts to obtain the ozone concentration specified for an experimental run. An ultraviolet absorption analyzer (Dasibi Environmental Corp., Glendale, California) was used to monitor the ozone concentration continuously and to establish steady state conditions.

Bubble Diffuser Contactor

The bubble diffuser contactor (Figure 1) consisted of three aluminum columns, each 3.7 m high and 300 mm in diameter, connected in series by PVC piping. The three columns were arranged in steps so that secondary effluent could flow by gravity from the first through the third column. The ozone

gas stream was split and the flow controlled by rotameters leading to each of the three columns. The ozone enriched gas was injected through a domed ceramic diffuser (Norton Chemical Process Products Division) at the bottom of each column. The flow was generally split with 50 percent of the gas being fed to the first column and 25 percent each to the second and third columns. The liquid residence time was approximately 3 minutes in each contactor for a total contact time of 9 minutes.

Sampling

Gaseous ozone concentrations were determined at the inlet and outlet from the diffuser columns using the iodometric method of Birdsall, Jenkins, and Spadinger (2). A wet test meter was used to measure gas volumes. The effluent leaving each column was measured for residual ozone using the Amperometric Back Titration Method.

RESULTS AND DISCUSSION

Ozone Transfer

The ozone transferred from the gas stream to the effluent was determined from a mass balance on gaseous ozone, as shown in Figure 2. The ozone balance on the liquid stream includes a reaction factor to equate with the ozone balance on the gas stream because the absorbed ozone can either react with substances in the liquid or undergo decay. This assumption is shown as ΔK.

Mass balance equations are given below:

$N - G_1(y_1-y_2) = L_2 (x_1-x_2) + \Delta K$

N = Ozone transferred, mg/min

y_1 = Inlet ozone concentration, mg/ℓ

y_2 = Outlet ozone concentration, mg/ℓ

$G_1 \approx G_2$ Gas flow (Oxygen), ℓ/min

x_1 = Outlet residual ozone concentration, mg/ℓ

x_2 - Inlet residual ozone concentration, mg/ℓ

L_1 L_2 Liquid flow, ℓ/min

ΔK Ozone consumed, or decayed mg/min

The ozone transferred can readily be calculated by the mass balance in the gas phase [i.e., $G_1(y_1-y_2)$]. An ozone mass balance in the liquid phase can be used to determine the quantity of ozone that reacted with the constituents in the influent and/or underwent decay, ΔK.

Experiments were conducted at various gas flow rates, G, to obtain the effect of gas velocity on mass transfer coefficients. The ozone concentration was also varied to obtain various ozone dosages at a specific gas flow rate. High ozone dosages were essential to calculate the gas mass transfer coefficients accurately, because a low off-gas concentration may indicate that the column is taller than required.

Gas Mass Transfer Coefficient Calculations

The method used to calculate mass transfer coefficients is summarized in Figure 3.

The ozone transferred is equal to

$$N = G\ (y_1-y_2)$$

This in turn is equal to the product of the overall gas mass transfer coefficient (K_gVa) and the log mean concentration difference between the inlet and outlet gas, (ΔC_{lm}). The overall ozone mass transfer coefficient is a measure of the rate at which a contactor can transfer ozone from the gas phase into the liquid phase. The log mean concentration difference is the driving force necessary to transfer the ozone from the gas phase into the liquid phase.

$$N = G\ (y_1-y_2) = (K_gVa)(\Delta C_{lm}).$$

The log mean concentration difference is calculated from the following equation, which uses the inlet and outlet conditions of both the gas and liquid phases.

$$\Delta C_{lm} = \Delta y_{lm} = (y_1-y_1^*) - (y_2-y_2^*)/\ell n\ [(y_1-y_1^*)/(y_2-y_2^*)]$$

Where

$y_1^* = Hx_1$ = equilibrium concentration at x_1 (bottom of the column)

and H is Henry's constant in (mg $O_3/\ell g$)/(mg O_3/ℓ_{liq}).

The value for Henry's constants was obtained by converting the published values in the International Critical Tables (ICT) into units that are conducive for calculating overall gas mass transfer coefficients.

$H_{20\ ^oC}$ = 2.86 x 10^6 mm Hg/mol fraction of ozone = 2.63 mg $O_3/\ell g$/mg O_3/ℓ_{liq}.

[To convert H into units that can be readily used, assume an ozone residual and multiply by H obtained in the ICT at a specific temperature to obtain the partial pressure (pp) of ozone in mm·Hg. Divide by 760 mm·Hg (1 atm) to obtain gaseous mol fraction. Convert mol fraction of ozone into mg $O_3/\ell g$.

Divide this value by the assumed ozone residual to obtain the H value in (mg $O_3/\ell g$)/(mg O_3/ℓ_{liq}) at the specified temperature. Repeat at various temperatures to obtain the curve shown in Figure 4.]

With the values of H obtained at several temperatures, a curve can be constructed covering the temperature range (2 to 40 °C) normally encountered at wastewater treatment plants.

Example of Gas Mass Transfer Calculation

Below is an example showing the method employed to calculate the ozone mass transfer coefficient from the data obtained at the T&E facility, using effluent from the Loveland, Ohio wastewater treatment plant.

The ozone transferred to the liquid from the gas in columns A, B and C were:

N_a $G_a(y_{1a}-y_{2a})$ = 16.6 (27.9-2.1) - 428 mg O_3/min

N_b $G_b(y_{1b}-y_{2b})$ = 8.3 (27.9-6.0) 182 mg O_3/min

N_c $G_c(y_{1c}-y_{2c})$ = 8.3 (27.9-7.1) = 173 mg O_3/min

The equilibrium partial pressure of ozone is calculated at the bottom of column A in terms of mg O_3/lg rather than mm Hg. The ozone residual was 2.3 mg/ℓ_{liq}. At 16.5 °C the H value is 2.2 (mg $O_3/\ell g$)/(mg O_3/ℓ_{liq}), which is multiplied by the residual, 2.3 mg/ℓ_{liq}, to obtain 5.1 mg/ℓ_{liq} as the equilibrium gas concentration of ozone at the bottom of the column. However, the values of both the incoming ozone gas and the equilibrium gas concentration at the bottom of the column need to be corrected because of the increased pressure caused by the water head of 3.2 m. The incoming ozone gas concentration, in reality the ozone partial pressure, is multiplied by the pressure correction factor of (10.4 + 3.2)/10.4 = 1.31. The inlet ozone partial pressure, or the effective ozone concentration, is 1.31 x 27.9 or 36.5 mg $O_3/\ell g$. The equilibrium ozone partial pressure for the ozone residual at the bottom of the column (2.3 mg/ℓ_{liq}) is 5.1 mg/ℓg. This value also requires correction because of the increased pressure at the bottom of the column. However, in this case the correction factor is less than one because the partial pressure of ozone is determined by the ozone residual. The ratio for the equilibrium concentration is 10.4/(10.4 + 3.2) = 0.765. The corrected equilibrium ozone concentration $y_1{}^*_c$ is 0.765 x 5.1 = 3.9 mg/ℓg. The concentration or partial pressure driving force at the bottom of column A is $y_1-y_{1c}{}^*$ = 36.5 - 3.9 = 32.6 mg/ℓg.

At the top of the column the liquid influent has no residual ozone so the driving force is only the concentration of the effluent gas, 2.1 mg/ℓg. Since the top of the column is at ambient pressure there is no correction factor applied to the ozone concentration leaving the column. The ozone concentration driving force over the entire column is the log mean concentration difference of the two end conditions.

$$\Delta C_{lm} = \Delta y_{lm} = \frac{\Delta y_1 - \Delta y_2}{\ln \frac{\Delta y_1}{\Delta y_2}} = \frac{(y_{1c} - y_{1c}^*) - (y_2 - 0)}{\ln \frac{y_{1c} - y_{1c}^*}{y_2 - 0}}$$

$$= \frac{32.6 - 2.1}{\ln \frac{32.6}{2.1}} = \frac{30.5}{\ln 15.5} = 11.1 \text{ mg } O_3/\ell g$$

and finally the ozone mass transfer coefficient is obtained by

$$N = (K_g Va)(\Delta y_{lm})$$

$$K_g Va = \frac{N}{\Delta y_{lm}} = \frac{428 \text{ mg } O_3/\text{min}}{11.1 \text{ mg } O_3/\ell g} = 38.6 \text{ mg } O_3/\text{min/unit concentration difference}$$

The calculation for columns B and C follow closely the method employed for column A. The only difference is that the liquid influent to columns B and C contain an ozone residual that must be taken into consideration at the top of the column to obtain the ozone concentration driving force, or partial pressure difference. The $K_g Va$ for column B was 20.8 mg O_3/min/unit concentration difference and for column C the $K_g Va$ was 17.0 mg O_3/min/unit concentration difference.

Mass Transfer Coefficients for Five Plants

The major objective of the pilot plant study with the bubble diffuser was to establish the ozone requirements needed to disinfect secondary effluent from six treatment plants. During these experimental runs, data were obtained that enabled the ozone mass transfer coefficients to be calculated. The data used to determine the mass transfer coefficients included only those results in which the effluent gas, y_2, was greater than 0.7 mg/ℓg. The Dasibi analyzer failed to show a measurable change when the effluent gas concentration was below 0.7 mg/ℓg.

The mass transfer coefficients obtained for the five plants are shown in Table 1.

TABLE 1. Mass Transfer Coefficients at Column A from Five Plants at a G of 17 ℓ/min

Plants	$K_g Va$	n	σ
Indian Creek	44	5	1.5
Muddy Creek	44	5	1.3
Loveland	41	8	2.3
Sycamore	39	15	3.0
Fairfield	39	10	3.7
Mill Creek	--	0	-
Mean	40	43	2.1

The mean for the 43 samples was 40 mg O_3/min/unit concentration difference. It should be noted that the mass transfer coefficient at Mill Creek could not be calculated because the highest y_2 value measured in 25 runs was equal to or less than 0.1 mg/ℓg.

Effect of Gas Flow on Mass Transfer Coefficients

Experimental runs were conducted with Indian Creek and Muddy Creek secondary effluents to obtain mass transfer coefficients at various gas flows. A plot of these data is shown in Figure 6. The rate of increase declined as the gas flow increased and it appears that very little, if any, improvement in mass transfer coefficient can be expected at gas flows above 30 ℓ/min.

Enhancement of Mass Transfer by Secondary Effluents

The bubble diffuser was operated on tap water to obtain mass transfer coefficients that were not influenced by ozone demanding material. Figure 6 shows the rate of increase was considerably less than that for secondary effluent. The mass transfer coefficient leveled off at gas flow rates above 25 ℓ/min. The difference in the mass transfer coefficients between tap water and secondary effluent can be attributable to the ozone demand of the secondary effluent, which resulted in a signficantly higher mass transfer coefficient.

Enhancement in Partially Satisfied Effluents

The bubble diffuser employed at the T&E Facility consisted of three columns. The effluent exited at the bottom of the third column. The ozone

enriched gas was split among the three columns, with 50% of the gas being fed to column A and 25% of the gas being fed to columns B and C. The mass transfer coefficient in column B was 34 at a gas flow rate of 16 ℓ/min and 29 in column C as compared to 40 in column A at a gas flow of 17 ℓ/min (Table 1).

A summary of these results is shown in Table 2.

TABLE 2. Mass Transfer Coefficients for Columns B and C

Plant	Column B				Column C			
	KgVa @ 8.3 ℓ/min	(n)	KgVa @ 16 ℓ/min	(n)	KgVa @ 8.3 ℓ/min	(n)	KgVa @ 16 ℓ/min	(n)
Sycamore	19	(15)	28	(2)	19	(15)	24	(2)
Fairfield	19	(9)	--	(0)	19	(12)	33	(1)
Loveland	21	(3)	38	(2)	20	(5)	32	(2)
Muddy Creek	19	(3)	--	(0)	17	(3)	35	(1)
Indian Creek	20	(1)	38	(1)	20	(2)	28	(1)
Mean	19	(31)	34	(5)	19	(37)	29	(7)
Mill Creek	--	(0)			26	(4)		

A series of experimental runs was conducted specifically for obtaining mass transfer coefficients in columns B and C at gas flow rates other than the flow rates used in the disinfection study. Column A was operated at the normal 17 ℓ/min to repeat the partial ozone satisfaction that prevailed during the disinfection experimental runs. The mass transfer coefficients for columns B and C were plotted against gas flow and again the coefficients increased, up to a plateau region, with increasing gas flow (Figure 7). When the coefficients are displayed with the coefficients from column A for secondary effluent and for tap water (Figure 8), the curve for column B lies under column A and the curve for column C lies slightly above tap water. These relative positions for the mass transfer coefficient for columns B and C, where they lie between column A and tap water, provide further evidence on the enhancement of the mass transfer coefficient by secondary effluent when compared with tap water.

Effect of Liquid Flow Rate

The liquid flow rate for secondary effluents and tap water was varied from 56 to 94 ℓ/min to determine the effect on the mass transfer coefficient.

The results displayed in Figure 9, showed that the mass transfer coefficients increased with increasing liquid rates, up to a plateau region, and that secondary effluent showed a similar enhancement in the mass transfer coefficient when compared with tap water. These results were expected since increasing the liquid flow rate increased the system's capacity to absorb additional ozone and thus improve ozone mass transfer.

Summary

The results from this study indicate that the secondary effluents from 5 of the 6 plants gave similar mass transfer coefficients. The mass transfer coefficients for ozone that were obtained at these plants can be used for the design and scale-up of ozone bubble diffuser contactors at plants where the primary source of the wastewater is of domestic origin. The coefficients should not be used where the source is primarily of industrial origin. The high absorptive capacity obtained from the Mill Creek effluent indicates that the enhancement of the transfer coefficient is considerably greater than the coefficient obtained from effluents of domestic origin.

This paper presents a method for calculating ozone mass transfer coefficients using steady state conditions in a bubble diffuser contactor. Although the calculations are tedious, the mass transfer coefficients provide the designer with the necessary tools to determine the effect of variables on ozone transfer, such as the effect of increasing the height of the contactor, or operating the contactor under pressure. The mass transfer coefficient allows the designer to optimize the bubble diffuser contactor without relying upon pilot plant operational data to determine contactor sizing and performance.

The relationship on the ozone mass transfer coefficients with increases in either gas or liquid rates shows a levelling off of the increase in the mass transfer coefficient as the gas or liquid flow rate increases. The designer can now employ these results to determine the rate of transfer at various flow rates and calculate the efficiency of ozone transfer at any specific rate.

Finally, the paper presents evidence on the enhancement of the ozone mass transfer coefficient by secondary effluent when compared with tap water, and the reduction in the enhancement when the effluent has undergone partial ozone demand satisfaction.

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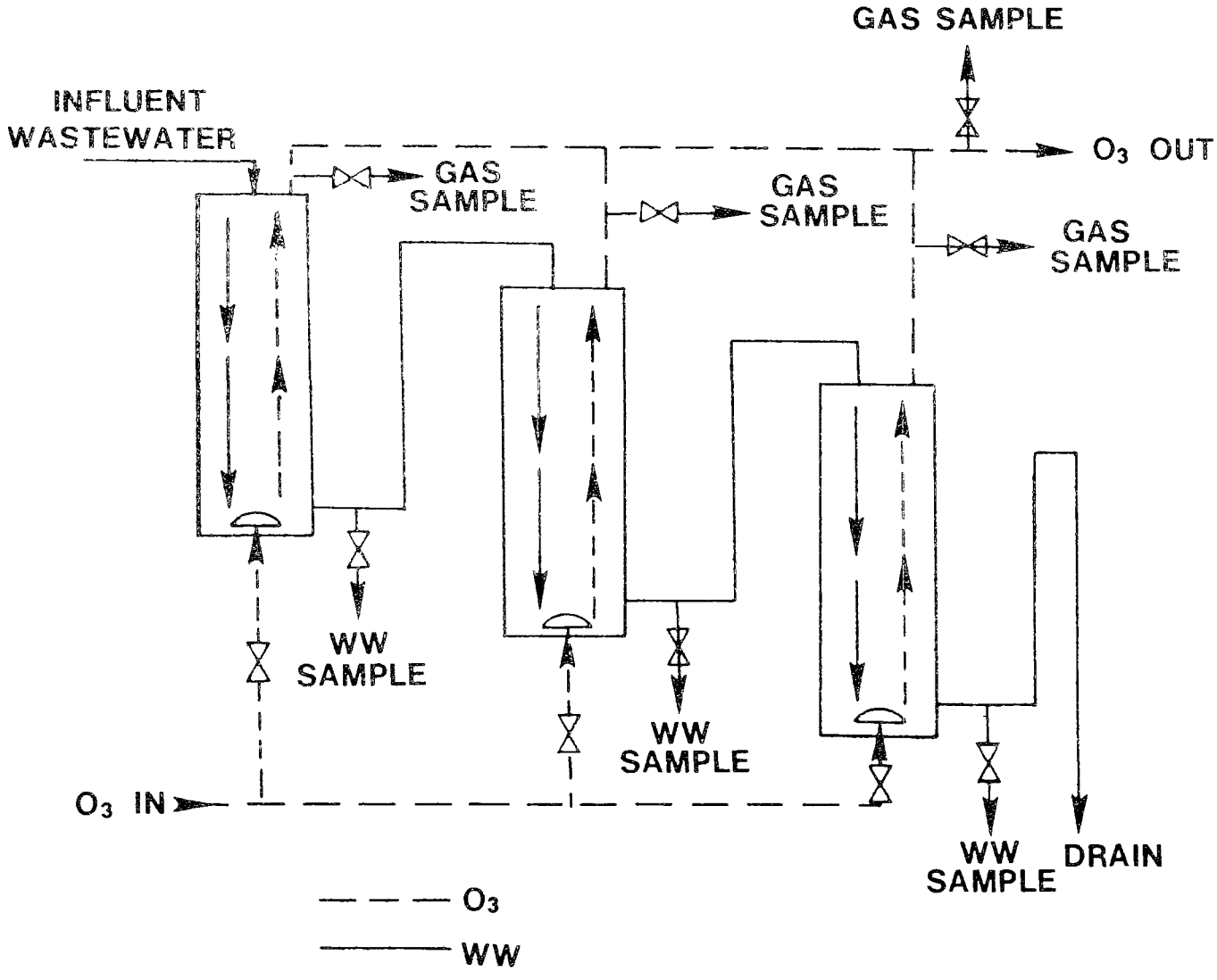

FIGURE 1. BUBBLE DIFFUSER OZONE CONTACTOR

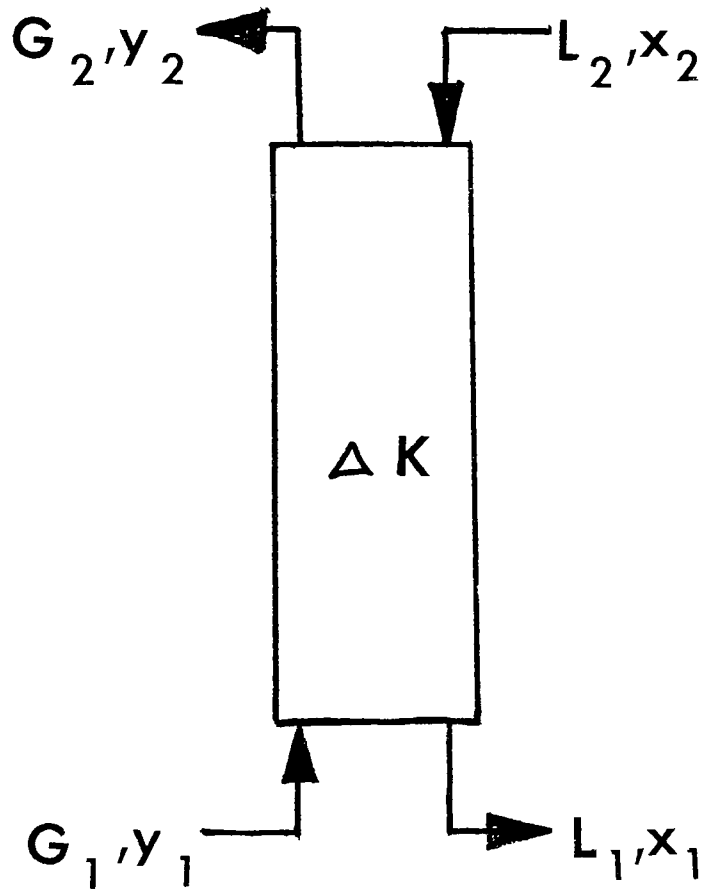

Figure 2

Ozone Mass Balance

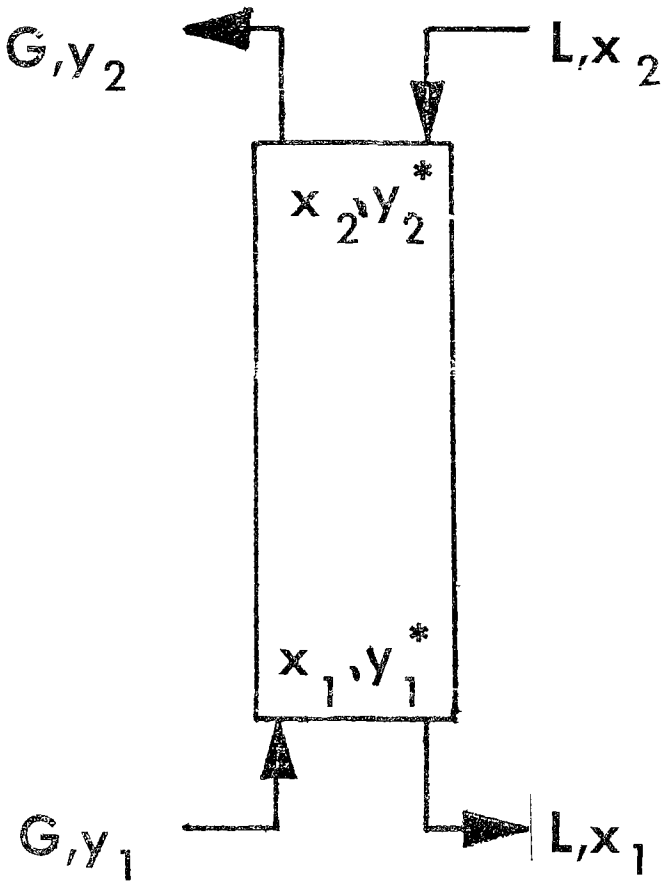

Figure 3
Mass Transfer Coefficients

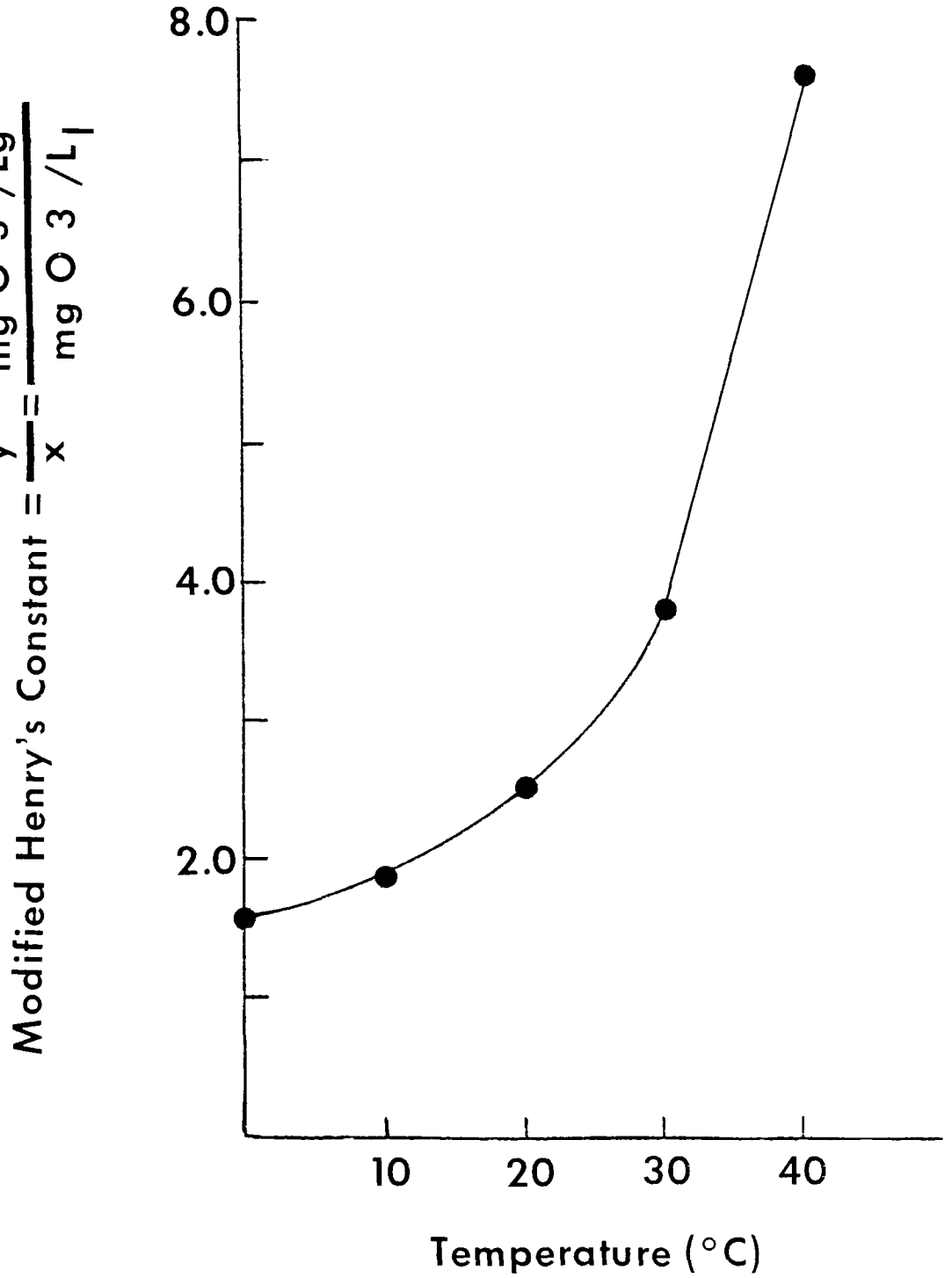

Figure 4. Effect of Temperature on Henry's Constant

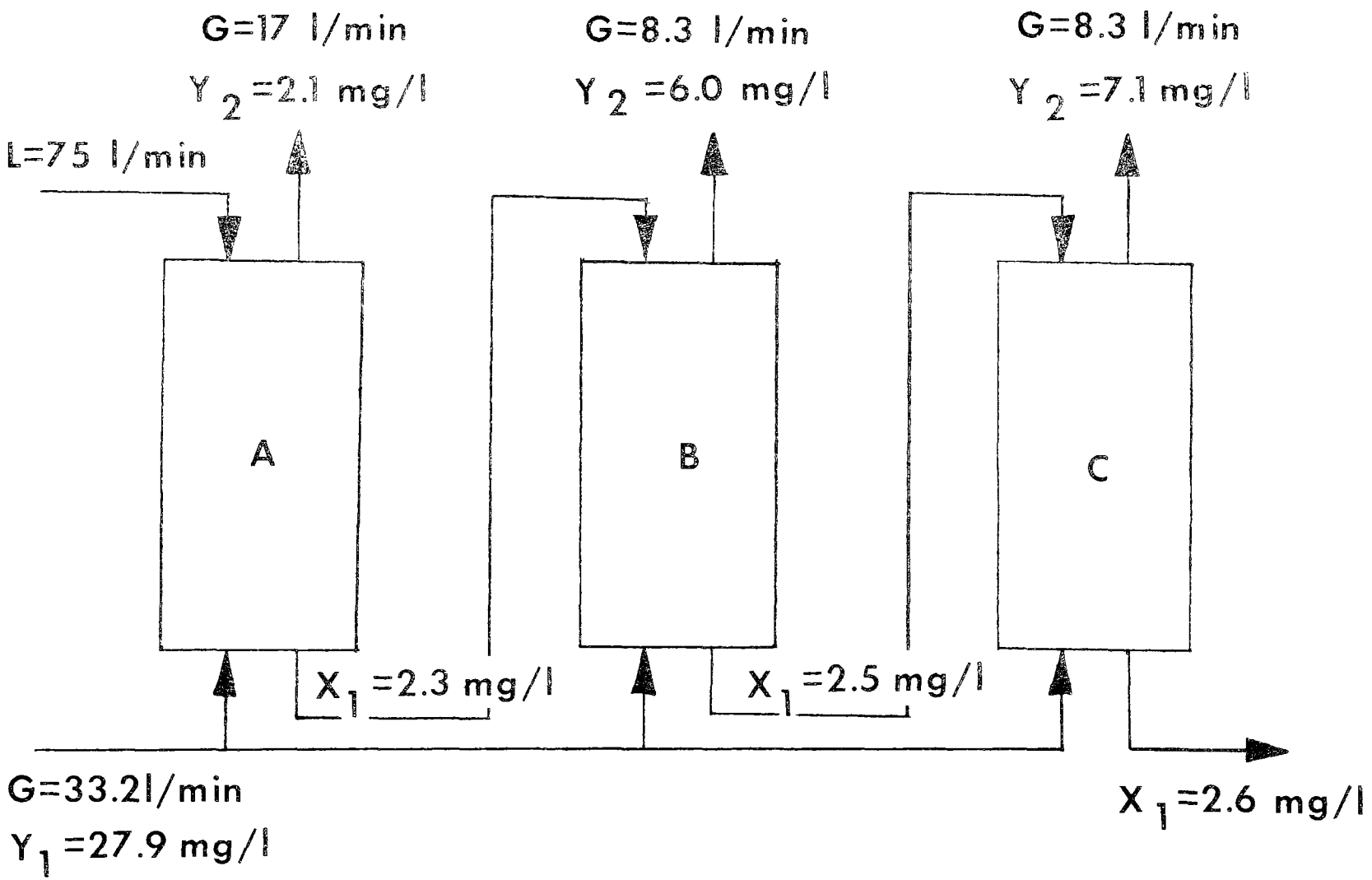

Figure 5. Ozone balance using Loveland effluent

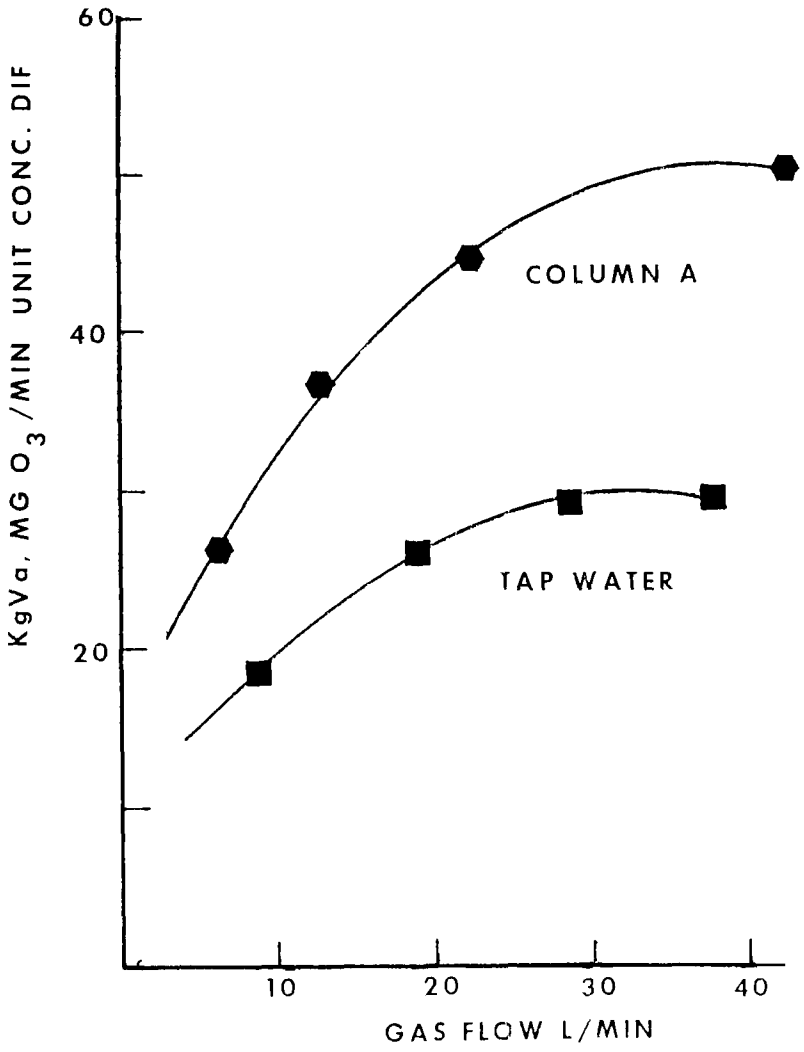

FIGURE 6

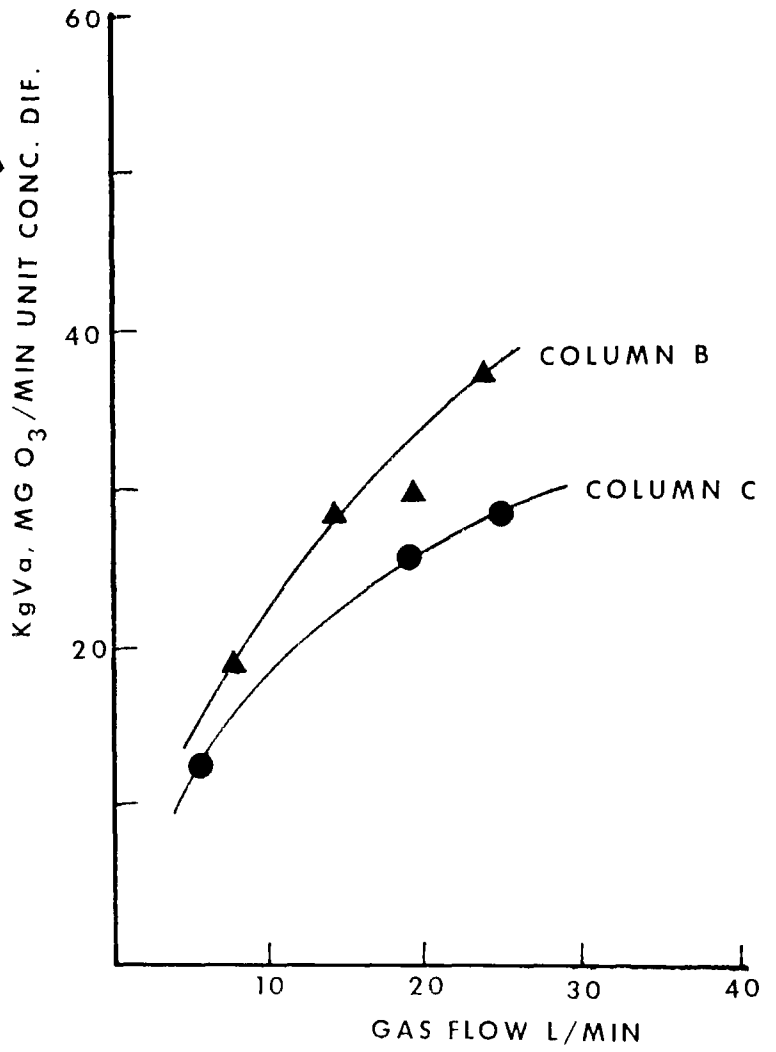

FIGURE 7

Figures 6 and 7. Effect of Gas Flow on Mass Transfer Coefficients

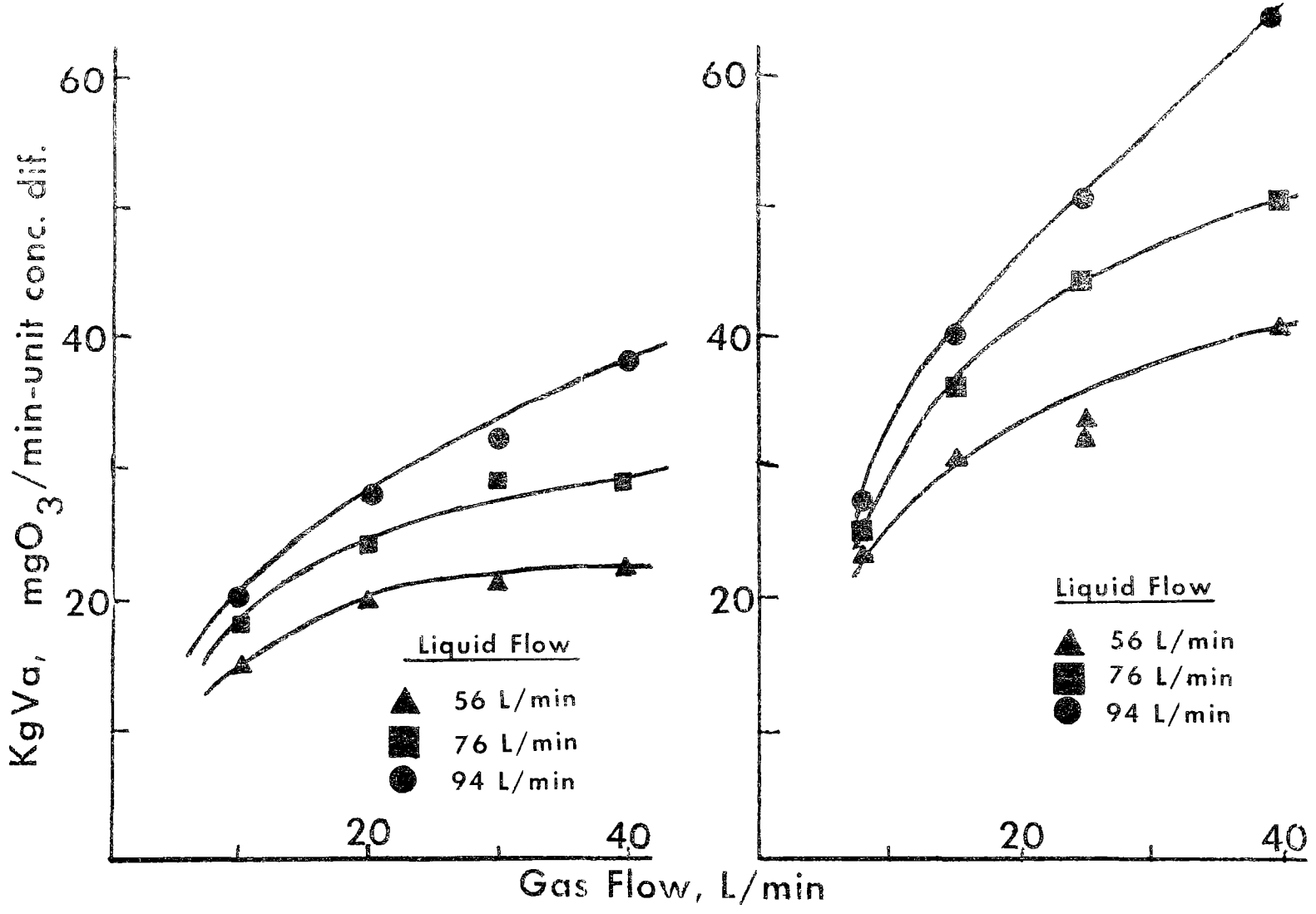

Figure 8. Effect of Gas Flow on Mass Transfer Coefficients

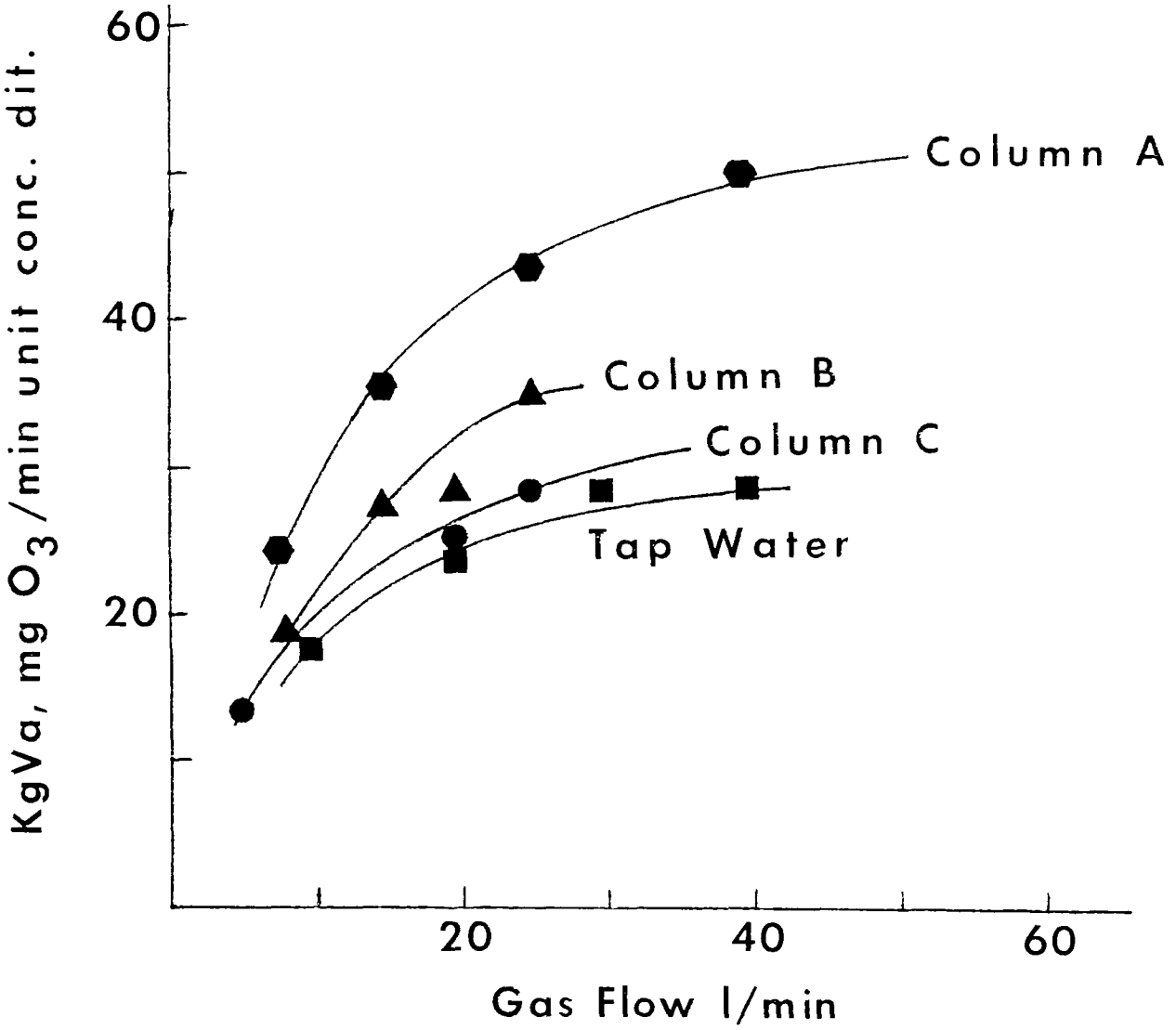

Figure 9. Effect of Liquid Flow on Mass Transfer Coefficients

12. INNOVATIONS IN THE ELECTROLYTIC GENERATION OF OZONE

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ABSTRACT

Though it has been known for well over a century that ozone may be generated through the electrolysis of aqueous electrolytes at inert anodes, only recently has research uncovered conditions that allow the process to be considered seriously as an alternative to conventional generation techniques. Innovations in anode material and electrolyte selection have resulted in acceptable current efficiencies at temperatures compatible with the use of energy-saving air-depolarized cathodes. Thus, in the overall process, feed air is reduced to water, which replaces that anodically decomposed (into ozone and oxygen). Advanced electrolytic ozonizers will be able to produce ozone concentrations all the way up to the limits of safety. Energy consumption is projected to be nearly equivalent to that of conventional air-fed corona discharge ozonizers, and will be independent of ozone concentration desired. The initial cost of electrolytic ozonizers may be substantially under that of conventional corona discharge equipment in that neither air pretreatment nor compression are required, D.C. power supplies are used, and that non-noble metal electrode materials can merely be stacked between injection-molded framing. Further engineering development is required before the technology can be commercialized.

1. DESCRIPTION OF THE PROCESS

Significant improvements to the electrochemical route for ozone generation have been demonstrated in recent research studies. Unlike conventional ozonators in which predried and compressed air (or oxygen) is passed through a high-frequency corona discharge, ozone is formed by electrochemical oxidation of water.

Certain aqueous fluoroanion electrolytes have been discovered, from which water may be oxidized to ozone at high current efficiency near room temperature (4-6). Advances in anode material selection also have contributed to making near ambient electrolysis temperatures possible (7). In previous work, attractive current efficiencies for ozone generation had only been observed at very low electrolyte (or anode surface) temperatures. Operation at such temperatures (-20 to -60^{o}C), in addition to requiring costly refrigeration, disallowed the use of reduction of oxygen as the corresponding cathodic process during electrolysis. The kinetics of oxygen reduction from air become very poor as temperature is decreased. Hydrogen evolution had been considered as the only available cathodic process, even though theoretically an additional 1.23V of cell potential is required. All economic projections for electrolytic production of ozone were most unfavorable. The electrolytic process

considered in this review (Figure 1) is composed of the following half-cell reactions. At the anode:

$$3H_2O \rightarrow O_3 + 6H^+ + 6e^-, V^o = +1.51V$$

and parasitically

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^-, V^o = +1.23V$$

At present there is no direct evidence for the two-electron reaction:

$$O_2 + H_2O \rightarrow O_3 + 2H^+ + 2e^-, V^o = +2.07V$$

At the cathode not:

$$2H^+ + 2e^- \rightarrow H_2, V^o = 0.0V$$

but

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O, V^o = +1.23V$$

The theoretical cell voltage for the production of ozone is 0.28V. Nothing even close to this voltage is achieved in practice, because one must suppress oxygen evolution by employing anode materials that have very high oxygen overvoltages. Similarly, the oxygen reduction reaction, which has been studied extensively in the development of fuel-cells, is notoriously slow. Cell voltages on the order of 1.8-2.1V are anticipated.

Overall, the process becomes: O_2(air) $\rightarrow$ O_3, and immediately certain inherent advantages may be pointed out. The air feed to the reactor need not be pretreated in any way. It need not be dried; in fact, slight humidification may be desirable to suppress water loss from the electrolyte. Compression also is unnecessary. Atmospheric CO_2 is rejected by the acidic electrolytes selected. On the anodic side, no NO_x is produced, only a mixture of ozone, oxygen and air serving as a carrier gas. Carrier gas (air fed to the electrolysis cells in excess of the stoichiometric requirements of the cathodes) is used to dilute the ozone formed as it evolves from the cells. Otherwise, ozone concentrations well over the explosion limit would be formed.

Here again an inherent advantage of electrolytic technology can be seen: the generated concentration of ozone is decoupled from power consumption, unlike in corona discharge technology. Ozone concentrations are determined first by current efficiency (which recent experiments (4,5) have shown may be obtained in the range of 30-50 percent) and second by the flow rate of diluent gas. Ozone concentrations of even 10 percent will be available using electrolytic technology. Air-fed corona discharge ozonators normally produce 2 percent ozone at best, many times at an energy efficiency lower than that found at concentrations approximating 1 percent.

2. HISTORICAL DEVELOPMENT AND RECENT RESULTS

Since ozone itself was first discovered by electrolysis of sulfuric acid in 1840 (18), approximately 25 publications have appeared dealing with its electrolytic generation. The field has developed slowly because until recently, the results have been uniformly discouraging and of academic interest only.

Work on electrolytic ozone generation may be characterized by electrolyte composition and by choice of anode material. The electrolyte must engage in no reactions other than oxygen and ozone evolution at the anode, and hydrogen evolution or oxygen reduction at the cathode. Chemical reactions with the ozone produced also must not occur. Such constraints led to the selection of acids of oxyanions and fluoroanions, as well as their alkali metal salts, as the most suitable electrolytes.

Very few anode materials are inert to ozone evolution conditions. Extremely high interfacial acid concentrations are produced during the anodic decomposition of water. High anodic potentials led to dissolution or passivation in the case of most metals. Platinum has been used commonly, and proves to be sufficiently inert. Certain of its noble metal alloys have been used, although their oxygen overvoltages are reduced. Conductive oxides in their highest oxidation states have been used (e.g., the alpha and beta forms of PbO_2 and SnO_2) and show promise. Pyrolytic carbons also prove to be inert in certain electrolyte compositions.

The platinum/sulfuric acid anode and electrolyte composition has been the subject of intense effort in two electrolysis regimes. Early authors used narrow filaments of platinum to achieve current densities on the order of 50-100 A/cm^2 (3,15). Current efficiencies (the fraction of ozone anodically evolved vs. oxygen) of up to 27 percent were reported from 0^oC electrolyte; however cell voltages of nearly 15V were observed. A glow discharge mechanism seems likely due to the high electric field encountered and the gas-blanketing that must occur.

The second ozone generation regime explored in the platinum/sulfuric acid combination was the electrolysis of eutectic electrolyte compositions at the lowest temperatures possible (2,19). Current efficiencies of up to 32 percent were reported; however, refrigeration costs (calculated as 1/3 to 1/2 of the energy consumed during the electrolysis itself) eliminated commercial consideration of the technology.

The platinum anode/perchloric acid combination was studied extensively in this same regime; however, maximum current efficiencies of 36 percent at -40^oC still were inadequate for scale-up (1,13,17).

A major advance in electrolytic ozone generation came with the use of PbO_2 anodes by three different groups of workers. Semchenko et al. first electrolyzed phosphoric acid and found that yields of 13 percent current efficiency can be obtained at temperatures of $10\text{-}15^oC$ (20).

Semchenko and co-workers next studied the use of perchloric acid, finding yields of 32 percent current efficiency at temperatures of -15°C (21,22). In conjunction with the use of PbO_2 anodes, a small quantity of fluoride ion was added to the electrolyte with the apparent effect of raising anode potential (and therefore ozone current efficiency). As of 1975 these were the most encouraging results yet obtained. However, with PbO_2 anodes, some erosion is observed during ozone evolution, following a combined chemical/electrochemical mechanism advanced by Foller and Tobias (8).

Fritz et al. (10) continued the characterization of phosphoric acid-based electrolyte systems, notably a neutrally buffered system in which PbO_2 erosion is suppressed. Yields of 13 percent current efficiency were obtained at ambient temperatures.

Foller and Tobias (5) studied the use of fluoroanion electrolytes, and continued to find yields using PbO_2 anodes much greater than those obtained with platinum electrodes. Further, it was found that the electrolytes HBF_4 and HPF_6 were particularly well suited to ozone evolution.

Figure 2 illustrates the current efficiencies obtained during the electrolysis of various concentrations of HPF_6 with beta-PbO_2 anodes at 0°C. Although the circumstances of this electrolysis (rapid weight loss and high PF_5 vapor pressure) are not compatible with commercial development, these experiments illustrate that high current efficiencies for ozone generation may indeed be obtained. The research and development problem is to find alternative conditions in which to run the oxidation of water so effectively.

The platinum anode was found to give very high ozone yields in HPF_6 as well, which led Foller and Tobias to propose a rationale for electrolyte selection based on anion electronegativity. Electrolyte anion adsorption on anode materials also was found to correlate with ozone current efficiency (9,16).

Foller et al. (7) found that a certain form of carbon, known as glassy carbon, also was capable of producing relatively high ozone current efficiencies at temperatures above 0°C in fluoroanion electrolytes. Under conditions ordinarily corresponding to ozone evolution, pressed carbon blacks (high-surface-area carbons) rapidly degrade, exhibiting CO_2 evolution and structural disintegration. Graphite also undergoes disintegration due to anion intercalation between its planes and consequent c-axis swelling.

Glassy carbon is much more resistant to oxidative processes and anion penetration due to its random, yet fully coordinated structure. This form of carbon is made by heat-treating certain resins under controlled inert atmosphere conditions. Attack is observed in oxyanion electrolytes and in low concentration acids of the fluoroanions, however not at all in high concentration electrolytes. The phenomenon is as curious as it is fortuitous, in that ozone yields reach their maximum at the highest concentrations of fluoranion acid electrolytes.

Figure 3 presents ozone current efficiencies as a function of current density for the electrolysis of various concentrations of tetrafluoboric acid electrolyte with glassy carbon anodes at 0°C. The highest yields are found at the highest acid concentration commercially available (48 wt percent). Figure 4 shows that these yields are stable over the periods of time investigated to date. No detectable weight loss is observed over 24 hours of accumulated running time in acid concentrations higher than 5 M.

There is a certain amount of confusion over ex-situ versus in-situ electrolytic ozone generation methods (when considering water treatment applications). What has been discussed to this point centers purely on gaseous ozone generation irrespective of contacting and end-use. Methods have been advanced, however, that propose in-situ ozone generation as an explanation of the efficacy of noble metal electrolysis as a treatment of potable water streams containing the chloride ion at levels on the order of hundreds of parts-per-million (23). Extraordinarily high voltages must be applied to pass minimal currents (due to poor solution conductivity). Actual anode potentials (independent of solution I-R) sufficient to oxidize chloride ion to chlorine (1.34V) and hypochlorite are achieved. These then, in conjunction with the adsorption and oxidation of organic substances on the electrodes themselves account for the levels of water sterilization observed.

From studies of ex-situ electrolytic ozone generation, it is clear that levels of ozone production in dilute electrolytes are quite small, and indeed may be attributable to analytical difficulties in separating the effects of the other chlorooxidants, which most certainly are produced. In any event, electrolysis at such high voltages (no matter what the assumed reaction products or current efficiencies) cannot be economic in comparison to ex-situ optimized ozone (or chlorine) generation processes.

3. PROJECTED COSTS

A accurate detailed cost estimate of electrolytic ozone generation technology is not yet possible. Projections, however, can be made, assuming that certain development milestones will be reached. Projections such as the following demonstrate why interest in electrolytic technology remains high.

3.1 Operating Cost

The operating cost of an electrolytic ozonator is almost entirely determined by the power consumption of the electrolysis cells. This power consumption may be derived from the current efficiency and cell voltage. Figure 5 is a plot of the amount of ozone produced per direct current (dc) kilowatt-hour as a function of various current efficiency levels and cell voltages. Two regions of operation are indicated, which correspond to projected cell voltages for either oxygen reduction or hydrogen evolution as the cathodic process. The ranges of cell voltage chosen as representative of the two process configurations correspond to operaton at 0.35-0.40 A/cm^2 (near the maximum of ozone

current efficiency, but at the same time avoiding the higher levels of polarization at higher current density). An anode potential of 2.2-2.4V vs. a standard hydrogen electrode (SHE), and an air-cathode potential of 0.55-0.65V vs. SHE were chosen for the purposes of this comparison. Electrolyte conductivity and a projected interelectrode gap of 5 mm also were included in the calculations.

Several current efficiency levels are indicated in Figure 5, which then may be used to determine power consumption. Horizontal lines on the figure indicate the power consumptions of conventional corona discharge ozonators. A fairly broad range is defined when the power consumption of all auxiliaries such as air drying and compression are added in, considering the entire spectrum of capacities commercially offered.

The energy efficiency of ozone production at a cell voltage of 2.0V (anode: 2.4V, cathode: 0.6V, heat disippation (I-R loss): 0.2V), and a current efficiency of 50 percent exceeds that of the best air-fed corona discharge ozonators. Similarly, a current efficiency of only 17 percent at 2.0V is required to undercut the energy consumption of some of the smaller air-fed units on the market today.

Projection of just where advanced electrolytic ozonators will lie within this range of energy consumption when fully optimized is problematical. The 2.4V anode potential and 50 percent current efficiency necessary to develop a 75g/kWh ozone electrolyzer have been demonstrated with platinum anodes at temperatures compatible with the flow of cooling water. It is possible to achieve these performance levels under laboratory conditions. Stable current efficiencies of 35-40 percent also have been achieved with the much less expensive glassy carbon anodes in a less volatile electrolyte (HBF_4), however, at somewhat higher anode potentials.

The optimization of energy efficiency in a commercially practical cell design will include the selection of a current density (the trade-off is that increasing current density increases current efficiency, but at the same time increases electrode potentials, I-R losses and heat generation), selection of an operating temperature (the trade-off is that increasing electrolyte temperature decreases air-cathode polarization, and increases electrolyte conductivity, but at the same time diminishes ozone current efficiency), and selection of anode, air-cathode, and electrolyte compositions. It is very likely that energy consumptions on the order of 45 to 50 g/AC kWh (95+ percent power supply efficiencies are common) can be achieved with non-noble metal electrodes and cooling water compatible anode temperatures.

Operating costs also will include maintenance. Both anodes and cathodes probably will need replacement at certain intervals. Even platinum-clad anodes probably will be subject to slow erosion. Glassy carbon anodes so-far have appeared extremely stable during 12- to 24-hour testing. The air-cathodes should exhibit lifetimes in excess of the 40,000 hours projected for high-temperature (190^{o}C) municipal power generation fuel cells. In these fuel

cells, catalyst area loss through agglomeration is a prime failure mode. At ambient temperature, longer lifetimes are expected, as migration is reduced. Periodic electrolyte rebalance through water or acid addition may also prove necessary.

3.2 Capital Costs

Electrolytic ozone generation should have initial cost advantages over conventional air-fed corona discharge technology for three reasons. First, the cell stack can be assembled from injection-molded polypropylene framework, and non-noble metal electrodes. The power supply required is very unsophisticated, a conventional dc source with minimal regulation. A 90-V, 3,500-A unit for a 1,000-lb/day ozonator can be purchased for $19,000 (1981). High-frequency and high-voltage power supplies for corona discharge ozonators are much more expensive. Finally, contacting costs can be reduced as higher concentration ozonizers can reduce contactor sizes and increase throughputs. However, mass transfer studies at the higher ozone concentrations available by electrolysis must be conducted first, to prove this hypothesis.

The higher concentrations of ozone in air available by electrolysis imply that a given quantity of ozone can be applied using a much lower volume of air. This will provide savings, because of smaller gas-handling equipment.

The size and capital cost of electrolytic ozonators may readily be estimated once some basic assumptions as to the progress of subsequent research are made. Assuming that 40 percent current efficiency can be achieved at cooling water temperature, and that a cell voltage of 2.0V will be encountered at 350 mA/cm^2, a 1,000-lb/day electrolytic ozonator may be sized.

A total current of 158,000 A is required. Therefore, if a 90-V power supply is used, two parallel stacks of forty-five 1,750-A cells may be envisioned. Each bi-cell would have an electrode area of 5,000 cm^2 (50 x 100 cm) and a thickness of approximately 3-4 cm, counting air and coolant flow provisions. Thus cell stack dimensions of 1.5 x 1.5 x 2.0 m appear likely.

Costs may be calculated on the basis of anode material ($50/$ft^2$), cathode material ($20/ft^2) and cell framing. A filter-press design seems most likely. Electrolyte, reservoirs, and auxiliaries such as monitoring equipment, air blowers and filters also must be added in along with assembly, overhead costs, and 40 percent mark-up. Figure 6 compares the projected cost of electrolytic ozonators with the costs of conventional air-fed ozonators as determined in a 1979 study of the U.S. Municipal Environmental Research Laboratory (11). A dramatic reduction in initial cost is forecast due to the basic simplicity of electrolytic technology. Whether this will be, in the end, the 75 percent reduction exhaustively calculated in the preparation of Figure 6, or only a 50 percent reducton, it is clear that significant advantages in cost are promised.

The reduction of the capital cost of ozonators is extremely important in that capital cost represents a very significant fraction of the total cost of ozonation. Amortization of equipment costs can outweigh operating cost (power consumption) for large installations. Figure 7, derived from the EPA-sponsored study of Gutmann and Clark (12), shows that even at 7 percent interest rates and 20-year amortizations, the fraction of capital-related costs in the total cost of ozonation (contacting costs included) rises rapidly. (An up-to-date detailed analysis of ozone cost alone follows.)

Contactor costs may be reduced because mass transfer rates from the gas phase to solution phase are inversely proportional to one minus mole fraction of ozone in the gas phase (14). Therefore, at the higher ozone concentrations that are produced by electrolytic technology (at no energy penalty), contactor sizes might be reduced, and/or a greater volume of solution may be treated per unit time. Such potential advantages of electrolytic technology must be analyzed in greater detail with regard to specific applications of ozone.

3.3 Total Amortized Cost of Ozone Produced

In order to more fully assess the economic impact of the development of an advanced electrolytic ozonizer, the following analysis of the cost of ozone on a per pound basis was performed.

Ozonizers of 1,000 lb/day capacity were used as a basis. These were assumed to have 20 year lives, and to be operated 24 hours per day 300 days a year. Replacement of anodes and cathodes of the electrolytic cells was scheduled for every five years. Twenty year financing at 15 percent interest, and maintenance cost of 5 percent the initial cost per year were assumed in each case. An identical power consumption of 60g/kWh (7.5 kWh/lb) at a $0.04/kWh electricity cost was further assumed. An initial cost of $800K was taken for an air-fed corona discharge ozonizer. An initial cost of $195K for the electrolytic ozonizer reflects a materials cost inclusive of applicable freight and taxes with 25 percent contingency, labor overhead at 200 percent direct, 15 percent general and administrative costs, and a 40 percent profit.

The annualized costs are then computed as follows.

Cost Element	Conventional Generator	Electrochemical Generator
Capital Costs	($800,000)	($195,000)
Interest on debt	$120,000	$ 29,300
Sinking fund for debt retirement	7,800	1,900
Operating Costs		
Maintenance	40,000	12,800
Electricity	90,900	90,900
Total Annualized Costs:	$258,700	$134,000
Cost Per Pound of Ozone:	86.2¢/lb	44.7¢/lb

Thus, a substantial difference in the cost of ozone is found. This difference is a key reason why the development of high-concentration electrolytic ozonizers is attractive.

4. DEVELOPMENT REQUIRED

To date, the electrolytic process has only been tested in small cells of 1-5 cm^2 electrode area. Further, testing has been performed only in half-cell configurations (i.e., hydrogen-evolving ozone cells and oxygen-evolving oxygen reduction cells). Only laboratory cell designs have been tested, but commercially practical configurations are on the drawing board.

Scale-up development involves coopting the desirable features of modern fuel-cell design (fuel-cells employ oxygen reduction) and advanced water electrolyzer design (water electrolyzers produce H_2 and O_2). Both technologies are now highly developed.

Assuming the glassy carbon anode and tetrafluoboric acid electrolyte system will be selected for final scale-up, the following development steps must be undertaken.

First, an optimized air-cathode formulation must be developed for HBF_4 electrolyte and high current density operation. Air-cathode technology is quite advanced, and such formulations have been developed for acid electrolyte fuel-cells, as well as alkali chlorine/caustic cells and metal/air batteries. The air-cathodes will consist of a Teflon-bonded Pt-catalyzed high surface area carbon. A catalyst loading of 1.0-1.5 mg Pt/cm^2, or less, on a carbon

such as Vulcan XC-72 bonded with approximately 20 wt percent Teflon-30 seems likely. Techniques of cathode manufacture, however, are complex, and in many instances proprietary.

The glassy carbon anodes also will have to be optimized for ozone evolution. Sensitivity to production methods, such as heat treatment temperature and starting resin, has been noticed in ozone current efficiency data (7), and to some extent accounts for differences in yield seen between Figures 3 and 4.

Most importantly, integrated cell testing to co-optimize operational temperature and current efficiency for minimal power consumption must be performed in practical cell designs. At this point, long-term testing of the cells would be begun.

Mass transfer studies should be conducted, using ozone-air combinations that contain higher concentrations of ozone, so that optimally sized ozone contacting chambers can be designed.

In addition, higher concentrations of ozone in air should be tested for compatibility with materials of construction. Higher concentrations of ozone probably will result in shorter lifetime of certain components of ozone-handling equipment.

5. POSSIBLE APPLICATIONS

Electrolytic technology probably will find applications in certain special-purpose fields well-suited to its particular characteristics in advance of its full optimization. These may be applications in which high concentrations of ozone are required (any concentration up to the limits of safety would be available), or in which relatively small quantities of ozone are needed (say 0.2-1.0 lb/day) at low initial cost. If 50 percent current efficiency at a 2.0-V cell voltage can be achieved in a commercial design at cooling water temperature, electrolytic technology will, of course, find the widest possible application.

Applications requiring very high concentrations of ozone are limited. Many current large-scale applications should benefit from increased concentration during contacting, but would, at the same time, require a fully optimized power consumption.

Hazardous waste treatment is a likely application, in that power cost is not a central issue in disposal of certain highly toxic materials. The high concentrations of ozone (previously unavailable) that electrolytic technology can provide most certainly will improve the oxidation kinetics of organics. An advantage of ozone in this field is that is is nonspecific; it can decompose many unsaturated aliphatics and aromatic organics even when chlorinated. Known pesticide, PCB, phenol, cyanide, surfactant, nitrocompound, dye waste,

higher alcohol, and organophosphate decomposition process should be more rapid at higher ozone concentrations.

Applications requiring very low initial cost may also be amenable to unoptimized electrolytic technology. A low-maintenance, continuous-treatment process for swimming pools may be devised, for example. The electrode area required to treat a 20,000-gallon pool at 1-mg/l-day works out to less than 500 cm^2, and the power requirements lie in the range of 2-3 kWh/day (well below filtration pumping costs).

6. CONCLUSIONS

Recent developments indicate that the electrochemical synthesis of ozone may become an economically feasible alternative to corona discharge. Additional basic research is required, along with substantial engineering development. However, the needed development centers on the available technologies of fuel cell and water electrolyzer (products: H_2 and O_2) design.

The possible outcome of continued efforts in electrolytic ozonator development is that high-concentration ozonators of quite low cost may become available with power consumptions equal to those of the best air-fed corona discharge technology. This new breed of ozone generators also may enable contacting costs to be reduced. Further, the technology will scale-up and scale-down with equal ease. Research in this field undoubtedly will continue.

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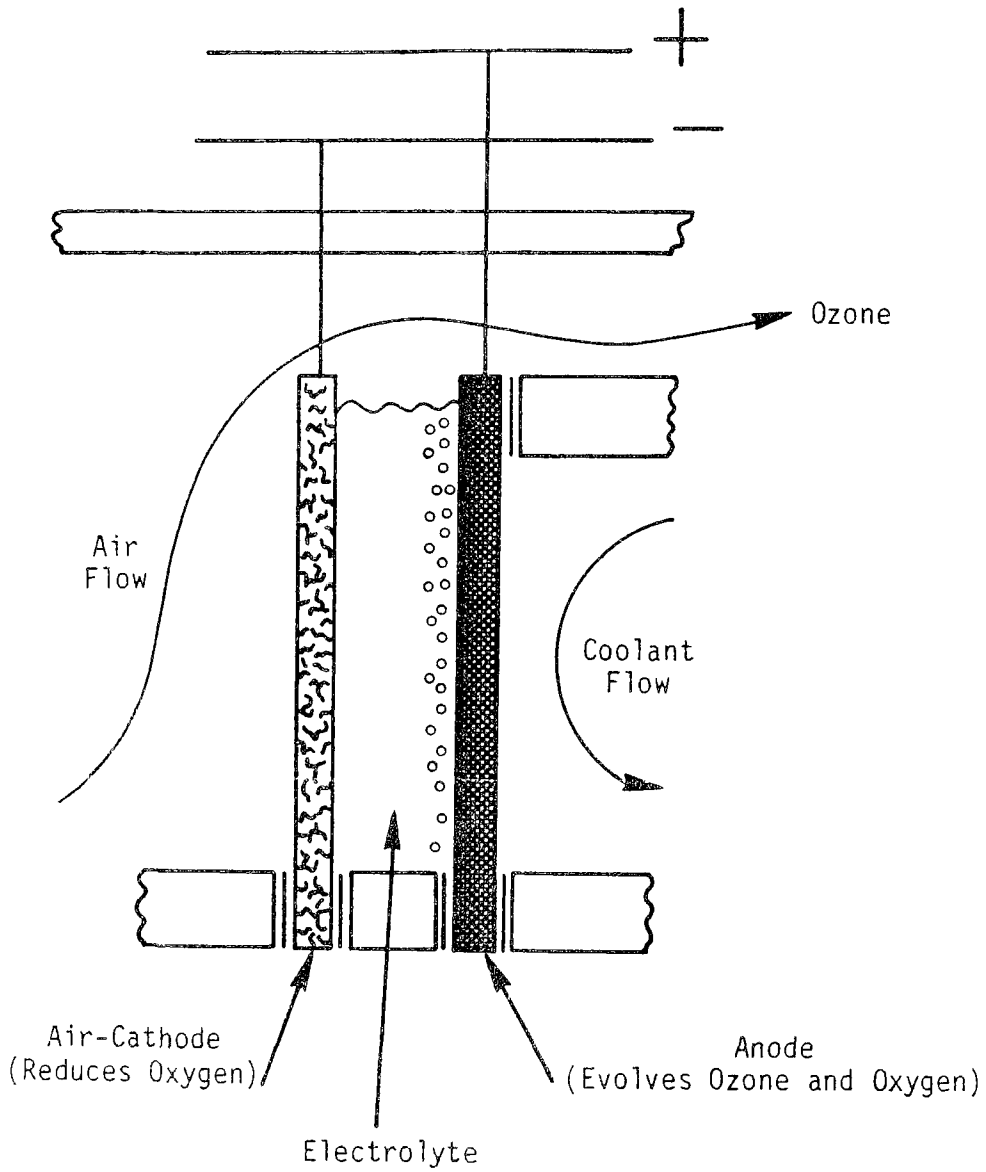

Figure 1. Schematic of ozonator cell design

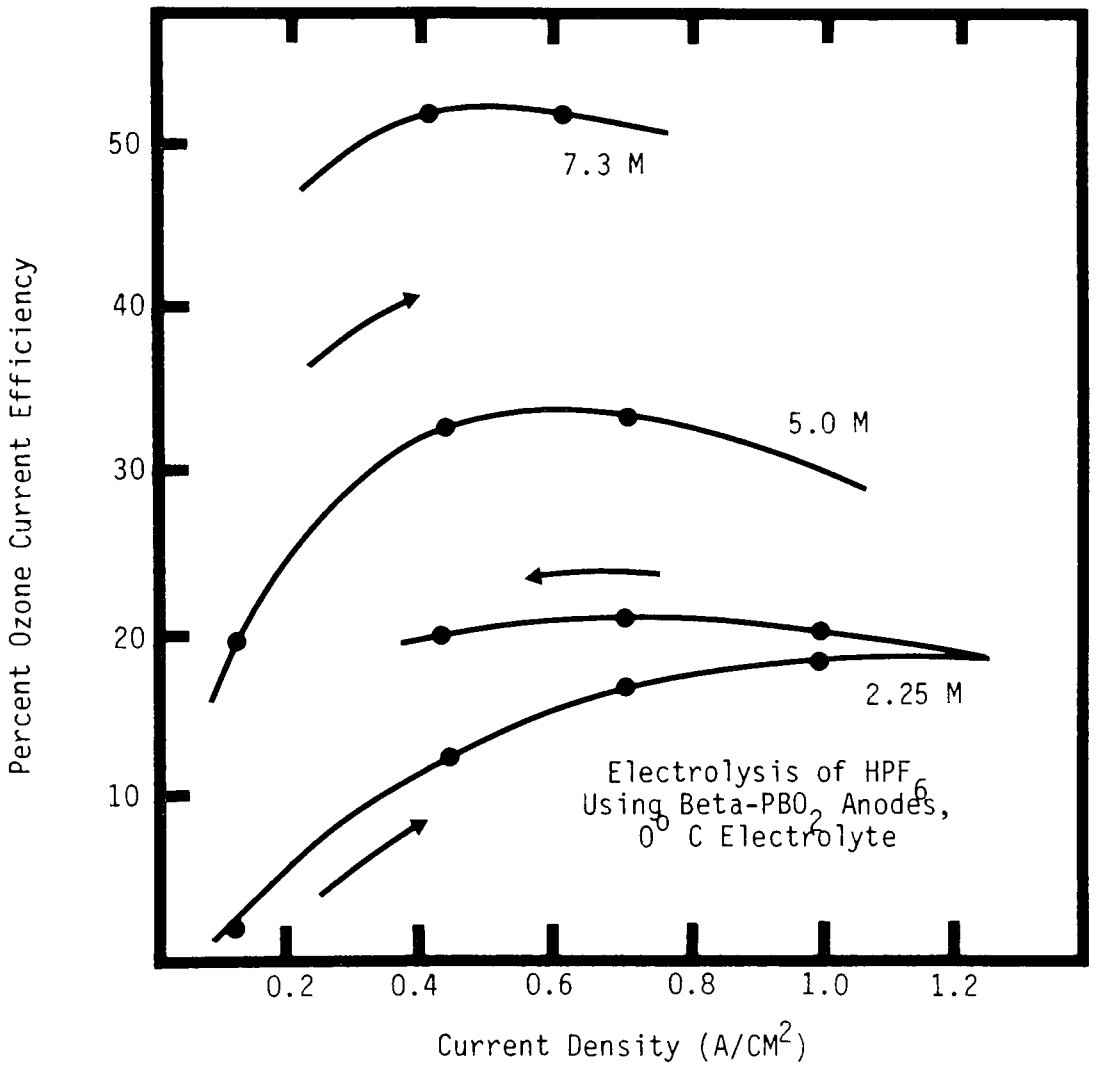

Figure 2. Current efficiency of the beta-PbO_2/HPF_6 anode/electrolyte combination as a function of current density and concentration at $0^{o}C$.

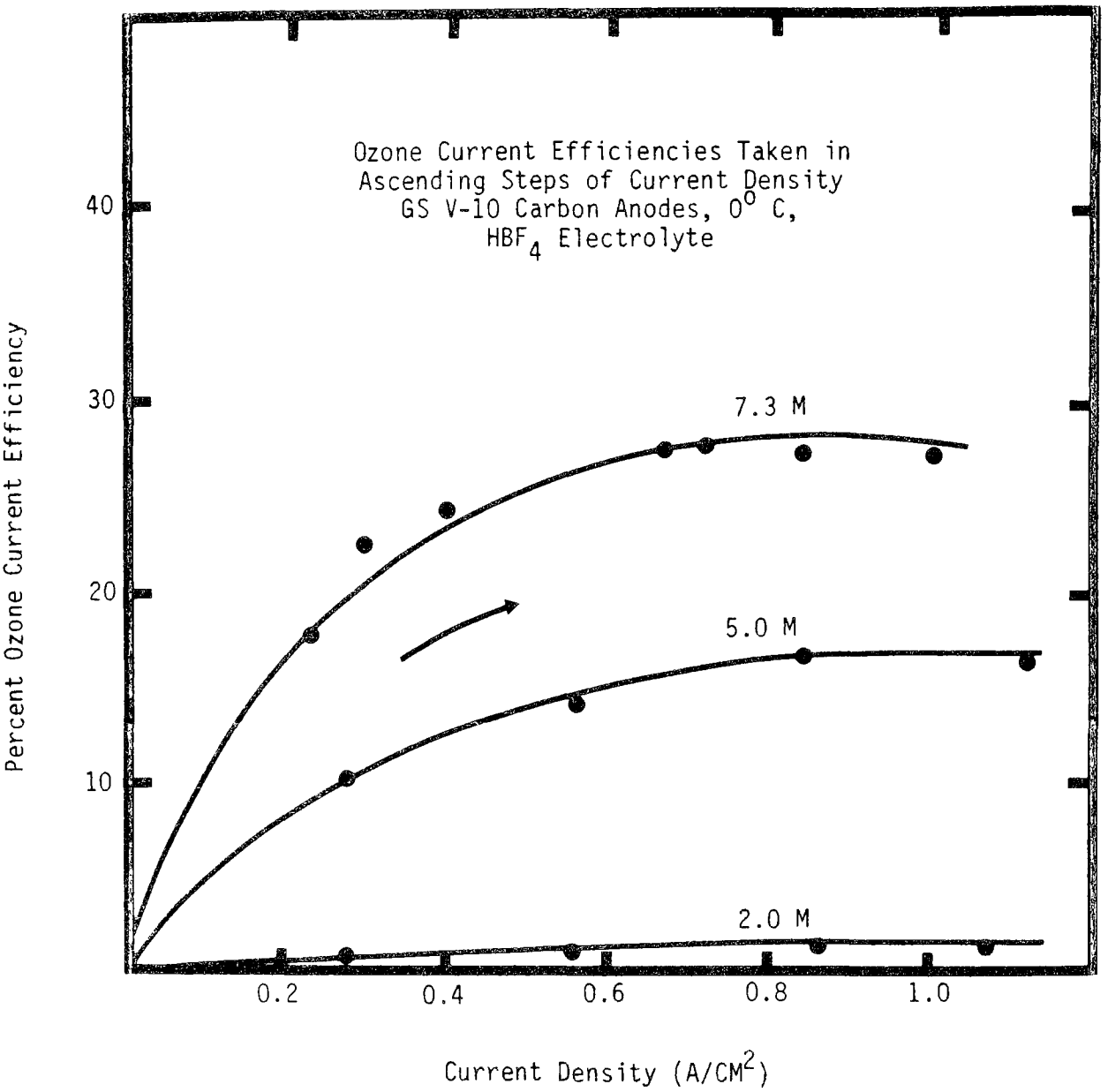

Figure 3. Current efficiency of the glassy carbon/HBF_4 anode/electrolyte combination as a function of current density and concentration at $0^{o}C$.

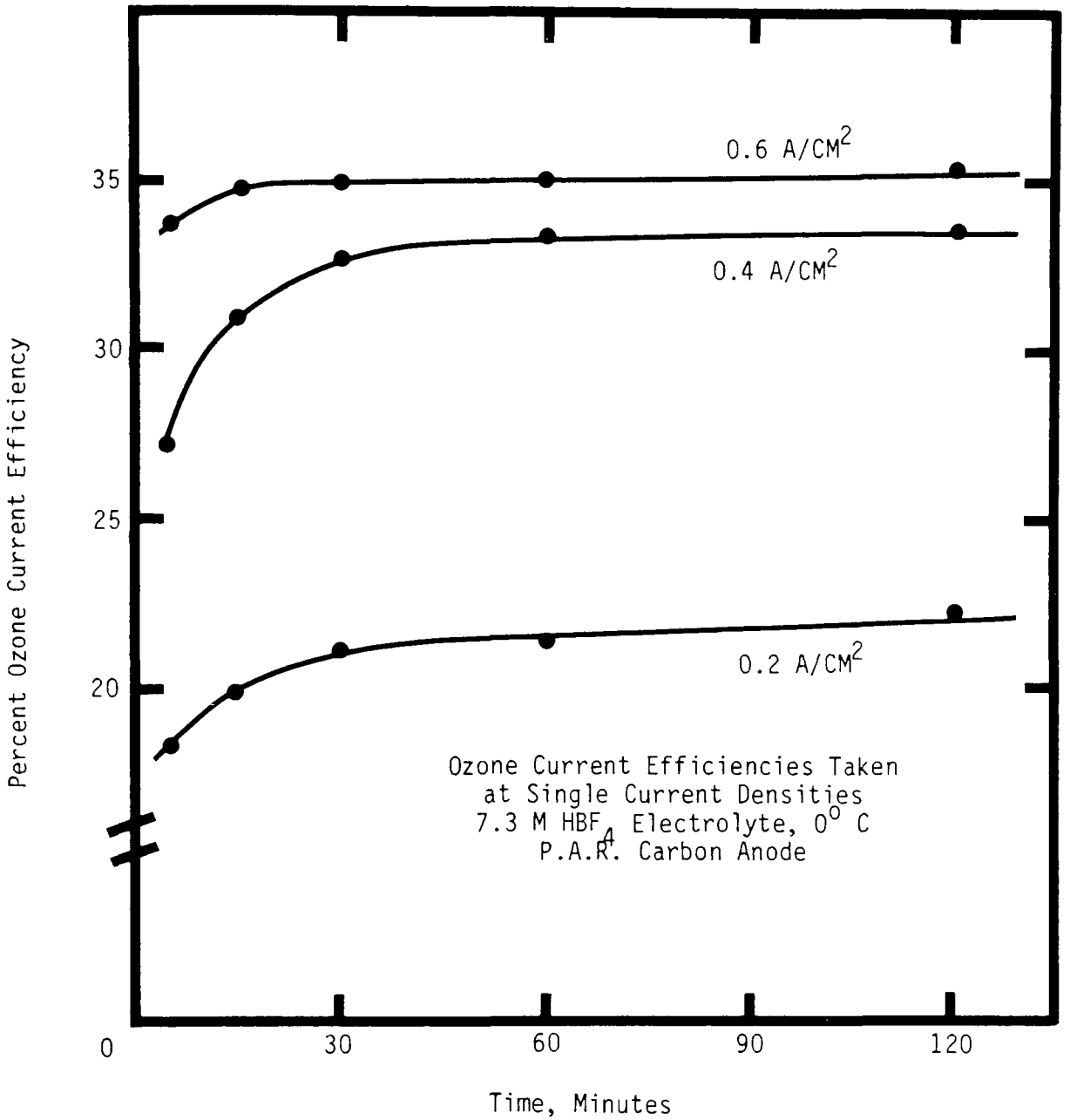

Figure 4. Current efficiencies of the glassy carbon/HBF_4 anode/electrolyte combination as a function of time at $0^{o}C$.

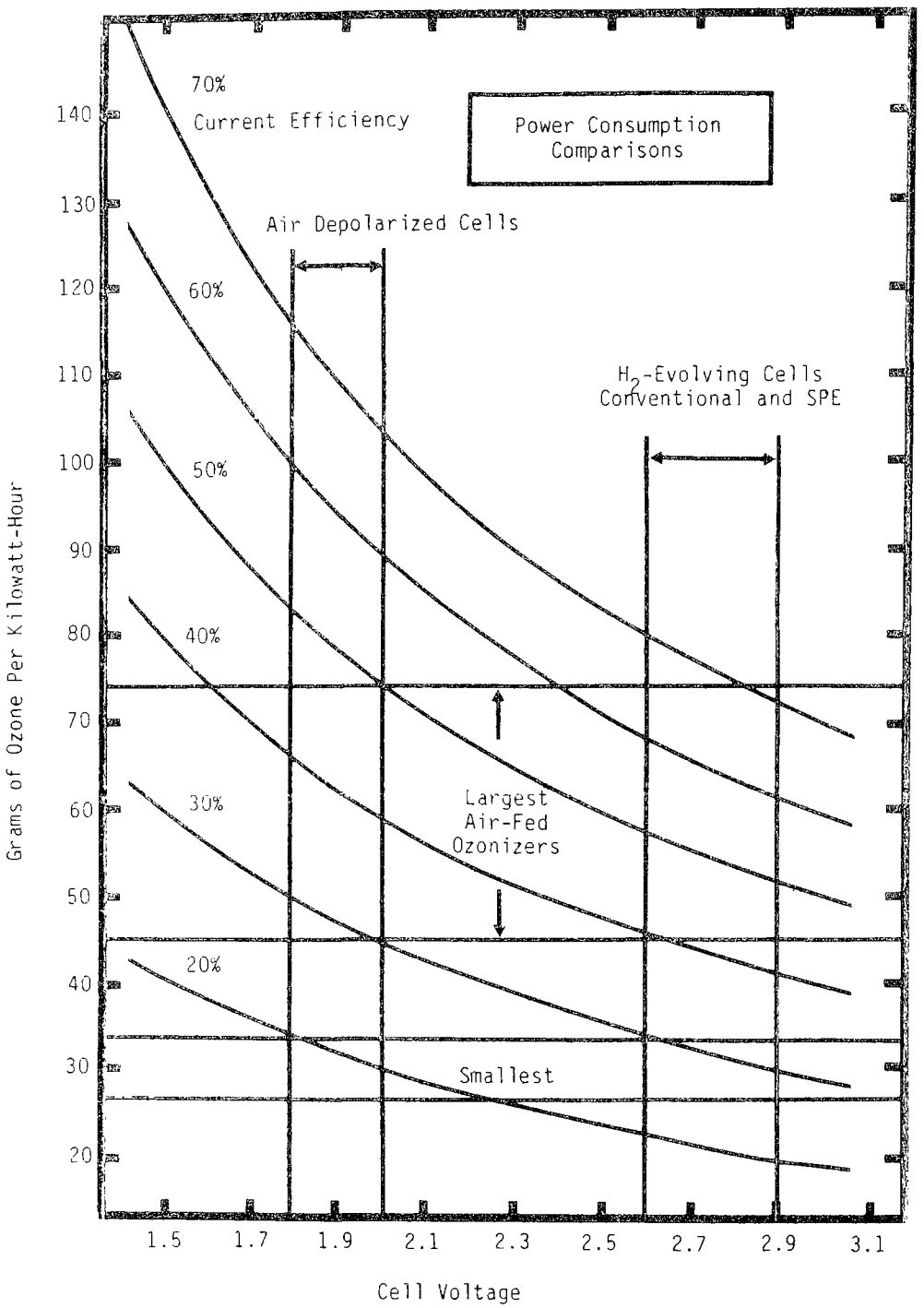

Figure 5. Analysis of the power consumption of electrolytic ozonators.

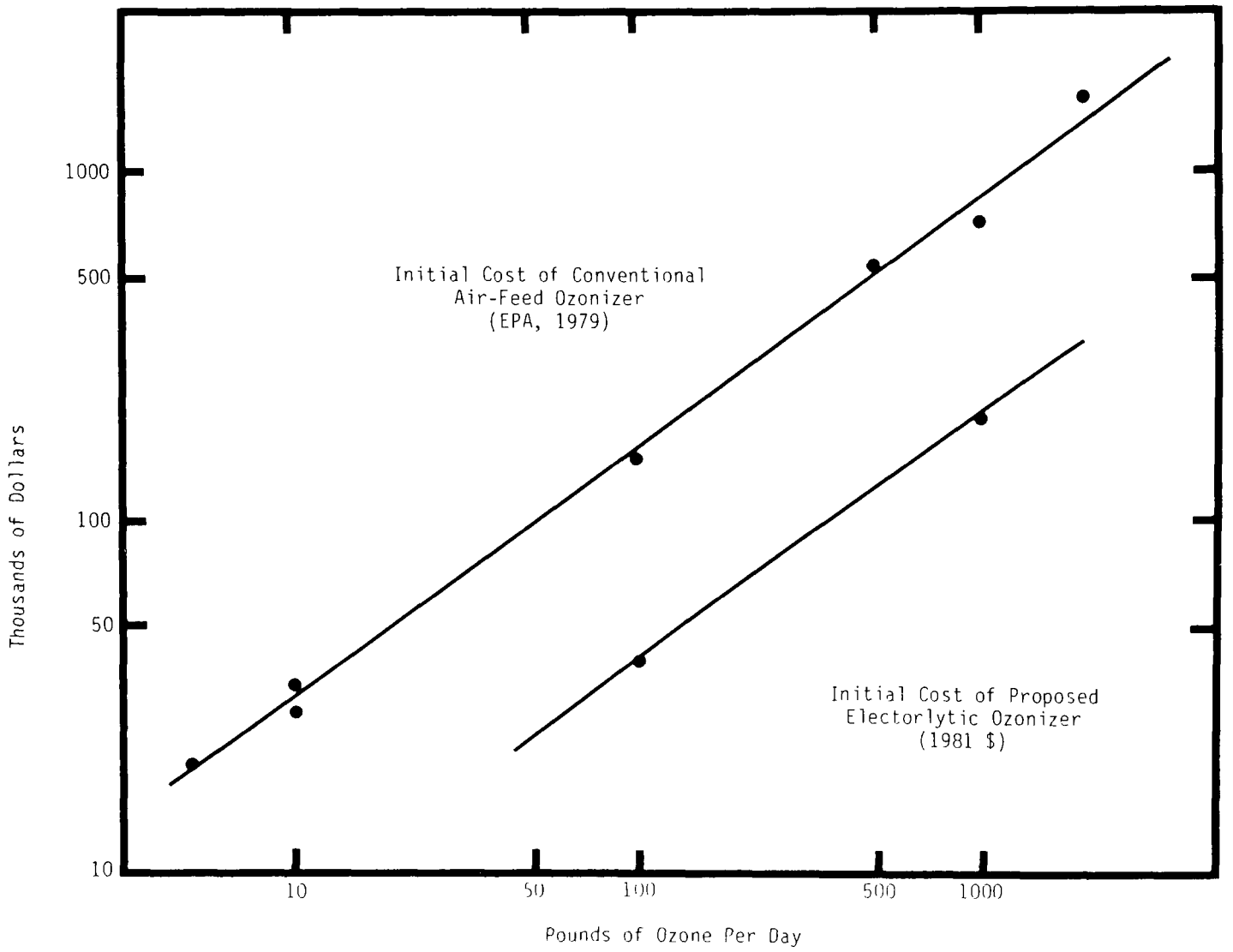

Figure 6. Comparison of capital costs (21).

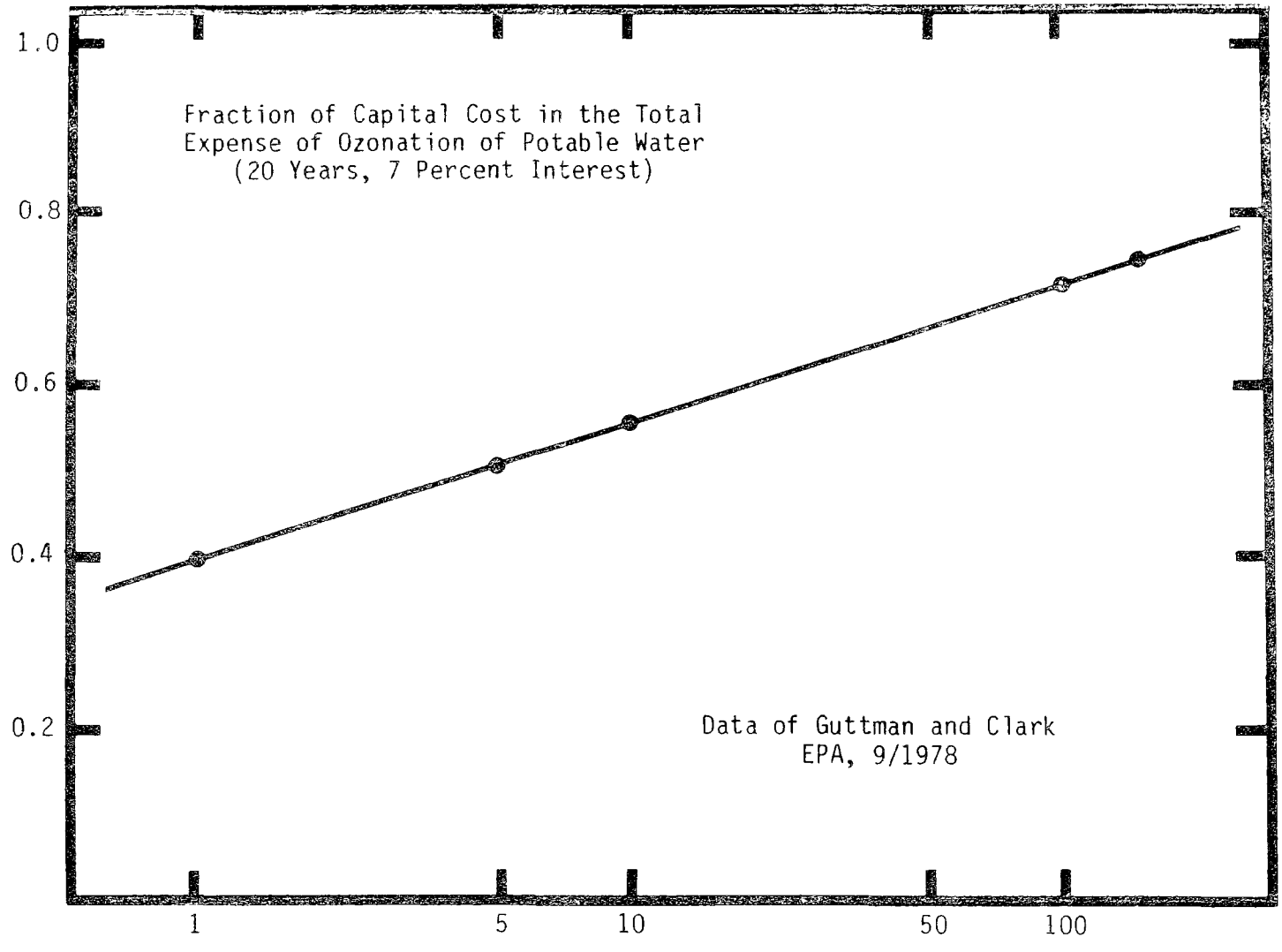

Figure 7. Fraction of capital cost in total expense of ozonation (22).

1. PRACTICAL CONSIDERATIONS IN THE USE OF HALOGEN DISINFECTANTS

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ABSTRACT

The various issues to be faced when designing and operating wastewater disinfection systems utilizing chlorine, hypochlorites, chlorine dioxide and bromine chloride will be reviewed, and areas of continuing uncertainty will be highlighted. These include dose estimation, contactor hydraulics, chlorine process control systems, and mixing conditions at the point of application.

INTRODUCTION

Halogens have been employed as disinfectants of wastewater for at least 150 years, since Averill (2) reported "When it is desirable to destroy the effluvia from drains, sewers, etc., or to purify the water of a cistern--dissolve about eight ounces of the chloride of lime in a pail full of water, and disperse it into them. Repeat the operation until the object is effected." Nevertheless, major issues relating to the design and operation of halogen disinfection processes remain only partially understood. This paper will review several of these as a preliminary step in the preparation of portions of a design manual on the subject.

ISSUES IMPORTANT AT THE DESIGN STAGE

When a wastewater disinfection system is to be designed, numerous problems present themselves, from those of dose estimation, to hydraulics, chemical supply and safety. Rather than enumerating all possible issues, several points of continuing uncertainty will be reviewed.

Chemical Dose Estimation

One of the basic questions in disinfection is how much of a given chemical is needed to attain a desired effluent standard. In wastewater, in particular, this question is complicated by the different upstream processes and resulting inputs to a disinfection system, and by the existence of substantial, competitive, demand-exerting reactions for all of the halogens employed.

While a number of references (18,35-38) present broad guidelines on the dose requirements needed to disinfect wastewater using chlorine, information on the analysis of microbial inactivation kinetics by halogens in wastewater remains sparse. Only two authors have considered the estimation of such process rates.

One model, of Selleck (24), which has been cited by other sources (18,35, 38), is of the form:

$$N/N_0 = (1 + ct/b)^{-a} \qquad (1)$$

In equation 1, c is the chlorine residual (generally, total residual) after a contact time t, while a and b are empirical constants. In this model of batch reactor wastewater chlorination kinetics, the empirical coefficients have been shown to vary in a poorly understood manner with the degree of prior treatment, and with the chlorine:ammonia-nitrogen dose ratio (24). The empirical Selleck model has been verified by Roberts et al. (21) for modelling the inactivation of coliforms in wastewater effluents by varying degrees of treatment using both chlorine and chlorine dioxide.

There appear to be at least three major problems with the above dose estimation procedure. The first, expressed by Roberts et al. (21), is that "...the model...has no rational, mechanistic basis in describing disinfection by chemical agents. Nonetheless, it does approximate empirically the behavior of the real system and as such provides a useful design tool." The lack of theoretical justification for this model makes it difficult to incorporate knowledge about contactor imperfections and mixing dynamics into the calculation procedure.

As a corollary to the above, a second problem with the approach of Selleck is the inability to extrapolate readily from kinetic parameters obtained on one effluent to those of another effluent. For example, in the studies by Roberts et al. (21), values for the a and b parameters were observed to differ between the treatment plants examined in the case of chlorine and chlorine dioxide, and the chlorine values differed from those reported by Selleck (24).

A third major problem with the Selleck approach is the need to estimate the chlorine demand and thus to calculate the initial chlorine dose required. Roberts et al. (21) have employed the empirical equation initially developed by Taras (33) to calculate the dose required for chlorine or chlorine dioxide disinfection in conjunction with the Selleck model. However, the Taras approach to chlorine demand calculations appears to share some of the disadvantage of the Selleck model in that it cannot readily be extrapolated to a different wastewater.

A second approach to the problem of determining disinfectant doses is the use of mass balance and reaction rate expressions for disinfection per se, chlorine-demand reactions, and other simultaneous processes which might occur (i.e., mixing of two fluid streams). The author (9) has described this process elsewhere, for the particular case of wastewater chlorination, and has contrasted such mass balance models with experimental data. The major drawback of this approach is the fact that the resulting mass balance models consist of several simultaneous ordinary differential equations, which may be non-linear, and thus are amenable only to numerical solution. In addition, the rate constants for many of the chlorine-ammonia and chlorine-amine reactions are not well characterized. The major advantage of this procedure, in principle, is that the inherent sensitivities of microorganisms to various disinfectant species and reaction rate constants with ammonia and amines might be expected to remain relatively constant among various wastewaters.

In the case of chlorine dioxide, the estimation of dose requirements for wastewater disinfection remains clouded by lack of knowledge regarding the chemical species responsible for chlorine dioxide demand. While White (38) indicates that chlorine dioxide demand of wastewater should be greater than that of chlorine demand, work of Roberts et al. (21) indicates that although this is true in conventional secondary effluent, the chlorine demand in nitrified filtered effluents may exceed the chlorine dioxide demand. The chemical reactions leading to chlorine dioxide demand are not known.

Influence of Disinfection Pretreatment

A related issue to that of dose estimation is the effect of treatment prior to disinfection. While efficient operation of secondary and tertiary treatment can directly remove microorganisms from wastewater, and thus reduce the necessary stringency of disinfection, several indirect effects upon this latter process have also been uncovered.

It is well known that the degree of nitrification, if any, and the presence of ammonia, organic amines, and various reducing agents can affect the efficiency of the chlorination process (37,38). Furthermore, the increase in efficiency of disinfection by chlorine with reductions in pH has also been reported (37,38).

More recent studies have indicated that the presence of certain cations may affect chlorination efficiency, although the significance of these effects in the field is unknown. For example, Kuzminski (16), Reid and Carlson (20), both working in laboratory demand-free systems, indicated that calcium concentrations could interfere with the chlorine inactivation of coliforms. In other work, a number of studies (12,14,22,27,31) have indicated that sodium, and perhaps potassium ions, can enhance the rate of inactivation of viruses as well as coliforms in laboratory, demand-free systems, and that the formation of a previously neglected ion-pair may explain this phenomenon (10,12). If coagulants or neutralizing agents are added prior to disinfection by chlorine, these indirect effects may be of significance, and may be amenable to manipulation with the objective of chlorine dose minimization.

With respect to chlorine dioxide, virtually no information exists which permits generalization regarding the effect of the surrounding menstruum on disinfection efficiency. While a number of authors have indicated that increasing pH increases the efficiency with which chlorine dioxide inactivates microorganisms in laboratory studies (5,23), the mechanism of this effect, and its applicability to full-scale wastewater treatment plants remain unknown. A very recent paper indicates that, when applied as a potable water disinfectant, chlorine dioxide inactivation efficiency also decreases as increasing amounts of humic color material are present (7).

With respect to bromine, and presumably bromine chloride, increasing the pH of a wastewater has been found to increase the efficiency of disinfection. This effect, and its contrast to the behavior of chlorine, has been attributed to the efficacy and stability of monobromamine, predominating at high pH, as compared with dibromamine, predominant at low pH (30).

A second type of pretreatment effect relates to the influence of prior conditions upon the innate sensitivity of microorganisms to inactivation by the halogens. These effects may relate to the selection of resistant strains of microorganisms or the alteration of innate physiological conditions so as to increase resistance. While the existence of these effects has rarely been investigated at wastewater treatment plants, a variety of studies in laboratory systems or in potable water treatment plants have suggested that *in situ* strains of microorganisms may be more resistant to chlorine than commonly used laboratory strains (26), may develop altered resistance upon repeated exposure and subculture (3,4,11), or that the antecedent growth conditions may alter the sensitivity of coliform organisms to chlorine (19). In wastewater, Aieta *et al.* (1) have shown that native populations of total coliform organisms are more sensitive than pure cultures of *E. coli* exposed to chlorine and chlorine dioxide under similar conditions; whether this is due to the importance of resistance, encapsulated coliforms, or to an inherent or induced strain resistance remains uncertain.

A peculiar example of this pretreatment effect appears to be emerging with regard to the chlorination of nitrified effluents. While it is well known that nitrites in such effluents may hinder chlorination due to exertion of a chlorine demand, White (39) has reported on the San Jose, CA plant, in which disinfection by chlorine was improved by addition of small amounts of ammonia nitrogen. The mechanisms for this effect are still unresolved, but it should be noted that in the vicinity of the breakpoint, the standard procedures for the analysis of chlorine forms may be subject to serious error (29). The experience of White in regard to disinfection of nitrified effluents is also supported by unpublished observations recorded at the Metropolitan Sanitary District of Greater Chicago (T.B.S. Prakasam, personal communication).

Mixing and Contactor Hydraulics

The hydraulic conditions at the point of mixing between the solution of disinfectant and wastewater, and in the subsequent contact chamber, have been shown to have a substantial effect on process performance. However, particularly with regard to the first effect, the mechanism of this phenomenon is not well understood.

The enhancement of chlorine disinfection of microorganisms in wastewater by intense mixing at the point of chemical addition has been documented by Longley (17). Recently, it has been suggested that reductions in chlorine dosage amounting to as much as 50 per cent can be achieved, in part, by optimizing the flash mixing conditions (25), and White (38) has advocated the use of an rms velocity gradient at the point of mixing ("G") of up to 1000 sec^{-1}. However, theoretical modelling of the wastewater chlorination process indicates that the observed enhancement is not due to the acceleration of contact between microorganisms and the rapidly reacting free chlorine (9), but may be due to an as-yet poorly understood shearing of microorganisms from protective particulates.

Since the disinfection process is positive order in microorganism concen-

tration, and since high performances are normally desired in such systems (i.e., efficiencies will in excess of 90 per cent), classical theory predicts that plug flow contactors should be vastly superior to complete-mix contactors (13,28). A corollary of this principle is that any small deviations from ideal plug flow behavior in a contactor will result in drastic deterioration in observed performance of a real system. The empirical length to width ratios resulting in close to plug flow conditions have been summarized by White (37,38) and in the WPCF Manual of Practice (35,36). However, the only quantitative synthesis of the effect of hydraulic imperfections upon chlorination contact chamber performance appears to have been that of Trussell and Chao (34), who combined the theory of reaction with longitudinal dispersion, under the assumption that inactivation is governed by the Selleck equation, and that chlorine residual is constant, with the assumption of segregated flow, to conclude that improvement in hydraulics which achieve a dispersion lower than 0.01 have little practical effect. However, it should be noted that the particular assumption of segregated flow used in the Trussell and Chao analysis, as well as the neglect of residual decomposition during contact and the limitations of the Selleck relationship introduce sources of error in this conclusion.

A second hydraulic aspect which has been briefly mentioned by White (38) is the ratio of volumetric flows of the halogen feed solution to the wastewater and its influence upon inactivation efficiency. It has been suggested that decreasing this ratio, i.e., using a low volume, highly concentrated, feed solution will improve efficiency. While this has been supported by theoretical modelling of the disinfection process itself (9), no experimental data appear to have been collected to elucidate this point.

ISSUES IMPORTANT FOR OPERATIONS

Following the start-up of a wastewater disinfection system, many operational factors become important in the performance of the process. Work is now underway to enumerate these various factors. Two major issues have received much attention, namely the control of chlorine dose and/or residual, and the behavior of the contact chamber as a sedimentation tank.

Process Control

The design of halogen disinfection processes generally precedes utilizing steady state assumptions and peak or average design flow conditions. In an attempt to meet an effluent microbiological constraint while minimizing the dose (and, in some cases, under regulation, the effluent residual) of halogen during conditions where flow and influent composition vary, it is necessary to introduce a control system. The most sophisticated version of this system is compound flow and residual control (35).

This system is only as strong as its weakest link, which would appear to be the chlorine analyser itself. Since there is a great difference in microbial sensitivity between free and combined chlorine forms, and since there is also a difference in sensitivity of microorganisms to mono- and di-chloramine (8), it would seem desirable to employ an analyser which could differentiate

among these distinct species and provide a sensitivity-weighted value of chlorine residual present. No such analyser exists, with the possible exception of the membrane polarographic electrode (15) which is sensitive primarily to HOCl. Snead et al. (29) have noted that all commonly used methods for chlorine analysis suffer from false positive indications of the presence of free chlorine under certain circumstances.

While the use of amperometric and automated wet chemical analysers is widespread in wastewater treatment plants practicing automatic control, there would therefore seem to be some room for future improvements in this area.

Solids Sedimentation

To prevent solids deposition in disinfection contact chambers, various sources recommend the use of a minimum horizontal flow-through velocity to promote scour (3,18,38). However, this approach has been questioned on the grounds that the occurrence of additional sedimentation in contact basins may promote the removal of microorganisms associated with the removed solids (32). There does not appear to have been a systematic study of this issue, which also directly influences the operation of contact basins, in that if sedimentation is promoted a means for solids collection must be provided.

SUMMARY

A number of issues associated with the design and operation of halogen disinfection systems have been discussed, and various areas of continued uncertainty highlighted. These include the following:

1) The estimation of halogen dose using procedures which are of a rational nature is still not entirely possible.

2) The influence of pre-disinfection treatment on the efficiency of the disinfection process, other than by alteration of pH or concentration of ammonia, remain to be investigated. In particular, the significance of cations in aiding or hindering wastewater chlorination should be determined, and the effect of various types of biological treatment on the inherent sensitivity of surviving microorganisms should be addressed. With chlorine dioxide, the basis for the effect of pH in altering disinfection efficiency should be explored.

3) The interaction of mixing at the point of chemical introduction and the inactivation process is not well understood from a mechanistic point of view, although the existence of this phenomenon is demonstrated. Until such mechanisms are understood, it is difficult to present any generalizations regarding the optimal amounts of such mixing.

4) Further attempts to model the influence of contactor hydraulics upon process efficiency should be made, and it is essential to obtain field verification of these results, to confirm the many necessary assumptions.

5) Continuing efforts are needed to develop chlorine analysers which are capable of distinguishing among the various forms of free and combined chlorine, and to incorporate such analysers in process control schemes. While the ideal analyser would be a rapid bioassay procedure, this does not appear feasible at present.

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2. DESIGN AND OPERATIONAL CONSIDERATIONS FOR WASTEWATER OZONE DISINFECTION SYSTEMS

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ABSTRACT

Ozone systems are usually air fed; once through oxygen fed; or recycle oxygen fed units. Air fed and recycle oxygen fed systems require dew point treatment processes that are extremely sensitive, yet critical to ozone production. Once through oxygen systems are desired, if the oxygen requirement of the downstream oxygen process (e.g., oxygen activated sludge process) can be balanced with the oxygen requirement of the ozone process.

A high dew point of the feed gas will decrease ozone production and may damage generator components. Typically, more than 99.9 percent moisture removal is required, and as little as 99.7 percent removal will cause problems. The design engineer and plant owner (as represented by the operator) should consider maintaining tight control of this critical and sensitive ozone system component.

Ozone systems are energy intensive, and energy consumption varies as ozone production rate varies. Power usage rate at start-up may be as much as 3 to 4 times the rate at design, unless system flexibility is provided. Both start-up and design conditions should be analyzed during design. Automatic control of ozone production and energy use may be employed, but the extra capital cost, imprecise control abilities, and intensive maintenance requirements for the control equipment may not be justified by the reduced ozone production rate achieved. Each situation should be thoroughly evaluated. Manual control may be quite complex or more simplified. A simple approach reduces, but does not eliminate the need for some process monitoring equipment.

Ozone transfer efficiency (T.E.) is proportional to absorbed ozone dosage, which is proportional to the disinfection level achieved. Ozone may be absorbed through chemical reaction or through ozone/liquid gas dissolution. The ozone chemical reaction must be satisfied before effective disinfection can occur. Municipal/industrial wastes which have known or suspected ozone reacting pollutants should be analyzed using bench or pilot scale studies to determine the required absorbed ozone dosage to achieve the desired disinfection level. In all plants, the minimum acceptable T.E. should be based upon ozone/liquid gas dissolution theory.

Ozone may be detected (smelled) at levels about 1/10 the typical 8-hour human exposure standards. This constitutes a safety aspect of ozone systems. However, operators may become desensitized or careless; thus, ambient ozone monitors with alarms should be provided. Ozone concentrations in the contact basin feed and exhaust gas is several thousand times greater than the human exposure standard, and a tiny leak can cause excessive ambient ozone concentrations. System design and operation must address this fact.

TYPES OF OZONE SYSTEMS

Ozone systems may be categorized in several ways, depending on the topic to be emphasized. If the ozone feed stream is emphasized, three broad types of ozone systems exist: 1) air fed, 2) once through oxygen fed, and 3) recycle oxygen fed. A flow schematic of each type is shown in Figure 1. Oxygen fed systems generate about twice as much ozone per unit of electrical energy used. However, oxygen fed units are typically not cost effective, unless the oxygen can be used for another purpose, for example in the activated sludge process.

Each type of ozone wastewater disinfection system has an ozone generator, contact basin, and destruct unit. The air fed and oxygen recycle systems also have dew point treatment equipment. Dew point treatment for ozone generation is very sensitive and will be discussed in more detail later. Dew point treatment is typically not required for once through oxygen fed systems, because direct feed high purity oxygen is normally much dryer than required for ozone generation (-51°C dew point or dryer is desired for ozone generation).

Once through oxygen fed systems can be used if the oxygen requirement downstream of the ozone process is balanced with the oxygen requirement of the ozone process. Figure 2 illustrates a balanced oxygen usage graph for a once through oxygen fed ozone system and an oxygen activated sludge process. Figure 2 shows that when the activated sludge oxygen requirement is 1.1 kg O_2/kg $(BOD_5)_R$ and the BOD_5 removal rate is 150 mg/l, then the ozone concentration will be about 3 percent when the required dosage is 5 mg/l.

No dew point treatment equipment and correspondingly fewer operation and maintenance tasks are required when a once through oxygen fed ozone system is used. If the downstream oxygen consuming process requires about as much oxygen as the ozone process, then a once through oxygen ozone process should be considered. If significantly more oxygen is required downstream, then a controlled amount of oxygen may be bypassed around the ozone system. If significantly less oxygen is required downstream, an oxygen recycle system may be considered.

DEW POINT TREATMENT

Ozone generation equipment must be supplied with dry, particle-free gas. Filters are typically used to remove particles. Desiccant dryers plus in some cases refrigerant dryers are used to attain dry gas. Feed gas treatment is recommended if its dew point is -51°C or higher. A high dew point will result in lower ozone production, as shown in Figure 3 (1). Further, a high dew point will cause more rapid fouling and require more frequent cleaning of the generator; nitric acid formation (air and oxygen recycle systems) and damaged generator components; and may cause electrical short circuiting.

The feed gas dew point varies with its moisture content. The relationship between moisture content and dew point is shown in Figure 4. For example, when the moisture content of the feed gas is about 20 ppm by weight (at 1 atm pressure), the dew point is -51°C.

A relatively small change in moisture content will cause a significant change in dew point, especially in the range of operating dew point levels for ozone generators. An example for an air fed ozone system is shown in Table 1. If 99.9 percent of the moisture is removed, the dew point is satisfactory. However, if only 0.2 percent less moisture is removed (99.7 percent removal), the dew point is marginal to unsatisfactory! The importance of a well-designed and operated ozone feed gas dew point treatment unit is apparent.

TABLE 1. DEW POINT TREATMENT SENSITIVITY TO MOISTURE CONTENT

Process Equipment	Moisture Content (ppm by Weight)*	Moisture Removal (%)	Dew Point °C
Compressor	23,000	----	27
Refrigerant Dryer	5,000	78.3	4.5
Desiccant Dryer	20	99.9	-51
	80	99.7	-40

*From Figure 4.

The ozone feed gas dew point treatment equipment is usually provided, but not manufactured by the ozone generator equipment supplier. The ozone equipment manufacturer will purchase the dew point treatment equipment from other manufacturers, as needed. Limited design engineer control of the dew point treatment aspect of the ozone system design will cause a myriad of system and equipment options available to the ozone equipment suppliers. Because of the important and sensitive nature of this process, as discussed above, the plant owner (as represented by the operator) and design engineer should consider maintaining tight control over this area of system design.

Air dew point treatment processes can be either low [103 kN/sq m (15 psig)], medium [206 kN/sq m (30 psig)], or high pressure [688 kN/sq m (100 psig)] systems. Each has specific operation and maintenance advantages and disadvantages, which should be evaluated on a case-by-case basis. Equipment reliability, air flow control, power usage, turndown capability, and maintenance requirements are a few of the issues which should be evaluated. Equipment duplication and system flexibility also should be provided, because a small upset in dew point treatment can result in major problems with ozone generation capability.

Monitoring devices should be provided to measure and record the feed gas dew point continuously. Alarms to indicate a high dew point level also should be installed. However, care must be taken to insure that the sensitive dew point measuring equipment is giving accurate results. A "dew point cup" measuring device may be used to check and calibrate the in-line meter.

A procedure for using the dew point cup is described below. Refer to Figure 5 for a schematic of the dew point cup.

> A small stream of air is directed to the outside of a polished, stainless steel cup. The cup is filled about half full with acetone, the temperature of which is measured with a thermometer. Dry ice is gradually added to the acetone to decrease the temperature of the acetone. The temperature of the acetone and dry ice mixture is then read, and that reading is the air dew point. This dew point reading is at atmospheric pressure and must be adjusted to the actual pressure dew point of the in-line dew point monitor in order to calibrate the monitor properly.

OZONE GENERATION

Several different types of ozone generators are available including air cooled, water cooled, or oil and water cooled; and voltage controlled or frequency controlled units. Each manufacturer has prescribed advantages of his brand, and the design engineer may decide to choose one type or consider all types equally. The ozone generator, however, is only part of the ozone process. Equally important is feed gas treatment, ozone contacting, and ozone destruction. All units should be evaluated independently and also as they interrelate, one to the other.

One consideration for ozone system design is power consumption of the process. The relationship between power use rate and ozone production for an air feed ozone process is shown in Figure 6 (1). The rate of power usage for the ozone generator alone increases as the ozone production rate increases. However, power use rate for the total system (generator, feed gas treatment, and ozone destruction) decreases as ozone production rate increases. The reason for this occurrence is the relatively high, constant power demand of the feed gas treatment and ozone destruction equipment.

For the ozone system represented in Figure 6, the lowest power usage rate occurs at the design point of the process. However, most ozone systems used in wastewater disinfection probably will be operated at outputs much less than the design output because: 1) conservative estimates of maximum ozone dosage required may be used to size the ozone equipment, and 2) start-up plant flow rate will probably be less than the plant design flow rate. Both reasons cause the ozone production requirement to be less than design, and will cause inefficient power consumption unless system flexibility is provided to achieve lower power usage rates at lower ozone production rates. Both the start-up and design power usage rate should be thoroughly evaluated.

Power consumption of the ozone system increases as the ozone production rate increases; thus, an energy savings is realized when the ozone production requirement is decreased. The ozone production requirement is established by the level of disinfection to be achieved (kill rate), the ozone demand, and the ozone contact basin T.E. Ozone demand and contact basin T.E. are discussed later. Venosa, et.al. (4)(5) and Stover, et.al. (3) have shown that the kill rate is directly proportional to the absorbed ozone dosage; thus, to reach a desired level of disinfection a certain absorbed ozone dosage must be attained. The required absorbed ozone dosage may vary for different plants, because of water quality, but in each plant the ozone production rate would be used to adjust the amount of ozone absorbed.

Optimum process control for any given plant is achieved when the ozone production rate is as low as needed to achieve the required disinfection level. The two desired goals of good effluent quality and minimum energy consumption are met. If ozone production is greater than necessary, good effluent quality will still be achieved. The required level of disinfection will be met, and the residual ozone caused by the overdose will decompose back to oxygen fairly quickly (2). However, more energy will be consumed. To minimize ozone production yet achieve good effluent quality, automatic control of the ozone supply rate is often a consideration in system design.

Some of the ways in which automatic control of ozone supply may be completed are:

- Effluent ozone residual control - Interloop between ozone residual analyzer and ozone production equipment.
- Ozone dosage with wastewater flow control - Interloop between wastewater flow measurement and ozone production equipment.
- Ozone off-gas control - Interloop between ozone contact basin off-gas residual analyzer and ozone production equipment.
- Combination off-gas and wastewater flow control - Compound interloop between ozone contact basin off-gas residual analyzer plus wastewater flow meter and ozone production equipment.

Each of the automatic control systems available have varying degrees of equipment problems and somewhat imprecise control abilities. Also, they add to the initial cost of an ozone system. This higher cost may not be recovered if the overall ozone production level is not reduced by a substantial amount. Thus, the cost of this additional control equipment and its intense maintenance requirements may not be justified. Each situation should be thoroughly evaluated.

The alternative to automatic process control of an ozone system is manual control. Manual control requires that the operators adjust the ozone supply rate as the wastewater flow rate varies to achieve the required level of disinfection. This procedure is similar to simple chlorination system control. However, manual control of ozone systems does not cause the prob-

lem with water quality due to overdosing as chlorine systems cause. The only drawback to manual control of ozone is the higher energy cost that may occur because of overdosing.

Manual control of the ozone system may be fairly complex or quite simplified. The degree of complexity is dictated by the number of parameters measured and analyzed before an adjustment is made to the ozone production rate. The operators may analyze ozone residual, wastewater flow rate, and contact basin off-gas concentration data before adjusting the ozone supply rate. These data provide information about the current operating condition of the system that most directly relates to the disinfection kill rate, but requires more complex and sensitive equipment which results in added maintenance requirements.

A more simplified manual control procedure may be used to reduce both the initial equipment costs and on-going maintenance costs. The approach requires that the operators develop, for their system, a relationship between ozone dosage to the wastewater and the desired level of disinfection. The ozone production rate can then be adjusted as the wastewater flow rate varies, to achieve the prescribed ozone dosage. A procedure for a simplified manual control approach for the Vail, Colorado, ozone process is as follows:

When the required ozone dosage is established the ozone production rate is varied to meet that dosage at various wastewater flow rates. The required ozone production for various wastewater flow rates is shown in Figure 7. The example shows that at a wastewater flow rate of 10,200 m^3/day (2.7 mgd) and an ozone dosage of 4 mg/l, the required ozone production is 41 kg/day (90 lb/day).

When the required ozone production is established the ozone system must be adjusted to produce ozone at that rate. Two main factors influence the production of ozone; the air flow rate and the power supply (power supply controls the ozone concentration from the generator), as shown in Figure 8. At a given air flow rate, for example 1.98 m^3/min (70 scfm), the power supply adjustment will cause the ozone concentration to vary and hence, ozone production to vary. To achieve a given production rate, power supply is adjusted and the air flow rate is left constant. The example in Figure 8 shows that to reach 41 kg/day (90 lb/day) production at an air flow rate of 1.98 m^3/min (70 scfm), the power should be adjusted until the ozone concentration reaches about 7,100 to 7,200 ppm by volume.

The Vail ozone system has air flow meters and an ozone concentration meter that can be used to set the ozone production at the desired rate. Several combinations of air flow and ozone concentration can be used to achieve the desired ozone production rate. The most economical operating point should be selected. This point may be determined by conducting a special generator mapping test, and then referring to the "map" each time the production

rate is changed. An example "map" for an ozone system is shown in Figure 9. The ozone production rate would be achieved using less electrical energy at air flow rate "B" versus air flow rate "A". Therefore, the operator would use the proper equipment in the system to get air flow rate "B", then adjust the generator power supply to achieve the required ozone concentration established from Figure 8.

It should be noted that the simplified manual control procedure has not eliminated the use of all process measuring equipment. At least two instruments are recommended; an in-line dew point monitor and an ozone concentration meter. The dew point cup is used to check and calibrate the dew point monitor, as discussed earlier. Wet-chemistry testing is used to check and calibrate the ozone concentration meter. The wet-chemistry procedure for the Vail, Colorado, ozone system is presented below. The approach is applicable to other systems. Note that the Vail system is at an elevation of 2,470 m (8,100 feet) above sea level.

1. Set ozonator at desired power setting. Record generator information on data sheet (see Figure 10).
2. Check High Concentration Ozone Meter zero, span, control, and sample frequency readings and adjust to manufacturer's recommended setting, if necessary.
3. Prepare wet test chemistry equipment (see Figure 11).
   a. Add 400 ml of 2 percent KI solution to each of two 500 ml gas washing bottles (Note: A fritted glass diffuser is not used on ozone-air inlet tube).
   b. Connect gas washing bottles in series and connect ozone supply line and wet test meter.
   c. Level wet test meter and adjust water level in the meter.
4. Open vent valve and vent test line for 2 minutes.
5. Read and record three consecutive Ozone Meter readings.
6. Set valve to direct ozone-air gas flow to the gas washing bottles at a rate of 2 liters/minute.
7. Run approximately 3.0 liters of gas flow through the bottles and record field data information on data sheet (see Figure 10).
8. Take gas washing bottles to laboratory immediately and have another person read and record three more Ozone Meter readings.
9. Quantitatively transfer liquid from gas washing bottles to two separate 1 liter Erlynmeyer flasks. Rinse tubes and bottles at least three times.
10. Immediately add 10 ml of 2N Sulfuric Acid (H_2SO_4).
11. Read initial buret volume which contains 0.1N Sodium Thiosulfate solution ($Na_2S_2O_3$). Note: Standardize $Na_2S_2O_3$ using the dichromate method. (Standard Methods Ed. 14, pp. 316.)
12. Quickly titrate the darker of the two flasks to a pale yellow color with the $Na_2S_2O_3$.
13. Add 5 ml starch indicator (see Standard Methods Ed. 14, pp. 314 for starch preparation) and carefully titrate until clear.
14. Add 5 ml starch indicator to second flask and again carefully titrate, dropwise, until clear.

15. Record final buret reading and determine total volume of titrant used. Record on data sheet (see Figure 10).
16. Complete calculations on data sheet (see Figure 10).
17. Adjust span setting on Ozone Meter by following calculation:

$$\text{New span} = \text{old span} \left(\frac{\text{Laboratory } O_3 \text{ concentration}}{\text{Meter } O_3 \text{ concentration}}\right)$$

OZONE CONTACT BASIN

The ozone contact basin plays a key role in achieving acceptable disinfection with ozone. Earlier it was mentioned that the level of disinfection is related to the absorbed ozone dosage (3)(4). Contact basin T.E. is directly proportional to absorbed ozone dosage, as shown below.

$$\text{T.E.} = \frac{(\text{Mass of Absorbed Ozone})(100)}{\text{Mass of Applied Ozone}}$$

where: Mass of Absorbed Ozone = Mass of Applied Ozone minus Mass of Ozone in Off-Gas

The relationship among applied ozone dosage, absorbed ozone dosage, and T.E. is shown in Figure 12. The example lines 1, 2, and 3 show the level of applied ozone dosage required to achieve the same level of absorbed ozone dosage as the T.E. decreases. When the T.E. decreases from 90 percent to 80 percent to 70 percent, the applied ozone dosage is 111 percent greater, 125 percent greater and 143 percent of the absorbed ozone dosage, resepectively. Indeed, if the T.E. is only 50 percent, a full 200 percent more applied ozone dosage is needed. The point is that the level of applied ozone dosage required, and resulting level of ozone production needed, to achieve a given absorbed ozone dosage increases at a faster rate than the T.E. decreases. Therefore, to minimize ozone production requirements, T.E. should be maximized.

For a given applied ozone dosage, the absorbed ozone dosage increases as the ozone T.E. increases. Ozone absorption can occur through a direct chemical reaction with the pollutants in the wastewater and through ozone dissolution to the wastewater. Extremely high ozone T.E. can occur if the chemical reaction predominates, for example as with potassium iodide (KI) or with certain kinds of industrial wastes. For these cases the T.E. and absorbed ozone dosage may be high, but the disinfection kill rate will probably be low. The ozone chemical demand must be satisfied before effective disinfection can occur.

When the ozone chemical demand is satisfied, the ozone dissolution rate (gas to liquid transfer rate) will control the level of absorbed ozone at a given applied ozone dosage. Based on this premise, the following recommendations are made:

- The minimum acceptable ozone contact basin T.E. should be based upon ozone/liquid gas transfer theory.

- Wastewaters which contain known or suspected ozone chemical reactants (i.e., municipal/industrial wastes) should be analyzed using bench or pilot scale studies to determine the absorbed ozone dosage required to achieve the desired disinfection level.

Venosa, et.al. (4)(5) have addressed ozone/liquid gas dissolution in detail. Based on their findings, the following points should be considered in design.

- Ozone T.E. is governed by Henry's law, like oxygen transfer to water.

- Deep contactors using "fine" or intermediate bubble diffusers appear to provide the best assurance for ozone dissolution.

- High applied ozone concentrations appear to yield better ozone dissolution efficiencies.

OPERATOR SAFETY AND OFF-GAS OZONE DESTRUCTION

Typical standards call for a maximum allowable atmospheric ozone concentration for an 8-hour work day of 0.0002 mg/l by weight/volume (0.1 ppm by volume). Usually a person can smell ozone at a concentration of 1/10 this level (2). Therein lies a built-in safety feature of ozone systems; the operators usually are not exposed to ozone concentration levels at or above the accepted standards when they do not detect (smell) ozone in the environment. However, operators may become somewhat desensitized to ozone or somewhat less careful when continuously around ozone systems; thus, all ozone systems should have one or more ambient ozone monitoring devices to measure and record the ambient ozone concentration, sound an alarm when concentrations exceed a predetermined level, and automatically shut-down the ozone system immediately, or after a pre-set time alarm is not acknowledged within that time frame. Note: The latter approach avoids unnecessary system shut-downs due to false readings.

The concentration of ozone from the generator is typically between 12 to 24 mg/l by weight/volume, or 60,000 to 120,000 times greater than the typical 8-hour human exposure standard. As such, a tiny leak in the ozone supply piping can cause excessive ambient ozone concentrations. Extreme care should be used in design and installation of ozone pipe and equipment. A remote location for the ozone system, stainless steel piping, and other special precautions should be considered.

A good ozone contact basin may have a T.E. of 90 percent. At that T.E. the off-gas ozone concentration would be 1.2 to 2.4 mg/l by weight/volume. This concentration is 6,000 to 12,000 times greater than the typical 8-hour human exposure standard. An exceptionally high, probably unrealistic, T.E. is 99 percent. Yet at 99 percent T.E. the off-gas ozone concentration is still 600 to 1,200 times greater than the typical human exposure standard. The ozone discharged in the off-gas will dissipate, in time, but the half-

life of ozone in air is as long as 12 hours (2). The need for destruction of ozone in the off-gas is apparent!

Some off-gas ozone destruction treatment options include heat destruct, heat/catalyst destruct, activated carbon, recycle to sewage or sludge, and discharge through a tall stack. Heat destruct provides positive control of off-gas ozone concentration but requires a high power consumption. Heat/catalyst destruct also provides positive control of off-gas ozone concentration but requires some power consumption and periodic catalyst replacement. Activated carbon has an explosive potential when combined with ozone, which should be thoroughly analyzed if considered. Recycle to sewage or sludge does not provide positive control over off-gas ozone destruction and could transfer the problem to another area of the plant. Discharge through a tall stack also does not provide positive control over potential off-gas ozone contamination of the work environment. The off-gas ozone treatment options normally used are heat or heat/catalyst destruct units.

Special precautions should be employed to insure that the off-gas containing ozone does not bypass the destruct unit. Also, foam suppression equipment should be installed in the off-gas removal piping to keep the foam from coating and contaminating the heating coils or catalyst equipment. The foam suppression equipment should be simple to operate, routinely checked by the operators, and easy to maintain.

The ozone concentration of the off-gas ozone destruction system should be measured on a periodic basis (weekly or monthly) to monitor the performance of the process. The procedure to measure the off-gas ozone concentration gas streams to and from the ozone destruct equipment is similar. The procedure for measuring the inlet off-gas ozone concentration for Vail, Colorado, is described below. The approach is applicable to other systems. Note that Vail is 2,470 m (8,100 feet) above sea level.

1. Prepare wet test chemistry equipment (see Figure 13).
   a. Add 400 ml of 2 percent KI solution to one gas washing bottle.
   b. Connect wash bottle to test line and wet test meter.
   c. Connect vacuum line to wet test meter vent.
   d. Level wet test meter and adjust water level in the meter.
   e. Open vacuum valve until moderate gas flow rate is established.
2. Run approximately 12 liters, or more if necessary, of gas flow through the bottle and record field information on data sheet (see Figure 14).
3. Take gas washing bottle to laboratory immediately.
4. Quantitatively transfer liquid from gas washing bottle to a 1 liter Erlynmyer flask. Rinse tube and bottle at least three times.
5. Immediately add 10 ml of 2N Sulfuric Acid (H_2SO_4).
6. Read initial buret volume which contains 0.1N Sodium Thiosulfate solution ($Na_2S_2O_3$). Note: Standardize $Na_2S_2O_3$ using the dichromate method. (Standard Methods Ed. 14, pp. 316).
7. Quickly titrate to pale yellow with $Na_2S_2O_3$.
8. Add 5 ml starch indicator (see Standard Methods Ed. 14, pp. 317 for starch preparation) and carefully titrate until clear.

9. Record final buret reading and determine total volume of titrant used. Record on data sheet (see Figure 14).
10. Complete calculations on data sheet (see Figure 14).

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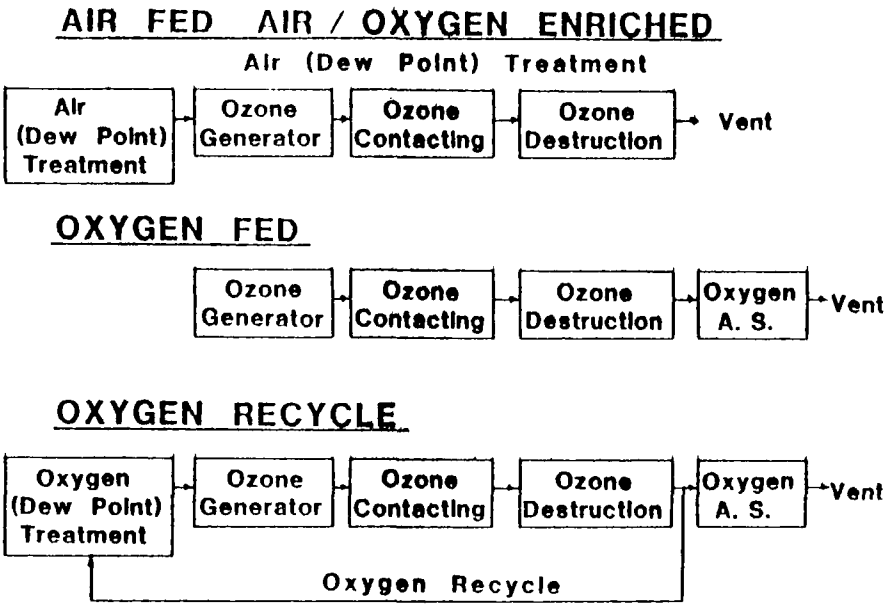


Figure 1. Line diagram for three types of ozone systems.

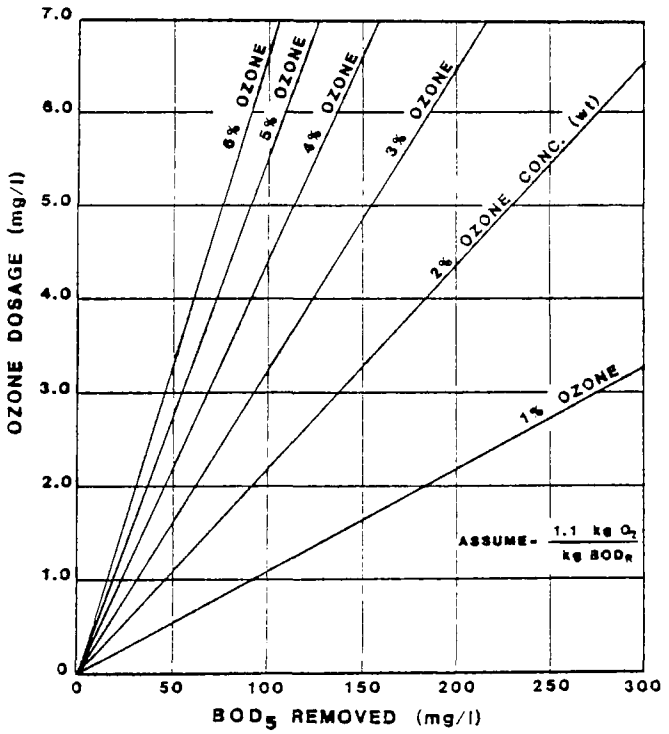


Figure 2. Required ozone concentration of various ozone dosages and BOD_5 removal rates for a once through oxygen/ozone and oxygen/activated sludge system.

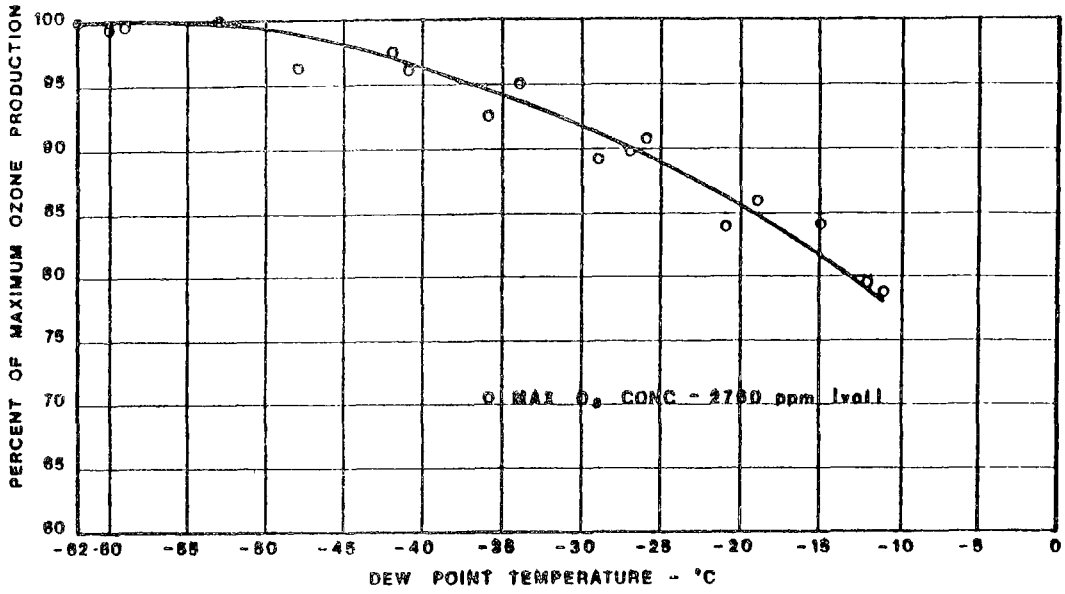
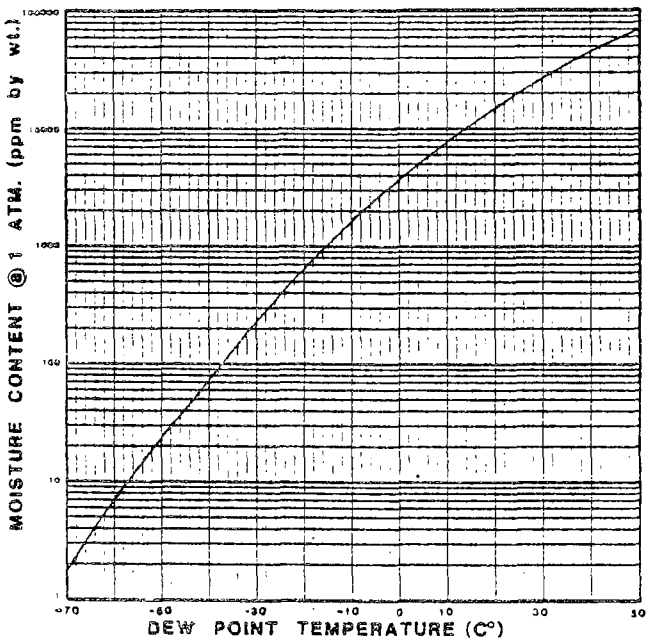


Figure 3. Ozone production rate decreases as feed gas dew point temperature increases.



DEW POINT vs. MOISTURE CONTENT

Figure 4. Feed gas moisture content increases as its dew point increases.

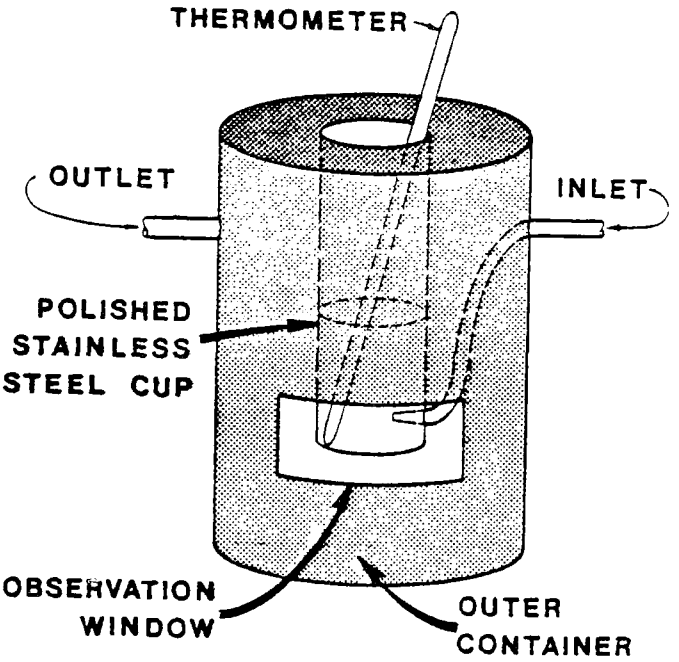


Figure 5. Diagram of dew point cup feed gas dew point measuring device.

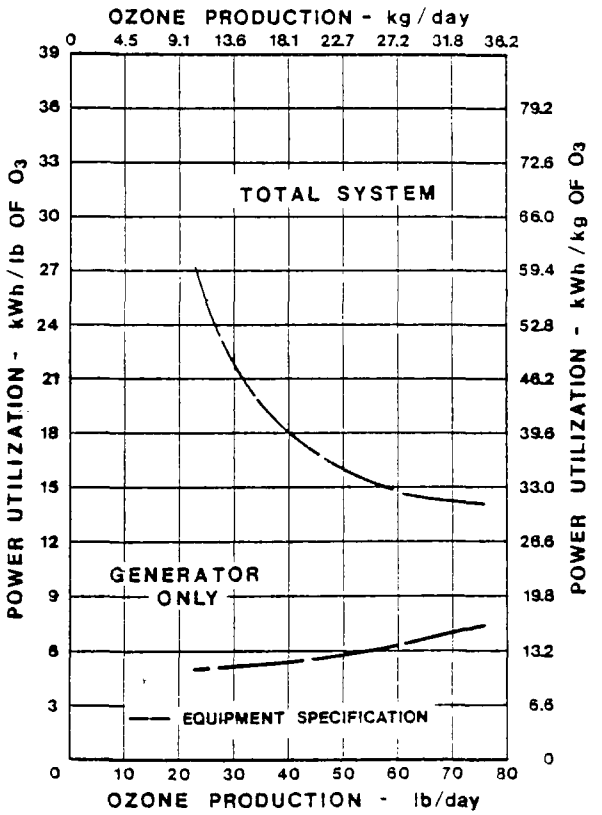
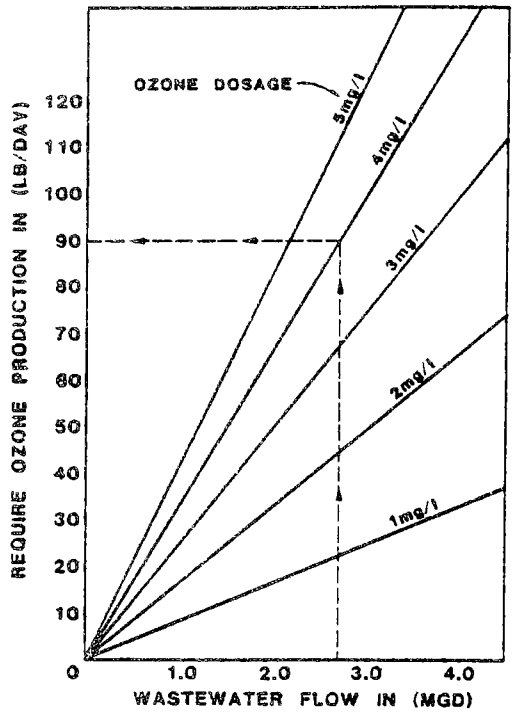


Figure 6. Power utilization varies as ozone production varies for the ozone generator and for the total ozone system.

Figure 7. Ozone production rate required at various wastewater flow and ozone dosages (lb/day x 0.453 = kg/day and mgd x 3785 = m^3/day



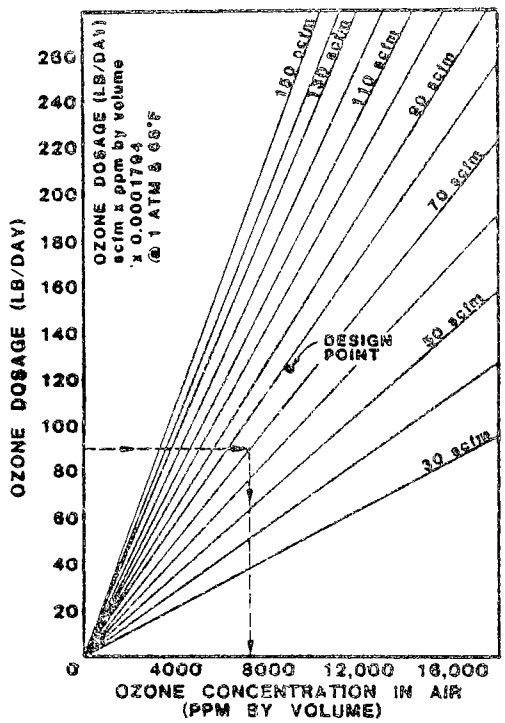


Figure 8. Ozone concentration required to achieve the desired ozone dosage at various feed gas flow rates (lb/day x 0.453 = kg/day).

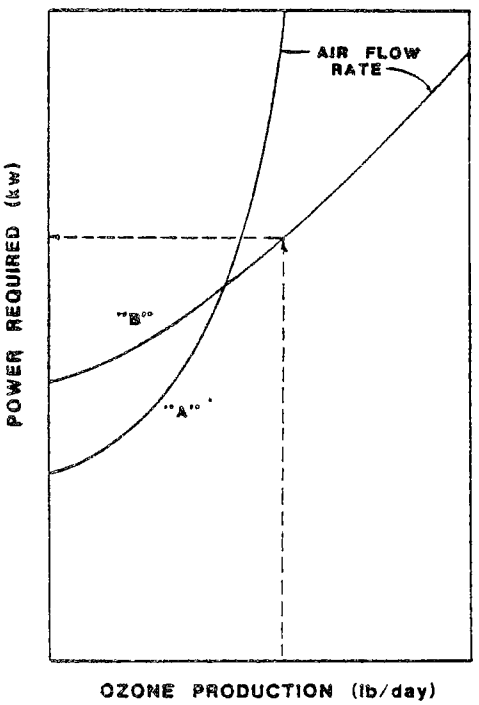


Figure 9. Example ozone generator "map" describing the most efficient operating point.

Date _____ Time of Analysis _____

FIELD INFORMATION

Generator: No. _____ Voltage _____volts Amperage _____amps

Air Pretreatment: Volume _____cfm Pressure _____psig Temperature _____°F Dewpoint (_____°F)

Ozone Meter: Span _____ Reading before _____ _____ _____ Reading after _____ _____ _____ Average _____

Wet Test Meter: Volume _____L Temperature ____°F Manometer ____in. H_2O (Suction is negative) Water Vapor Pressure _____in. H_2O (see Table)

LAB INFORMATION

Wet Test Meter: Titration _____mls N of $Na_2S_2O_3$ _____mole eq/L

GENERATOR OUTPUT AND APPLIED OZONE DOSAGE CALCULATIONS

Ozone Concentration:

Calculate weight of ozone trapped in KI solution.

$$WT = \left(\frac{N \text{ mole eq}}{L}\right)(\text{Titration ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{gm ml}}\right)$$

$$WT = \left(\frac{\text{\_\_\_\_\_ mole eq}}{L}\right)(\text{\_\_\_\_\_ ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{gm ml}}\right) = \text{\_\_\_\_\_ mg}$$

Calculate volume of gas that passed through wet test meter.

$$V_2 = (V_1)\left(\frac{P_1}{P_2}\right)\left(\frac{T_2}{T_1}\right) = (\text{\_\_\_\_\_ L})\left(\frac{\text{\_\_\_\_\_ in } H_2O}{406.8 \text{ in } H_2O}\right)\left(\frac{527.6°R}{\text{\_\_\_\_\_ °R}}\right) = \text{\_\_\_\_\_ L}$$

Where: V_1 = Actual volume in L

P_2 = Standard pressure = 402.8 in. H_2O

P_1 = Adjusted pressure = (Plant atmospheric pressure (8100 ft) of 301 in H_2O) - (water vapor pressure) + (wet test manometer pressure - Note: suction is negative). P_1 = _____ - _____ + _____ = _____ in. H_2O

T_2 = Standard temperature (absolute) = 68°F + 459.6 = 527.6°R

T_1 = Actual temperature (absolute) = _____°F + 459.6 = _____°R

Calculate ozone concentration.

$$mg/L \text{ air} = (\text{\_\_\_\_\_ mg})\left(\frac{1}{\text{\_\_\_\_\_ L}}\right) = \text{\_\_\_\_\_ mg/L air}$$

$$ppm/vol = \left(\frac{\text{\_\_\_\_\_ mg}}{L \text{ air}}\right)(1{,}000{,}000)\left(\frac{1}{1997}\right) = \text{\_\_\_\_\_ ppm/vol (20°C)}$$

Ozone Supply:

Calculate ozonated air flow rate.

$$V_2 = V_1 \left[\left(\frac{P_1}{P_2}\right)\left(\frac{T_2}{T_1}\right)\right]\frac{T_3}{T_2} = (\text{\_\_\_\_\_ cfm})\left[\frac{\text{\_\_\_\_\_ psia}}{20.7 \text{psia}}\right]\left(\frac{529.6°R}{°R}\right)(1) = \text{\_\_\_\_\_ scfm (20°C)}$$

Where: V_1 = Actual Volume in Ft^3

P_2 = Standard pressure (absolute) = Gauge pressure + atmospheric pressure = 6 psig + 14.7 psi = 20.7 psia

P_1 = Actual pressure (absolute) = Gauge reading (psig) + plant atmospheric pressure (8100) of 10.88 psi

_____psig + 10.88 psi = _____ psia

T_2 = Standard temperature for rotometer (absolute) = 70°F + 459.6 = 527.6°R

T_1 = Actual temperature for rotometer (absolute) = _____°F + 459.6 = _____°R

T_3 = Standard temperature of 20°C (absolute) = 68°F + 459.6 = 527.6°R

Calculate ozone supply rate.

$$lbs/day = (\text{\_\_\_\_\_ } ft^3/min)(\text{\_\_\_\_\_ mg/L air})(28.32 \text{ L/ft}^3)\left(\frac{1}{453{,}400}\frac{lbs}{gm}\right)(1440 \text{ min/day}) = \text{\_\_\_\_\_ lbs/day}$$

Water Vapor Pressure

Wet Test Meter Temperature (°F)	Water Vapor Pressure (in H_2O)
42-47	4
47-53	5
54-58	6
59-63	7
64-66	8
67-69	9
70-72	10
73-75	11
76-77	12
78-79	13
79-80	14

Figure 10. Example wet-chemistry, ozone production concentration data sheet.

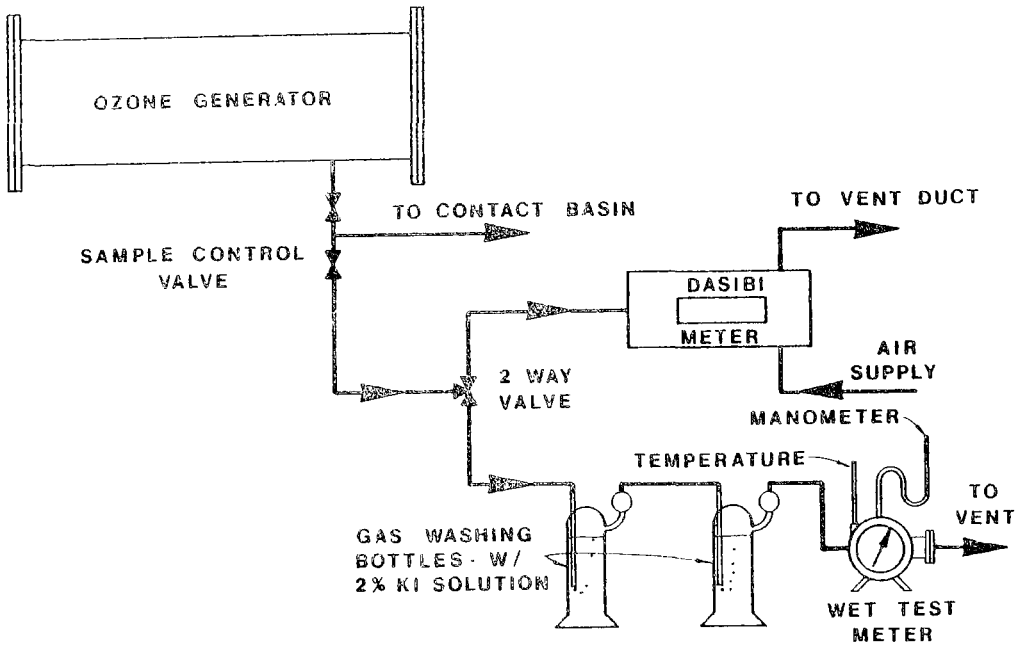


Figure 11. Example wet-chemistry, ozone production concentration testing equipment set-up.

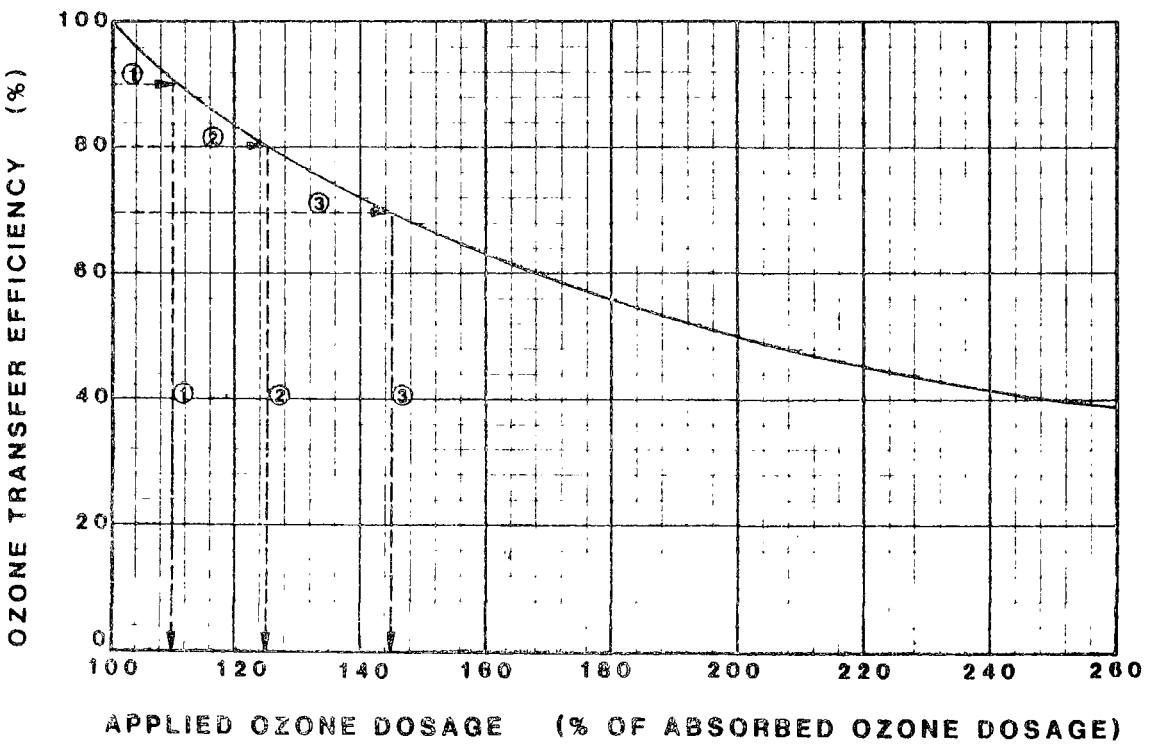


Figure 12. Applied ozone dosage increases significantly as ozone transfer efficiency decreases.

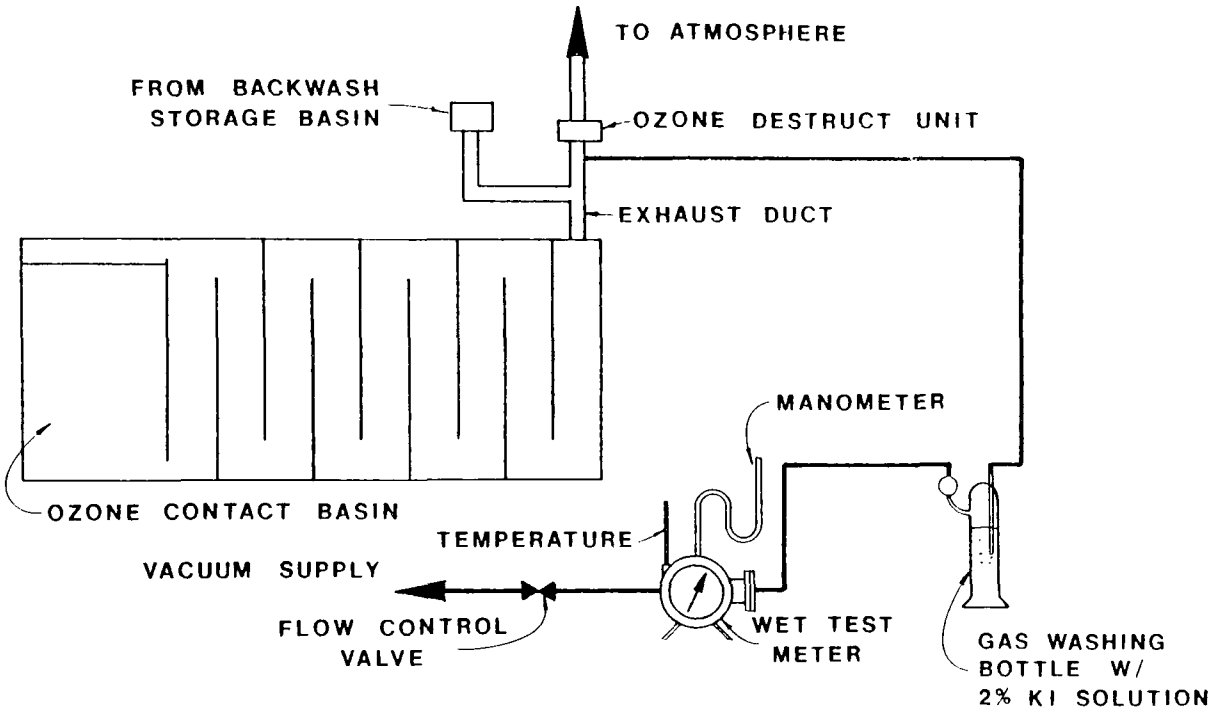


Figure 13. Example wet-chemistry, off-gas concentration testing equipment set-up.

Date _____ Time _____ Location _____

FIELD INFORMATION

Off-Gas: Flow _____ cfm $\left(\frac{301}{406.8}\right)\left(\frac{527.6}{\ ^{\circ}F+459.6}\right)$ = _____ scfm (20°C)

Wet Test Meter: Volume _____ L Temperature _____ °F Manometer _____ in. H_2O (Suction is negative) Water Vapor Pressure _____ in. H_2O (see Table)

LAB INFORMATION

Wet Test Meter: Titration _____ mls N of $Na_2S_2O_3$ _____ mole eq/L

CALCULATIONS

Ozone Concentration in Off Gas:

Calculate weight of ozone trapped in KI solution.

$$WT = \left(\frac{N \text{ mole eq}}{L}\right)(\text{Titration ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{gm ml}}\right) = (\text{\_\_\_\_\_ mole eq/L})(\text{\_\_\_\_\_ ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{mg gm}}\right) = \text{\_\_\_\_\_ mg}$$

Calculate volume of gas that passed through wet test meter.

$$V_2 = (V_1)\left(\frac{P_1}{P_2}\right)\left(\frac{T_2}{T_1}\right) = (\text{\_\_\_\_\_ L})\left(\frac{\text{\_\_\_\_\_ in } H_2O}{406.8 \text{ in } H_2O}\right)\left(\frac{527.6\ ^{\circ}R}{\text{\_\_\_\_\_}\ ^{\circ}R}\right) = \text{\_\_\_\_\_ l}$$

Where: V_1 = Actual volume in L

P_2 = Standard pressure = 402.8 in. H_2O

P_1 = Adjusted pressure = (Plant atmospheric pressure (8100 ft) of 301 in H_2O) − (water vapor pressure) + (wet test manometer pressure - Note: suction is negative). P_1 = _____ − _____ + _____ = _____ in. H_2O

T_2 = Standard temperature (absolute) = 68°F + 459.6 = 527.6°R

T_1 = Actual temperature (absolute) = _____°F + 459.6 = _____°R

Water Vapor Pressure

Wet Test Meter Temperature (°F)	Water Vapor Pressure (in H_2O)
42-47	4
47-53	5
54-58	6
59-63	7
64-66	8
67-69	9
70-72	10
73-75	11
76-77	12
78-79	13
79-80	14

Calculate ozone concentration in off gas.

$$mg/L \text{ air} = (\text{\_\_\_\_\_ mg})\left(\frac{1}{\text{\_\_\_\_\_ L}}\right) = \text{\_\_\_\_\_ mg/L air}$$

$$ppm/vol = \left(\frac{\text{\_\_\_\_\_ mg}}{\text{L air}}\right)(1{,}000{,}000)\left(\frac{1}{1997}\right) = \text{\_\_\_\_\_ ppm/vol (20°C)}$$

Residual Ozone Concentration:

Calculate weight of ozone trapped in KI solution.

$$WT = \left(\frac{N \text{ mole eq}}{L}\right)(\text{Titration ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{gm ml}}\right) = (\text{\_\_\_\_\_ mole eq/L})(\text{\_\_\_\_\_ ml})\left(\frac{24\text{gm } O_3}{\text{mole eq}}\right)\left(\frac{1.0 \text{ L mg}}{\text{mg gm}}\right) = \text{\_\_\_\_\_ mg}$$

Calculate residual ozone concentration.

$$mg/L\ H_2O = (\text{\_\_\_\_\_ mg})\left(\frac{1}{\text{\_\_\_\_\_ ml of sample}}\right)\left(\frac{1000 \text{ ml}}{L}\right) = \text{\_\_\_\_\_ mg/L } H_2O$$

Ozone Lost in Vent:

Calculate ozone lost in vent.

$$lbs/day = (\text{\_\_\_\_\_ ft}^3/\text{min})(\text{\_\_\_\_\_ mg/L air})(28.32 \text{ L/ft}^3)\left(\frac{1}{453{,}600}\ \frac{lbs}{gm}\right)(1440 \text{ min/day}) = \text{\_\_\_\_\_ lbs/day}$$

Ozone Transfer:

$$\text{Percent ozone transfer} = \frac{(\text{ozone supply rate (lbs/day)} - \text{ozone lost in vent (lbs/day)})(100)}{\text{ozone supply rate (lbs/day)}}$$

$$= \frac{(\text{\_\_\_\_\_ (lbs/day)} - \text{\_\_\_\_\_ (lbs/day)})(100)}{\text{\_\_\_\_\_ (lbs/day)}} = \text{\_\_\_\_\_ \%}$$

Figure 14. Example data sheet for ozone off-gas concentration measurements.

3. THE EFFECTS OF OPERATION AND MAINTENANCE PRACTICES ON SELECTED OZONE AND ULTRAVIOLET DISINFECTION SYSTEMS

by: Randy Junkins, Manager O&M Section
WESTON, Designers-Consultants
West Chester, Pennsylvania

INTRODUCTION

Increased attention has been given in recent years to the disinfection of municipal wastewaters via methods other than conventional chlorination. Two alternative approaches that have generated particular interest are ozone and ultraviolet light (UV) disinfection. As part of EPA's efforts to compile and subsequently promulgate design and operational information concerning these two technologies, Roy F. Weston, Inc. was contracted (EPA Contract No. 68-03-3019) to identify operations and maintenance factors affecting the performance of ozone and UV disinfection systems. This paper presents the study methodology utilized by WESTON and discusses the project results to date.

The objective of the nine-month study is to determine, analyze, and prioritize those O&M factors that affect the operational efficiencies of ozone and UV disinfection systems. During the study, on-site evaluations will be conducted at 15 municipal wastewater treatment plants that utilize either ozone or UV disinfection. During these plant visits, operating personnel will be interviewed, operational practices will be observed, and operating data reviewed in order to establish O&M causative factors relating to poor and efficient process performance. This information will be documented in individual plant evaluation reports.

The project final report will integrate the data collected and observations made at the 15 treatment plants into a comprehensive O&M document. The report will present the O&M problems encountered, conclusions drawn concerning their cause, and recommendations made toward their resolution. Recommendations presented will address operating practices, process changes, monitoring and sampling techniques, staffing requirements, operator training, and maintenance procedures.

STUDY METHODOLOGY

The study methodology formulated to accomplish the project goals is shown in Figure 1. Initially, existing data concerning the operation of ozone and UV disinfection systems and descriptive information about the 15 treatment plants to be evaluated will be collected and reviewed. Simultaneously, a preliminary telephone survey of the treatment plants will be conducted in

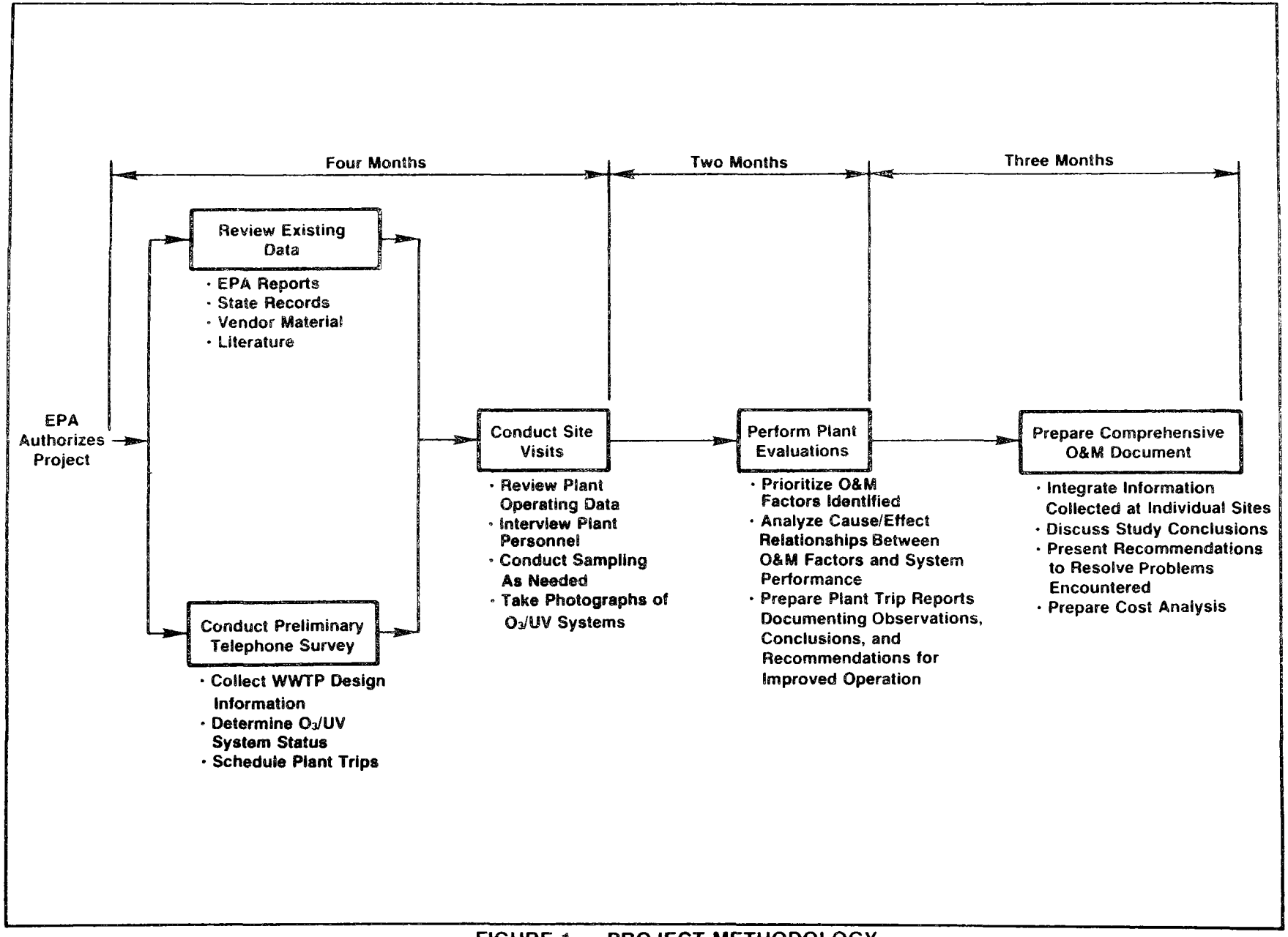


FIGURE 1 PROJECT METHODOLOGY

order to gather design information and schedule plant trips. Site visits will subsequently be made to gather plant operating data and identify those O&M factors which affect the performance of ozone and UV disinfection systems. Following completion of the field trips, the data collected will be analyzed and cause/effect relationships between the O&M factors identified and system efficiencies will be formulated. Finally, the project results will be reported in a comprehensive O&M document that includes recommendations for optimizing process performance.

PROJECT STATUS

Originally the nine-month project was to be initiated during the first few weeks in 1981. However, due to EPA budget cutbacks, the project was delayed and WESTON was not authorized to proceed until late December 1981.

The project is presently in the initial data collection and review phase. The preliminary telephone survey has also been started and is the primary source of the data presented in this paper. It is anticipated that the treatment plant visits will begin within the next two weeks. The project is scheduled for completion in September 1982.

PRELIMINARY PROJECT RESULTS

Information collected to date concerning the treatment plants to be visited, and specifically their disinfection unit operations, is discussed below. The preliminary project results presented will be confirmed, expanded, and refined following the individual site visits.

PLANTS UTILIZING OZONE DISINFECTION

Descriptive information about the plants which use ozone disinfection, and design data concerning their disinfection systems are presented in Tables 1 and 2. It can be seen that plant hydraulic sizes ranged from 303 to 71,915 m^3/d (0.08 to 19 mgd) and ozone system capacities varied from 6.35 to 1905 kg.d (14 to 4,200 ppd). It is also noted that fifty percent of the plants contacted utilized a pure oxygen activated sludge process which included second stage nitrification. The number of plants which used air and oxygen as the ozone carrier gas was also evenly divided.

Information concerning disinfection systems performance and various operation and maintenance considerations is presented in Tables 3 and 4. It can be seen that both operational efficiencies and costs varied greatly.

TABLE 1. OZONE WWTP'S - PLANT DESCRIPTIONS

Plant no.	Plant age yrs.	Type*	Avg. flow m^3/d (mgd)	Eff. quality BOD mg/l	Eff. quality SS mg/l	Pre-treatment
1	2	RBC	303 (0.08)	12	3	Filt.
2	2	O_2-A.S. + Nit.	45,420 (12.0)	20	30	None
3	1.5	A.S.	1,325 (0.35)	3	2	Filt.
4	1.5	O.D.	17,033 (4.5)	20	20	None
5	4	O_2-A.S. + Nit.	71,915 (19.0)	2	2	Filt.
6	1	RBC + A.S.	7,570 (2.0)	10	12	Filt.
7	2	O_2-A.S. + Nit.	18,925 (5.0)	9	8	Filt.
8	3	O_2-A.S. + Nit.	10,977 (2.9)	3	3	Filt.

*RBC - Rotating Biological Contactor
A.S. - Activated Sludge
O_2-A.S. - Pure Oxygen Activated Sludge
Nit. - Nitrification
O.D. - Oxidation Ditch

TABLE 2. OZONE WWTP'S - OZONE SYSTEM DESCRIPTIONS

Plant no.	Number generators	Cells/gen.	Capacity total kg.d (ppd)	Carrier gas	Oz. transfer efficiency
1	2	8	6.35 (14)	Air	---
2	17	90	1,905 (4,200)	O_2	---
3	2	16	77.1 (170)	Air	---
4	3	109	272.2 (600)	Air	60%
5	13	90	1,562.7 (3,445)	O_2	86%
6	2	72	113.4 (250)	Air	67%
7	3	72	340.2 (750)	O_2	84%
8	3	90	571.5 (1,260)	O_2	---

TABLE 3. OZONE WWTP'S - OZONE SYSTEM PERFORMANCE

Plant no.	Operational mode		Contact time min.	Ozone dosage mg/l	Coliform count/100 ml	
	Auto.	Manual			Inf.	Eff.
1	x		15	9	---	3,800
2	x		15	22	---	1,000
3		x	300 (60 min. design)	3.5	18,000	12
4	x		50	3.0	350,000	200
5		x	30	3.0	---	<1
6	x		60	10.0	30,000	3,000
7		x	40	7	---	100
8		x	75	6	20,000	100

TABLE 4. OZONE WWTP'S - O&M CONSIDERATIONS

Plant no.	Maintenance		Power requirements kwh/kg oz (kwh/lb oz)	Capital $	Annual ¢/m^3 (¢/1,000 gal)
	Downtime	Time reqts. hrs/wk			
1	---	---	---	74,000	---
2	Minimal	---	---	---	0.71 (2.7)
3	10%	8	---	---	2.01 (7.6)
4	Minimal	4	22 (10)	500,000	---
5	70%	8	11 (5)	---	0.34 (1.3)
6	High	8-12	26 (12)	200,000	0.84 (3.2)
7	30-50%	8	48 (22)	---	0.92 (3.5)
8	---	4	---	---	---

Typical O&M problems encountered, and associated suggestions for improved operation (determined during conversations with plant personnel) are listed below. Most of the problems identified were maintenance-related items.

Ozone WWTP's - Typical O&M Problems

- Multiple and frequent Ozone Generator cell failures.
- Silicone control rectifier (SCR) failures.
- Severe foam problems with contact tank gas recovery system.
- Ozone system electronics are complicated, making it difficult for WWTP personnel to perform routine maintenance and repair work.
- Corrosion problems with O_3 analyzer valve components.
- Ozone contact tanks were constructed below the system control room and O_3 leaks cause instrumentation rubber seals to corrode.
- System equipment is very noisy.
- The system includes much equipment which must be maintained.
- Much time is required to continually calibrate system instrumentation.
- Ozone generators are a high maintenance item and continually blow fuses.
- Catalyst poisoning in O_3 destruction system.
- Dew point indicators are not reliable.
- Excessive heat build-up in O_3 generator room caused generator heaters to shut down units.
- A full-time instrumentation person is needed to monitor and maintain O_3 system.

Ozone WWTP's - Operator Suggestions

- Ensure an adequate air filtration system is installed.
- An experienced and qualified instrumentation person must be part of the WWTP's operating staff.
- Ozone contact tanks should be sited away from other buildings and equipment to avoid corrosion and safety problems.
- An O_3 destruction unit may not be required if contact tank off-gases can be vented in an isolated area away from WWTP buildings.
- System should include a carrier gas O_3 monitor to aid in determining when unit needs to be cleaned.
- Provide a dry gas purge system for compressors.
- Provide a foam suppression system in contact tank and for gas destruction system.
- Provide ample air circulation in O_3 generator room to prevent excessive heat build-up.
- Equipment should be housed inside a building for protection and prevention of corrosion and freezing problems.
- The ozone generator room must be kept very clean, otherwise the generator cells will short-out and blow fuses.

PLANTS UTILIZING UV DISINFECTION

Design information about the two plants contacted that utilize UV disinfection is presented in Tables 5 and 6. Data concerning the performance of their UV systems is shown in Table 7. It can be seen that both systems operate very effectively. Although no cost data were available from the individual plants, cost information presented in a previous EPA report on UV disinfection is indicated in Table 8. Typical O&M problems reported during the telephone survey are listed.

TABLE 5. ULTRAVIOLET WWTP'S - PLANT DESCRIPTIONS

Plant no.	Age mo.	Type*	Avg. flow m^3/d (mgd)	Eff. quality BOD mg/l	Eff. quality SS mg/l	Pretreatment
1	3	A.S.	5,678 (1.5)	15	15	None
2	4	Aer. Lag.	7,570 (2.0)	2	2	Filt.

*A.S. - Activated sludge
Aer. Lag. - Aerated lagoon

TABLE 6. ULTRAVIOLET WWTP'S - UV SYSTEM DESCRIPTIONS

Plant no.	No. sections	No. lamps/sec.	Lamp output $\mu w/cm^2$	Cleaning mechanism	Flow/unit m^3/d (mgd)
1	2	90	---	Pneumatic Scraper	5,678 (1.5)
2	4	32	190	Mechanical Wiper	8,327 (2.2)

TABLE 7. ULTRAVIOLET WWTP'S - UV SYSTEM PERFORMANCE

Plant no.	Contact time sec.	Dosage $\mu W\text{-sec/cm}^2$	Coliform count/100 ml. Inf.	Eff.
1	3.5	---	---	70
2	1.5	30,000	5	<1

TABLE 8. ULTRAVIOLET WWTP'S - PREVIOUS EPA STUDY COST DATA

Plant flow mgd	Capital cost $	Annual O&M ¢/m^3 (¢/1,000 gal)
1	80,000	0.66 (2.5)
10	700,000	0.53 (2.0)
100	5,200,000	0.48 (1.8)

Ultraviolet WWTP's - Typical O&M Problems

- Ballasts on UV lamps overheat and shut system down.
- Foam build-ups interfere with operation of cleaning mechanism.
- Low flow rate caused unit to overheat.
- Algae accumulations on unit interfere with system operation.

COMPARISON OF OPERATING COSTS

Estimated operating cost for various alternative disinfection processes are compared in Table 9. These estimates were prepared as part of a previous EPA study on wastewater disinfection. The data indicate that UV disinfection appears to be the most cost-effective strategy for smaller treatment plants that treat 1 mgd or less, while chlorination (even with de-chlorination) is the most economical approach for larger plants.

TABLE 9. COMPARISON OF ALTERNATIVE DISINFECTION PROCESSES OPERATING COSTS

Flow mgd	Ultraviolet	Chlorination	Chlorination/ De-Chlorination	Ozonation-air
1	1.19 (4.5)	1.69 (6.4)	2.19 (8.3)	3.41 (12.9)
10	0.95 (3.6)	0.74 (2.8)	0.82 (3.1)	2.01 (7.6)
100	0.82 (3.1)	0.58 (2.2)	0.61 (2.3)	1.45 (5.5)

Costs defined as ¢/cm^3 (¢/1,000 gal.).

GENERAL OBSERVATIONS

The following preliminary general observations are made:

Ozone Systems

1. Many of the ozone systems surveyed had only been in operation three to four months.

2. Minimal cost and maintenance requirements information is currently available for full-scale installations.

3. Very few of the same problems were encountered at the treatment plants surveyed.

4. The consensus of opinion among the plant operators interviewed is that chlorine disinfection systems are more reliable, less expensive to operate, and require less maintenance than ozone systems.

Ultraviolet Systems

1. Presently there are very few full-scale UV systems on-line.

2. Available O&M data concerning UV disinfection are minimal.

3. Those UV systems which are on-line were reported to be reliable unit operations.

REFERENCES

1. EPA Project Summary Report; EPA 600/52-81-152, Sept. 1981.

4. SECOND NATIONAL SYMPOSIUM ON MUNICIPAL WASTEWATER DISINFECTION - SUMMARY AND CLOSING REMARKS

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INTRODUCTION

Water, more perhaps than any other medium, illustrates the recycling process that takes place in nature. All waste that is discharged to the biosphere - biological,chemical,and physical-sooner or later finds its way into the earth's water. That water must be cleansed by nature or by man before it is again safe for human use and consumption. For 2-1/2 days some 250 persons from 33 states and 2 foreign countries have been discussing this phenomenon as it relates to wastewater treatment plant effluents. Fifty percent of these persons are representatives of government agencies, 20 percent equipment manufacturers and suppliers, 20 percent consulting engineers and 10 percent academicians. During the next few minutes I will try to capsulize the papers and discussions of this conference and along the way add some comments that reflect my own point of view on wastewater disinfection. To do this I will divide my remarks into two general segments - health consideration associated with discharge of wastewater and the concerns of this conference, and the technological considerations related to disinfection practices.

HEALTH CONSIDERATIONS

Health considerations we have discussed embrace - arguments for and against chlorination of sewage effluents. Jim Coulter presented strong arguments and factual data against chlorination of Wastewater Treatment Plant effluents citing the negative impact on fish populations in Maryland. Henry Ongerth countered forcefully citing the generally recognized preventive health arguments against the uncontrolled discharge to the environment of waste potentially harmful to humans. The question is now raised as to whether there must be this confrontation between fish and people? Before I attempt an answer lets look at other discussions that shed light on the basic question of what public health evidence supports the need for disinfection of these effluents.

Elmer Akin provided information on "Infective Dose of Waterborne Pathogens." He reported on studies involving bacterial, protozoal and viral pathogens. The data indicated that all these categories of enteric pathogens can produce infection and/or illness at very low exposure levels. All the doses were administered to the human volunteers by the oral route. As recognized by one participant, the lesson learned in this case is clear - don't drink contaminated water.

Dr. Dolin related a case study on an outbreak of viral gastroenteritis caused by a Norwalk-like virus. The outbreak accurred after a water supply source was contaminated with sewage from a broken sewerline. Once again we prove the point - don't drink contaminated water.

Now I am not being facetious in my comments on these two studies. Unfortunately, humans inadvertently do ingest water when swimming and these waters when contaminated are capable of producing disease. The question would appear to be what level of contamination can be considered reasonably safe under these conditions?

Dr. Hubly provided some incite into risk assessment as related to the use of chlorine in wastewater disinfection. His investigation uncovered very limited to nonexistant historical data. We all know that risk assessment of potentially harmful environmental impacts is an emerging science and in the absence of background data little reliance can be placed on predictions of risk associated with the use of chlorine in wastewater disinfection. All in all though it would seem that the risk is minimal.

The luncheon speaker Dr. Arthur Lane of the Jet Propulsion Laboratory took us on a most interesting and exciting photographic voyage to Jupiter and Saturn. The pictures were simply fantastic.

Once we were back to earth, Walter Jakubowski reported on a series of epidemiological studies of community exposures to aerosols from wastewater treatment plants and a study of worker exposure to aerosols, sewage liquids, and solid contacts. One could not conclude from these study results that the communities were harmed by aerosol spray eminating from the wastewater treatment plant. With respect to sewage treatment plant workers, inexperienced workers evidenced higher rates of gastrointestinal symptoms than did experi enced workers or controls. The symptoms were mild and transitory and did not result in time lost from work. Pathogen isolation did not indicate any increase risk from sewage exposure. The results of two studies from Israel related to spray irrigation of sewage were found to be inconclusive because of the poor quality of the data. A third study is underway.

Dr. Cabelli's epidemiological studies enabled the development of a predictive model which is intended to aid in the determination of requirements for wastewater disinfection of bathing beaches. While the model has not been validated it offers a tool that is otherwise unavailable for this purpose. Perhaps the validation should and can be obtained through its use.

While Dr. Cabelli's work was associated with marine waters, Dr. Dufour investigated fresh recreational water quality and swimming associated illnesses. General conclusions would suggest that as water quality deteriorates the potential for disease increases. Further, as in marine waters, entercocci probably represent the best indicator organism for recreational waters. Because fresh water swimming associated gastrointestinal rates are lower than in marine water, different water quality standards should apply. One additional thought is worth recalling. The unanticipated illnesses associated with waters of relatively high quality may be associated with particle ingestion. This observation certainly warrants further study.

We should ask ourselves how is all this related to discussions by Coulter and Ongerth? It tells me that the state of knowledge with respect to the potential for harm to the public health from discharge of unchlorinated sewage to the environment is a big question mark. On the other hand the discharge of chlorinated sewage as presented by Coulter appears to be harmful to fish. Now while we do not know what the harm to persons is, no one argues that a benefit to people is associated with such a practice. Further we will soon hear that disinfection of Wastewater Treatment Plant effluents does not automatically require chlorination. Finally when the economic value of protecting the fish, or the accepted public health risk to the people, dictates a change in current practices, these practices will be changed. Until then we will continue to accommodate to the maximum extent possible the desires of the Coulters and the Ongerths on a case by case basis.

TECHNOLOGY CONSIDERATIONS

Now we can't really get Jim Coulter out of his dilemma unless we have useable alternatives to existing disinfection practices. Also economics and improved efficiencies in current practices are always welcomed by operators of Wastewater Treatment Plants. With this in mind chlorination, ultraviolet, and ozone disinfection practices were discussed.

Chlorination

Better mixing means better disinfection. Dr. Longley presented a paper which showed that rapid mixing of chlorine with the wastewater stream initially provides increased contact between the bacteria and virus with chlorine before the chlorine is dissipated in other reaction pathways. He studied several models claimed to be useful in disinfection process design. He concluded that the Prandt Eddy frequency and the mean velocity gradient have properties which make them useful for disinfection process design. He offered a pipe mixer and a venturi mixer as advances in technology for this purpose.

A simple, inexpensive, but effective modification to existing chlorination contact chambers was presented by Fred Hart. Baffling to produce plug flow and eliminate short circuiting was illustrated. The result was a 9 to 15 percent savings in chlorine usage.

George White reported on problems of disinfecting nitrified effluents at the San Jose/Santa Clara Water Pollution Control Plant. It was noted that the nitrified effluent created an exceptionally high chlorine demand. The application of amonia nitrogen reduced the chlorine demand and still enabled the plant to meet its NPDES Permit requirement of 2.2/100mL-MPN Total Coliform in the effluent. The surprising factor in this investigation was that the combined chlorine residual was found to be much more reliable in its germicidal efficiency than a free chlorine residual.

Ultraviolet Disinfection

Many of us were hoping, even expecting, that discussions on ultraviolet units would offer an immediate alternative to chlorination as a disinfectant. While this may be true under limited circumstances there would appear to be considerable work and study required before UV becomes fully competitive with chlorination.

Mr. Nehm presented results of pilot plant studies of 4 manufacturers' UV units. The test indicated that all units were capable of achieving the effluent fecal coliform goal of 200/100 mL. However, the useful in-service time of all units was limited by the formation of scale on the tubes. While this experience is limited to one quality water, this has always been a problem with UV disinfection, even with relatively cleaner drinking water. Until a solution is found that extends the useful life of the tubes to an acceptable period of time, it would seem that costs associated with operation and maintenance of the units will limit their application in wastewater treatment.

Donald Johnson looked at UV disinfection of filtered and unfiltered effluents under thick and thin film conditions. Given the proper dose (intensity x time) the UV units tested produced a 200/100mL fecal coliform count from both filtered and unfiltered effluents. I don't think anyone questions the ability of UV to disinfect. The question is for how long and at what cost.

Perhaps the work now getting underway at the Port Richmond Plant in New York will shed more light and provide more answers to these operation and maintenance problems. Karl Scheible says they are in phase one of an 18 month study. Their protocol provides for study of water quality, system geometry, system hydraulics, and equipment specification.

Ozone

Discussion of ozone technology essentially concerned analysis and control methodology. It is believed by some that analytical methods for determining residual ozone require some attention. Gilbert Gordon's paper suggests that electronic monitoring is simply not satisfactory and in wet analysis the stability of some indicator solutions is a problem. Indigo and arsenic were determined to be the most reliable. The effects of ozone decomposition and the pH of the effluent also were discussed.

Control of the ozone disinfection process by monitoring of exhaust gas was presented by Al Venosa. This approach is said to measure true ozone, require only one measurement, capable of being automated, accurate, sensitive, and stable. It requires a constant gas to liquid flow ratio. When this is done the dose can be controlled by monitoring gas in the exhaust and automatically signaling changes in the power to the generator.

Enos Stover told us how to optimize operational control of ozone disinfection. This should be considered in the design stages of the facility. The disinfection process must be effective, reliable, economic, safe, and require

minimal power, and maintenance. He noted that maximum dose applied may not permit maximum ozone transfer and lowest power consumption. He offers 2 ways to optimize your situation.

After investigating ozone disinfection and transfer in wastewater, Patrick Given shared results of a Canadian experience with us. The study involved screened, dilute wastewater. The results indicated not only a reduction of 99.9 percent in indicator organisms but a substantial wastewater strength reduction produced high dissolved oxygen levels in the effluent. Wastewater strength was an important factor in these studies.

Ed Opatken offered to convince us that it is possible to calculate ozone mass transfer coefficients. Then he told us there is no such thing (it varies with gas flow), and also that mass balance is enhanced by secondary effluent when compared with tap water. He says that mass transfer coefficients for ozone that were obtained from these studies can be used for the design and scale-up of ozone bubble diffuser contactors of plants where the primary source of the wastewater is of domestic origin.

Peter Foller presented a future new process for generation of ozone. It promises reduced capital cost (reductions of 50 percent) and easier scale up and scale down of equipment. Use is approximately 2 year away for a 10 pound per day level of ozone use.

The last session, just completed, related some practical considerations in the use of halogens, disinfectants, and in the design, operation, and maintenance associated with ozone and ultraviolet disinfection. Charles Haas reminded us that problems still exist in the design of disinfection systems using halogens. Some of these problems are associated with dose estimation, contactor hydraulics, process control, chemical supply, and safety.

Kerwin Rakness was concerned with the application of the correct ozone dose and the reliable and economical production of ozone. The significance and sensitivity of pretreatment of air was emphasized. Use of air in the ozone generator with a dew point of less than -55°C is considered optimum. In some situations manual control of ozone application may be preferable to use of automated controls.

Randy Junkins reported on a study just getting underway that will evaluate and document the effects of operation and maintenance practices on the performance of ozone and UV systems. It is much too early to draw any conclusions from the effort, either as related to efficiency, reliability or costs.

Karl Scheible says that disinfection of treated wastewater by ultraviolet irradiation has emerged as an accepted, feasible, cost-effective alternative. In my opinion this statement is open to considerable question. Regardless, initial steps to produce a UV design manual, sponsored by the EPA, are underway. It is proposed to feature the latest developments in the state of the art as they are recognized and practiced today.

CONCLUSION

During the course of this symposium a great deal of information has been presented and we now should be convinced that chlorination, ultraviolet, and ozone processes can be used under varying degrees of difficulty to disinfect wastewater effluents. Unfortunately no information has been supplied as to the relative economics associated with the capital cost and operation and maintenance cost for these processes. Until this is done Jim Coulter is hard put to press his desire to eliminate chlorination as the primary disinfectant for wastewater effluents.

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