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Environmental Protection Technology Series

**SYMPOSIUM ON FUGITIVE EMISSIONS
MEASUREMENT AND CONTROL
(May 1976, Hartford, CT)**



**Industrial Environmental Research Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
Research Triangle Park, North Carolina 27711**

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(May 1976, Hartford, CT)

E. M. Helming, Compiler

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Washington, DC 20460

FOREWORD

These proceedings for the symposium on "Fugitive Emissions: Measurement and Control" constitute the final report submitted to the Industrial Environmental Research Laboratory for Task Three of Environmental Protection Agency Contract Number 68-02-2110. The symposium was held at the Sheraton-Hartford Hotel, Hartford, Connecticut, May 17-19, 1976.

The objective of the symposium was to support the Environmental Protection Agency's efforts to develop methods for the measurement and control of industrial fugitive emissions. Papers were presented that described the scope and regulatory aspects of fugitive emissions in general, reviewed the impact of fugitive dusts on the atmosphere and indicated the need for future measurement and control programs. Recently developed measurement methods for air borne fugitive emissions and the results of measurement programs utilizing the methods in specific industrial applications were described. Existing fugitive emissions control technologies in typical industries were reviewed.

Dr. Robert M. Statnick of the Industrial Environmental Research Laboratory, Environmental Protection Agency, Research Triangle Park, North Carolina, was the Project Officer and General Chairman of the symposium.

Elizabeth M. Helming, Project Scientist at TRC - The Research Corporation of New England, Wethersfield, Connecticut, was the Symposium Coordinator and Compiler of the proceedings.

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Session I:
INTRODUCTION

Fugitive Emissions Problems in Perspective*

by J. E. Yocom

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ABSTRACT

Primary emphasis on control of air and water pollutants has been placed on point sources. Fugitive emissions, or those emitted from non-point sources, are important contributors to environmental degradation in many areas and in relation to many types of industrial operations. Because of the potentially high cost of controlling such emissions it is important that their significance be accurately assessed.

This paper discusses many types of fugitive emissions and the methods for assessing them. Fugitive emissions are extremely site specific in respect to their measurement and control, and this paper presents examples of measurement programs that put fugitive emissions in proper perspective in relation to other source categories and other environmental impacts.

*For Presentation at a Symposium on "Fugitive Emissions - Measurement and Control". Sponsored by the Environmental Protection Agency (IERL-RTP), Sheraton-Hartford Hotel, Hartford, Conn., May 17-19, 1976.

1.0 INTRODUCTION

The pollution control programs currently being developed and implemented in this country have, for the most part, been directed at emission sources that release pollutants into the environment from well defined points. These emission points are represented by stacks and ducts emitting air pollutants; and pipes, culverts, and channels discharging water pollutants. Until recently little attention has been given to fugitive emissions, or emissions that enter the environment through other, ill-defined routes.

This brings up the question of terminology. It is easier to say what fugitive emissions are not than what they are. The term "fugitive" is not descriptive. It implies pollutants that have escaped and must be captured, brought to justice, and incarcerated. But in considering fugitive air pollutants that have already escaped from a roof monitor or an outdoor material handling pile or fugitive water pollutants that have already entered a natural stream or the ground water, it is out of the question technologically and economically to attempt capturing and controlling them at this point in their escape route. There are countless types of fugitive emissions and all of us could provide many examples related to type of pollutant, mechanism of release, and behavior in the environment. When we consider this class of emissions we immediately recognize that each is extremely source and site specific and the assessment and control of such emissions must recognize these fundamental factors. While I do not advocate a change in the title of this conference, nor will I stop using the term "fugitive emission", I believe that we could come up with a more descriptive term for the type of pollutant emission we will be discussing. At the moment I would vote for "non-point source emissions", a term currently used by the water pollution control people.

The strategies for control efforts on point sources have been based upon

a logical progression starting with quantifying the deleterious effects of pollutants on human health and ecological systems, followed by prioritization of sources to be controlled based upon the quantities of important pollutants released. The programs for point source control are well advanced, largely because such releases are already or have traditionally been released from well defined conduits and uncontrolled emissions from these points often represent the largest potential emission from a process. Furthermore, such emissions can be accurately measured using well established techniques.

In spite of this emphasis on point source control, EPA and the industrial community have long recognized that fugitive emissions from certain types of processes can be important contributors to environmental degradation. Many obvious fugitive sources have already received considerable attention and control efforts have been implemented or initiated; for example, fugitive air emissions from coke ovens, and non-point source emissions from abandoned coal mines in the form of acid mine drainage. As point sources are controlled more stringently, fugitive emissions (even those that are not now obvious) will become increasingly important in determining environmental quality. The principal deterrents to the control of fugitive emissions have been essentially in two areas:

1. Difficulties in measuring fugitive emission rates and thereby assessing their impact on the environment.
2. The generally high costs of controlling such emissions by consolidating them through process modification or capturing them for removal in emission control systems.

2.0 IMPORTANCE OF FUGITIVE EMISSIONS

How important are fugitive emissions? Those living adjacent to an active coal storage area on a windy day might tell you that the living conditions are intolerable. Ecologists concerned about eutrophication of small lakes in rural

areas will tell you that fugitive or non-point sources of nutrients are the culprits. On the other hand, an industrialist faced with controlling fine particulate emissions leaking from furnace buildings might tend to look at these fugitive emissions as only an aesthetic problem. This is perhaps understandable since he might have already spent several million dollars in enclosing furnaces and installing air pollution control systems and the leakage, which he considers unavoidable based upon present control technology, is thought to be a small part of the problem.

In view of the tendency for many fugitive emissions to take on an element of emotionalism there is a need to develop some perspective about their relative importance. First of all, it is difficult to generalize. As stated earlier, each fugitive emission problem is different and tends to be quite site specific. One aspect that makes air fugitive emissions especially important is the tendency for such emissions to be emitted over a broad frontal area and at ground level. This means that fence line effects tend to be significant over much of the plant's perimeter. Furthermore, except for large particles which settle out rapidly as they move away from the source, concentrations of fugitive emissions do not fall as rapidly with distance as do emissions from tall stacks. In general, ground level concentrations of pollutants from tall stacks decrease with the square of the distance from the source since such emissions are able to disperse both vertically and laterally. On the other hand, ground level concentrations of pollutants emitted from low-level fugitive sources covering a broad frontal area tend to be related to the first power of distance from the source. This results from the inability of the plume to disperse downward and the lack of significant dilution laterally because of the breadth of the plume.

In short, fugitive emissions as a class are important but their relative importance in specific situations depends upon a variety of factors.

3.0 SOURCES OF FUGITIVE EMISSIONS

While fugitive emissions are site specific, there are certain types of situations that provide maximum potential for their release.

1. Open Operations

Such operations as mining, quarrying, outdoor materials handling, and coke making produce fugitive air emissions containing both particulates and gaseous pollutants. Fugitive water emissions also emanate from such operations, for example, acid mine drainage from coal mining, and surface runoff from all such outdoor operations.

2. Leaks and Spills

Leaky piping, pump and valve glands, and loss of material from spills are important sources of fugitive air and water emissions. Gases and high vapor pressure liquids that leak to the atmosphere are sources of air fugitive emissions. Liquids leaking to the ground are capable of finding their way into surface and ground waters.

3. Storage and Disposal of Materials and Wastes

Material storage piles (both worked and quiescent) and dried sludge beds are sources of atmospheric particulate matter. Sludge disposal beds are sources of contaminated runoff and can also degrade ground water. Disposal ponds containing volatile materials are sources of fugitive air emissions.

4. Incompletely Controlled Point Sources

While a control device may adequately control the basic process emissions from a source, certain operating conditions, which may be planned or unplanned, create emissions that the control system cannot adequately handle. In the case of air pollutants an example would be an electric steel furnace whose basic emissions are controlled by hooding or furnace evacuation followed by a high efficiency bag house, but during charging and tapping the fumes are not captured and they escape through the roof monitor. An example in the water area would be an aeration or stabilization lagoon that releases fugitive water pollutants during heavy rains when the capacity of the system is exceeded.

5. Poor Housekeeping

Accumulations of materials that are likely to be carried into the environment by the winds or rain water are important sources of fugitive emissions. In rendering plants the accumulation of putrescible materials is a significant source of fugitive odors.

In some industrial processes examples can be found for many of these categories of sources. Figure 1 shows a simplified schematic diagram of a typical iron foundry. Even this relatively simple operation has many sources of fugitive air emissions. As part of an EPA study, TRC identified a group of industrial processes that have significant potential for fugitive air emissions. These are shown in Table 1.

4.0 METHODS OF MEASUREMENT

A very simple method for categorizing fugitive emissions is by means of the feasible methods of measuring them. In our work for EPA we have evolved three basic approaches to the measurement of atmospheric fugitive emissions. These will be discussed in greater detail by Hank Kolnsberg.

4.1 Quasi-Stack

In this method the fugitive emission is temporarily hooded or encapsulated and a temporary duct or stack and fan is installed on the duct to permit sampling by means of standard stack or duct sampling methods.

As an illustration of how the quasi-stack method is used to assess fugitive emissions, we present Figure 2 from a TRC study. This test configuration was set up in a ship building establishment to assess welding emissions. There were three basic objectives of the study:

1. Determine indoor concentrations of important particulate and gaseous releases.
2. Utilize the data to design a ventilation system.
3. Assess impact of fugitive and process emissions on the surroundings.

Emission factors for each of 10 welding configurations were developed in terms of weight rate of pollutant (particulates, metals, pollutant gases) per pound of a welding rod. The reproducibility of results from duplicate samples was excellent and since the plant maintained accurate records on amount and

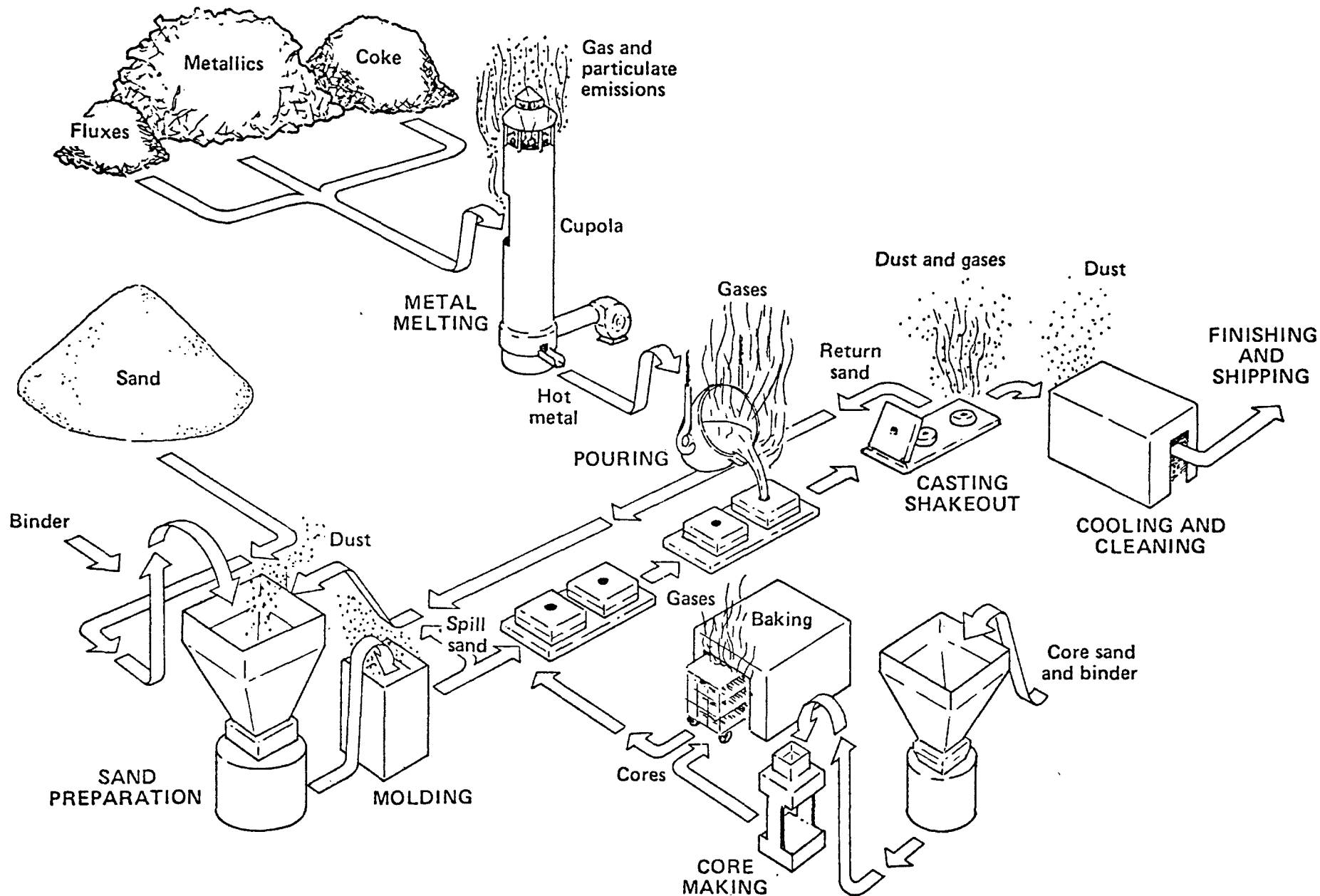


Figure 1. Iron foundry process flow; sources of emissions.

Table I. Classification of Industries With High Potential for Fugitive Emissions

<u>Category</u>	<u>Industry/Process</u>	<u>Major Fugitive Emissions</u>
Metallurgical	Primary Aluminum	Fume, Fluorides, PNA*
	Primary Copper	Fume, SO ₂ , Dust
	Steel Making	Fume, Kish, CO, Odors
	Iron & Steel Foundries	Fume, Odors
	Coke Making	Smoke & Fume, Hydrocarbon Gases & Vapors, Odors, CO, PNA
Energy/Fuels	Coal Mining & Processing	Dust
	Coal Gasification	Hydrocarbons, Smoke & Fumes, CO, PNA
	Char-Oil-Gas	Hydrocarbon Gases & Vapors, CO, PNA
	Shale Oil	Fumes & Dust, Hydrocarbon Gases & Vapors, CO, PNA
	Petroleum Refining	Hydrocarbon Gases & Vapors, Odors, PNA
	Oil Production	Hydrocarbon Gases & Vapors, Odors
Chemical Products	Plastics	Hydrocarbon Gases & Vapors, Odors
	Tire & Rubber	Hydrocarbon Gases & Vapors, Odors
Rock Products	Phosphate Fertilizer	Dust, Fluorides, SO ₂
	Lime	Dust
	Sand & Gravel	Dust
	Asphalt Batching	Dust, Odors, PNA
Other	Agricultural Operations	Dust

*PNA - Polynuclear Aromatics

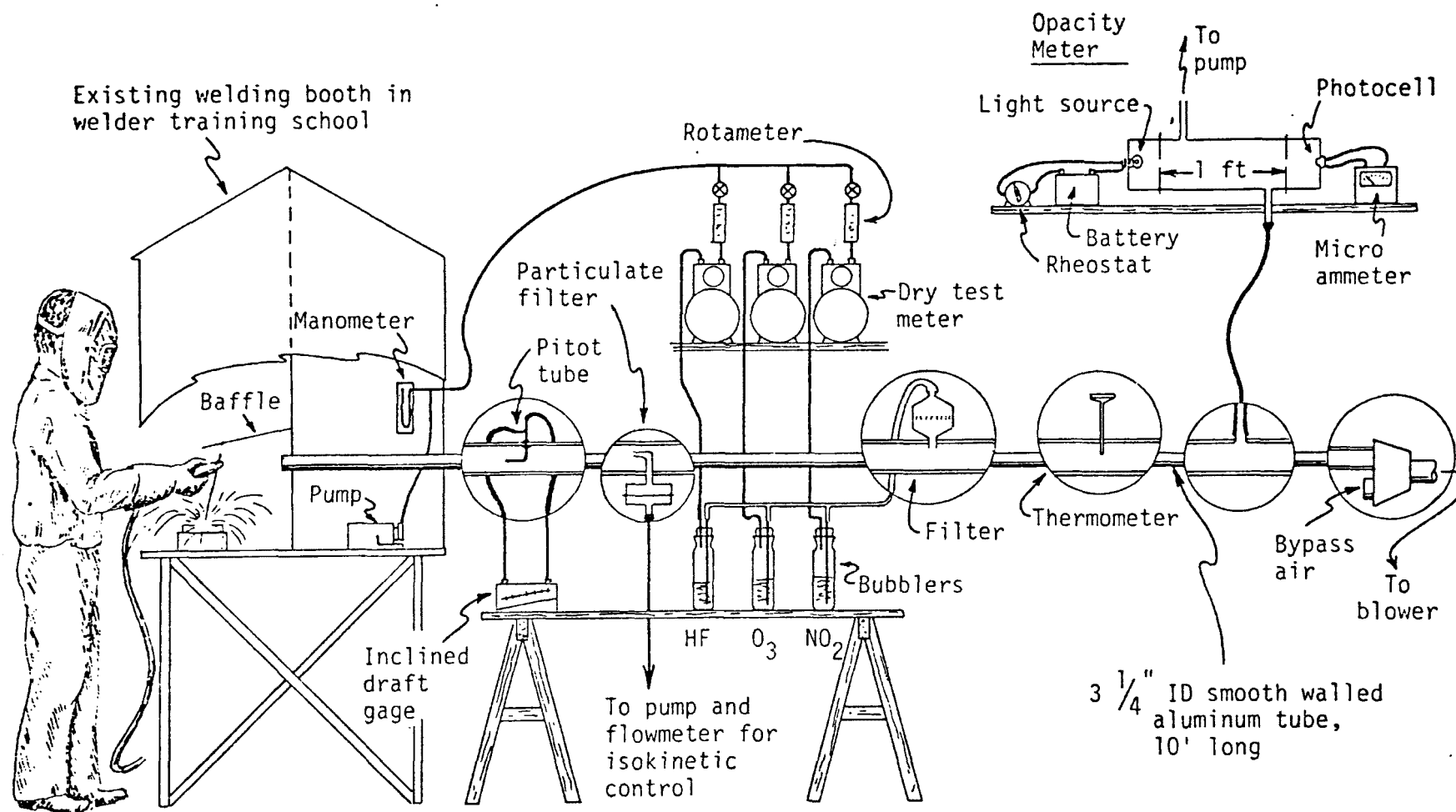


Figure 2. Schematic diagram of test configuration for field measurements of welding emissions.

type of welding rod used, we were able to provide an accurate estimate (probably $\pm 25\%$) of plant emissions. Furthermore, we were able to assess the impact of changes in the quantity and mix of welding types. This approach, using the quasi-stack method and computing emissions, was considered to be far more accurate and cost-effective than sampling in the building openings. By application of this method, we were able to place the impact of fugitive emissions on the outdoor atmosphere in perspective as compared with the effect of the emissions on the indoor environment.

Another advantage of the quasi-stack method is that it often can provide data on the design of a control system, since sources of this type are most often controlled by installing hoods, ducts, control systems and fans.

4.2 Roof Monitor

The term "roof monitor" denotes the generalized situation where there are open sources inside of a building and the fugitive emissions are to be measured at building openings, including the roof monitor, windows and doors, and ventilation openings. This method is applied when sources inside the building are too numerous, too large, or too inaccessible to permit the use of the quasi-stack method. This method tends to be less accurate than the quasi-stack method since it is necessary to produce an accurate air and material balance on all building openings. Since we will be hearing about an example of a roof monitor measuring program and since Hank Kolnsberg will be giving further details on our methods development program, I will not discuss this method further.

4.3 Upwind-Downwind

This term describes the general approach of sampling a fugitive emission in the free atmosphere. The most common application is that of sampling simultaneously upwind and downwind of sources of fugitive emissions and using the difference in sampling values as an indication of source contribution. This

generalized method would also encompass sampling directly in free plumes.

We will hear more about this aspect from Bob Jacko.

The application of the upwind-downwind method is not straightforward and carrying out such measurements and interpreting the data must be done with great care. In general, a pair of simple measurements at ground level upwind and downwind of a fugitive source will indicate at best only that there is a source between the sampling sites. It will not provide even an estimate of emission levels.

The sophisticated route is to utilize three-dimensional sampling and wind flux arrays upwind and downwind of the source. A good cost-effective compromise using ground level monitors can be used but it requires supplementary data gathering efforts including

1. Accurate emission inventories for point sources.
2. Detailed knowledge of the chemical and physical properties of point and fugitive emissions and the collected pollutants.
3. Mathematical models that take into consideration the decay, deposition or reaction of emitted pollutants.
4. Tracer studies to calibrate the models.

Table II shows the results of a study that pieces together from the above elements an assessment of the relative contribution of fugitive and point source emissions of a mineral based operation in relation to background particulate levels. Such an analysis puts the problem of plant fugitive emissions in perspective, and, in this case, the results of the study showed that control of fugitive emissions was a more cost-effective method of air quality control than further control of point sources.

Table II. Distribution of Particulate Matter from Process Emissions
and Fugitive Dust at Several Downwind Sampling Points

Distance from Edge of Plant Property, mi	0	0.8	1.4	4.8
Process Contribution ¹ $\mu\text{g}/\text{m}^3$	126 (37%)	26 (18%)	12 (10%)	1.6 (2%)
Plant Fugitive Dust ² $\mu\text{g}/\text{m}^3$	170 (49%)	40 (27%)	23 (20%)	2.4 (3%)
Background, $\mu\text{g}/\text{m}^3$	49 ³ (14%)	80 ⁴ (55%)	80 ⁴ (70%)	70 (95%)
TOTAL $\mu\text{g}/\text{m}$	345	146	115	74

¹Based on source inventory and diffusion modelling.

²Calculated by difference.

³Based on upwind-downwind sampling at plant.

⁴Based on data for winds other than from direction of plant.

⁵Air quality data for downwind periods.

5.0 CONCLUSIONS

Fugitive emissions are a complex and challenging category of sources that have important impact on the environment. Their nature and relative importance are exceedingly site specific and reliable methods of measuring such emissions are the key to putting their relative importance into perspective. Control measures are closely related to the processes generating the fugitive emissions and are likewise site specific. Since fugitive emission control can be costly, there is a great need for measurements for assessing the importance of the emissions and the effectiveness of control systems.

REGULATORY ASPECTS OF FUGITIVE EMISSIONS

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May 17, 1976

In a June 1974 letter to Oregon's Department of Environmental Quality concerning emission standards, the Asphalt Pavement Association of Oregon concluded with this statement: "If God wanted perfectly clean air he would have made the earth out of asphalt not dirt and he would not have created fire."

There are a few problems with this concept, but the writer does have a point. Where there's fire, there's smoke; and where there's dirt, there's dust. Windblown dust can comprise as much as 90 percent of total ambient particulate concentrations, ⁽¹⁾ agricultural open burning is by far the largest stationary source of both particulate and hydrocarbon emissions out of 200 source categories evaluated in a recent study, ⁽²⁾ and industrial fugitive emissions may in many cases exceed controlled stack emissions from the process. What, then, are fugitive emissions, how much of a problem do they represent, and what are EPA and other agencies doing to regulate them?

To begin with, it is necessary to define what is meant by the term fugitive emissions. Probably the most widely used definition is that they consist of air pollution emissions which have not passed through a stack or duct. The distinction is a fine one, since SO₂ from a culm pile is considered fugitive, while SO₂ passing uncontrolled through an electrostatic precipitator is not. Lillis

and Young⁽¹⁾ define two separate problems: industrial fugitive emissions (gaseous and particulate emissions that result from industrial related operations such as metallurgical furnaces and materials transfer and storage and which escape to the atmosphere through windows, doors vents, etc.) and fugitive dust emissions (dust storms and windblown particulate from unpaved roads, tilled land, etc.). In discussing regulatory aspects, it is convenient to further define fugitive emissions on the basis of controllability, as:

1. Confined - fugitive emissions which were initially contained in a duct, hood, building, or other facility, but which escape to the atmosphere without passing through a control device or stack, and

2. Confinable - fugitive emissions from storage piles, open conveyor belts, material transfer operations and other sources, such as coke ovens, which escape directly to the atmosphere, but for which confinement or other control measures are possible.

Fugitive dust emissions constitute a third category, unconfined, since they originate from large tracts of tilled farm land, dust storms, active volcanos, and other sources for which control measures are usually unavailable, unjustifiable, or too costly. There is some overlap in these categories. Paving dirt roads, for example, may be feasible in one area and unjustifiable in another, so unpaved roads may or may not be considered "unconfined." In the context of this paper, the term fugitive emissions has the same definition used by Lillis and Young and refers to the first two of the three controllability categories:

confined and confinable emissions.

The Problem

Fugitive emissions constitute an increasingly important problem in air pollution control strategy. Limited but persuasive data implicate fugitive emissions as a major proportion of total emissions, especially where stack emissions are reasonably well controlled. Fugitive emissions of particulate during electric arc furnace charging, for example, are estimated to be from five to fifty times well-controlled stack emissions from the furnace on a pounds per ton basis, and tests at a lead smelter measured three times as much lead per hour emitted from fugitive sources as from the controlled stack emissions.⁽³⁾

Preliminary estimates indicate that 128 of the 247 Air Quality Control Regions (AQCR's), 52 percent, are not expected to attain the total suspended particulate (TSP) national ambient air quality standard (NAAQS) in 1976.⁽⁴⁾ Additional control of stack emissions will be adequate for some, but not all, AQCR's. Assume that fugitive emissions from a process equal two percent of total uncontrolled emissions from that operation; if the captured emissions are treated with a 98 percent efficient control device, then the amount of fugitive emissions equals stack emissions. Also, the fugitive emissions would likely have a greater local effect on ambient air pollutant concentrations since they generally are emitted nearer ground level with less vertical velocity than are stack emissions. Ambient air measurements at a coke oven plant, for example, indicate that emissions from this facility increase downwind TSP levels by 200 $\mu\text{g}/\text{m}^3$ half a

mile from the facility and by $100 \mu\text{g}/\text{m}^3$ a mile away.⁽⁸⁾

The impact of fugitive emissions on air quality over a large area has recently been estimated. Over 30 percent of all industrialized urban monitoring sites analyzed in a recent study⁽⁵⁾ are influenced by particulate fugitive emissions; they average on an annual basis $25 \mu\text{g}/\text{m}^3$ TSP higher than the other industrial sites. Fugitive emissions in heavily industrialized cities are tentatively estimated to increase annual citywide ambient TSP levels by roughly $10 \mu\text{g}/\text{m}^3$. These results are from 20 sites in five cities.

Many sources of hydrocarbon emissions are essentially fugitive emission sources. The magnitude of these emissions is difficult to determine, but the Office of Air Quality Planning and Standards (OAQPS) has a task force studying the problem and establishing model regulations. Oxidants, for which hydrocarbons are precursors, are the second most serious nonattainment problem (Table 1).

Regulation Under SIP

During the development of SIPs, states quantified emissions from all sources using the best information available to them. Few fugitive emission sources were specified in these plans, primarily due to the magnitude of stack emissions from poorly controlled sources and the lack of even rough emission estimates for most fugitive emission sources. Even such general regulations as process weight curves are usually applied only to stacks, not total emissions. This probably is a direct reflection of the extreme costs, difficulties, and uncertainties involved in testing most fugitive emission sources. No widely accepted test methods exist, and testing wasn't even being attempted on most of these sources until very recently.

Many agencies did, however, utilize general regulations to address the fugitive emissions problem. An analysis of all 50 state and 26 major local agency regulations showed that 63 percent require that reasonable precautions be taken to prevent fugitive emissions; 5 percent have nuisance provisions; 14 percent limit property line concentration, particle size, or fallout; 11 percent regulate specific sources; and 7 percent have no fugitive emission regulations.⁽⁵⁾ Compliance is a matter of following equipment specifications and approved operating procedures, meeting a visible emission limitation, or meeting an ambient air property line fall-out or concentration requirement (Table 2).

A year-long revision of SIPs, in areas where a need for revisions is indicated, begins July 1976. The initial SIPs, as Figure 1 indicates, when fully implemented will do a remarkable job of reducing process emissions. Particulate emissions shown are those projected for 1985. The top line represents uncontrolled emissions; the next delineates emissions with present (1975) SIP requirements; the third represents a combination of 1975 SIPs and best control of all post-1975 sources; the final line is a lower limit representing zero emissions from all post-1975 capacity. This 1985 profile shows that SIPs provide the major impact on particulate process emissions (including combustion). Many of the SIP revisions in non-attainment areas, therefore, are expected to concern fugitive emissions.

Several EPA projects are underway to assist states in this effort. One is a report on the best estimates of both fugitive and stack emissions from all iron and steel mill operations, including windblown dust from storage piles. These factors are being formulated by a joint EPA-AISI

task force formed in January 1976 to determine whether fugitive emissions are, as has been claimed by industry, essentially "cosmetic." Finalized factors are expected in mid-1976, but longer-range task force projects include acceptable fugitive emission test methods, tests of specific fugitive emission sources (at least partially through existing EPA-ORD contract studies), and dispersion modeling a medium size well-controlled integrated steel mill to determine ambient air impact.

Guidance on other sources is expected in an EPA report due January 1977 which will summarize all existing fugitive emission information for the states' use. A similar study on fugitive dust is expected in July 1976. The reports will include model regulations. The present control strategy recommended by EPA is an equipment standard which uses visible emissions as an enforcement tool.⁽¹⁾

Regulation Under NSPS and NESHAPS

New Source Performance Standards (NSPS) are applicable to new or modified stationary sources under Section 111 of the Clean Air Act (CAA), and have been directed principally at stack emissions. Figure 2, which is drawn by expanding the Figure 1 ordinate, permits an evaluation of the impact of NSPS on total 1985 particulate emissions. As Figure 2 shows, the impact - with the exception of the power plant standard - is hardly noticeable in terms of national emissions. This is at least partially due to the stringent SIPs, which lessen the impact of an NSPS representing best control.

For other criteria pollutants, NSPS impact is much more dramatic, and present policy emphasizes a need for hydrocarbon and nitrogen oxide NSPS, a decision supported by growing concern over widespread non-attainment

of the oxidant NAAQS, an anticipated rapid growth in nitrogen oxide emissions, and a growing realization that both of these pollutants represent difficult control technology/enforcement problems.

Increasing interest in and information on fugitive emissions has refocused attention on particulate sources and will affect NSPS priorities. It will also intensify efforts to establish NSPS for total source emissions, both stack and fugitive. Promulgated NSPS have usually included at least a visible emission limitation applicable to both fugitive and stack emissions. A few have actually established mass limitations, such as the fluoride emission NSPS for primary aluminum production, which applies to both stack and roof monitor emissions and specifies test methods for both emission points. One serious limitation to NSPS for fugitive emissions is the lack of specific authority to promulgate equipment standards. In certain cases, such standards would be by far the most efficacious from the viewpoint of both industry and control agencies.

Addressing fugitive emissions more directly than do NSPS, national emission standards for hazardous air pollutants (NESHAPS) under Section 112 of the CAA require installation of control equipment with specific performance characteristics and/or require implementation of certain operating and maintenance practices. The asbestos NESHAPS, for example, specifies control equipment parameters and mandates how asbestos tailings piles are to be operated. The regulation also requires fencing (or natural barriers) and even describes warning signs to be posted. The mercury and beryllium NESHAPS utilize similar approaches.

Enforcement

EPA's Division of Stationary Source Enforcement (DSSE) has identified fugitive emissions as a major compliance issue, and has initiated projects to quantify total emissions from such difficult-to-measure sources as coke oven battery coke-side leaks and pushing emissions⁽⁷⁾ and quench tower emissions.⁽⁸⁾ These and other studies planned or underway in EPA's Office of Research and Development and other groups will help quantify the seriousness of fugitive emissions from many sources. Fugitive emission sources in the 128 AQCRs where ambient TSP standards are not being attained are of special concern to DSSE. Labeling these emissions as too difficult to assess, insignificant, or cosmetic is insufficient; efforts to control fugitive emissions will, in nearly all cases, be expected.

Until much less elaborate and costly test methodology is available, opacity will remain the primary fugitive emission enforcement tool. Opacity tests (such as the NSPS Method 9) have been upheld in court litigation, give nearly instantaneous results, require only one or two qualified observers, and eliminate the need for test equipment and laboratory analysis. Use of visible emission standards is by far the most practical enforcement approach in terms of minimizing both control agency and industry testing resources.

It is surprising, then, that court action continues against SIP visible emission standards and that a recently proposed CAA amendment⁽⁹⁾ would preclude EPA enforcement of such SIP regulations. The alternative, for emission sources, is much worse: EPA under Section 114 of the CAA can, among other things, require an owner or operator to sample emissions

(as prescribed by EPA) and install, use, and maintain monitoring equipment to determine whether a source is in violation of Section 110, 111, or 112 standards (or even to aid in developing such standards). Such requirements have to date been minimal only because visible emission regulations provided an equivalent, less costly, alternative.

Summary

In conclusion, EPA's regulatory offices are becoming increasingly concerned about fugitive emissions and their impact on ambient air pollutant concentrations. Quantification and regulation of these emissions will likely constitute a major EPA effort during the next five years. The most practical approach appears to consist of equipment standards enforced by visual observation; present sampling technology is too difficult and costly as long as opacity regulations remain a viable option.

TABLE 1. PRELIMINARY ASSESSMENT OF NUMBER AND PERCENT OF
AQCR's WHICH MAY NOT ATTAIN NAAQS IN 1976⁽⁴⁾

<u>Pollutant</u>	<u>Number</u>	<u>Percent</u>
TSP	128	52
SO ₂	27	11
CO	48	19
O _x	65	26
NO _x	13	5

FIGURE 1. Projected 1985 Particulate Emissions Under Various Control Strategies (2,6)

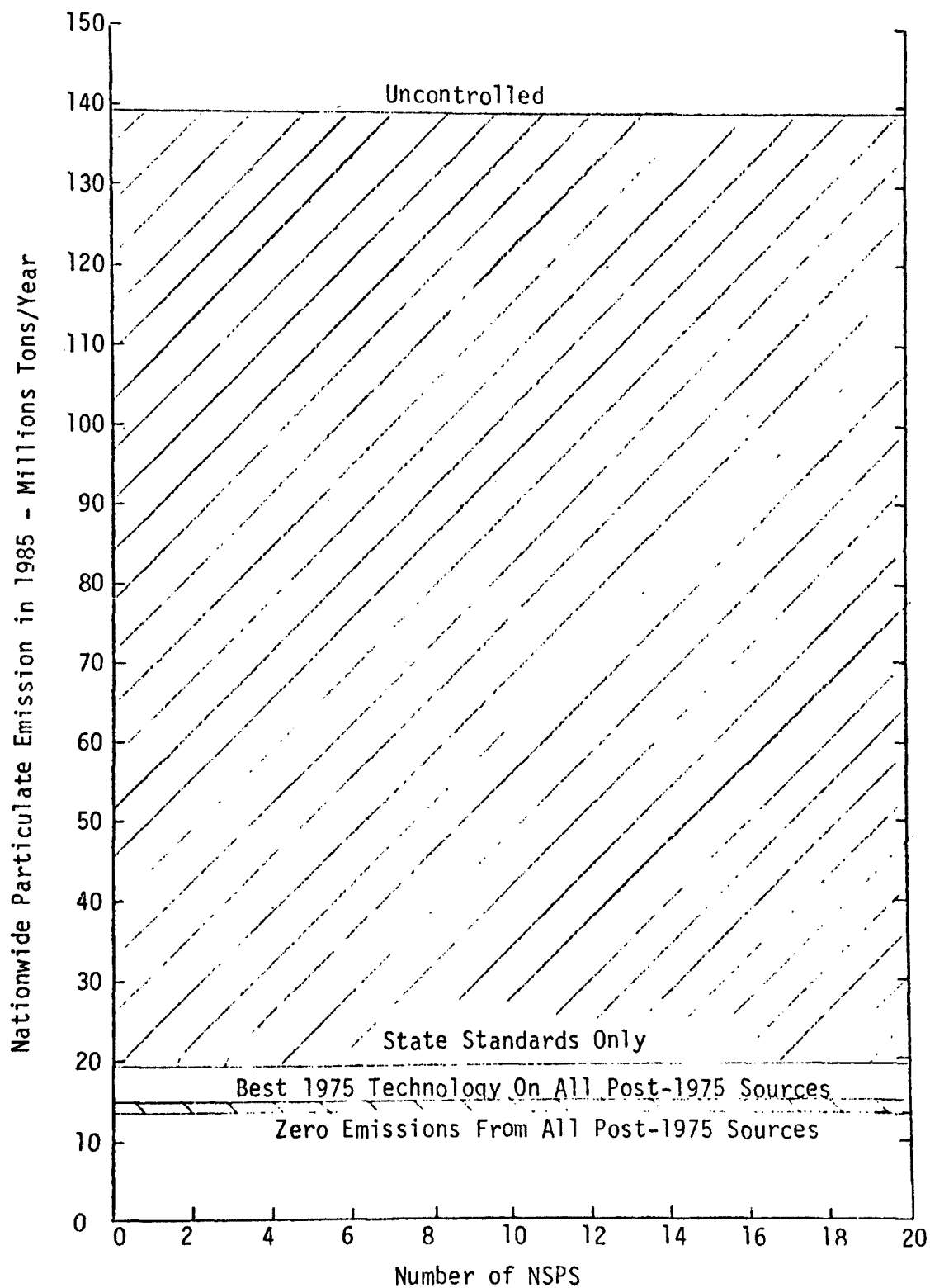


FIGURE 2. Estimated Impact of NSPS Over a Ten-Year Period
on 1985 National Particulate Process Emissions

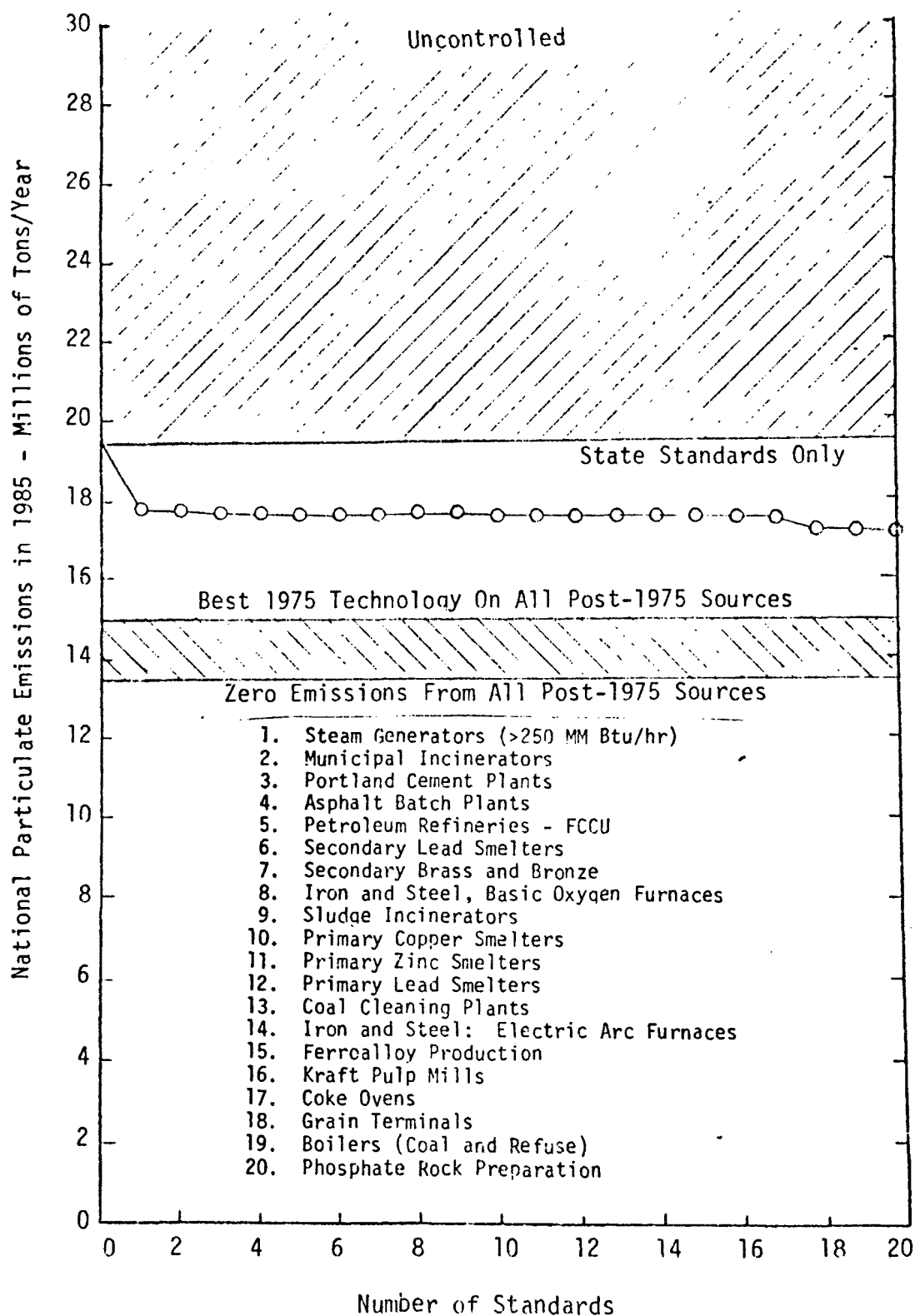


TABLE 2. PROPERTY LINE STANDARDS⁽⁵⁾

Jurisdiction	Standards
Hawaii	150 $\mu\text{g}/\text{m}^3$, above upwind concentration, 12-hour average 3.0 g/m^2 , fallout above upwind concentration, 14 day period
Kansas	2.0 mg/m^3 , above background concentration, 60 minute average
Texas	100 $\mu\text{g}/\text{m}^3$, 5-hour average 200 $\mu\text{g}/\text{m}^3$, 3-hour average 400 $\mu\text{g}/\text{m}^3$, 1-hour average
Omaha	500 $\mu\text{g}/\text{m}^3$, 60 minute average
Missouri	80 $\mu\text{g}/\text{m}^3$, 6 month geometric mean 200 $\mu\text{g}/\text{m}^3$, 2-hour arithmetic mean 0.4 coh/1000 linear feet soiling index, 6 month geometric mean 1.0 coh/1000 linear feet soiling index, 8-hour arithmetic mean >40 μm prohibited
Mississippi	5.25 g/m^2 , fallout above background
Nevada	2 tons/ mi^2 , 24-hour period
Illinois	>40 μm prohibited
Cleveland	500 $\mu\text{g}/\text{m}^2$, 60-minute average

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3. Fugitive emission results were from PEDCo's Silver Valley /Bunker Hill Smelter Environmental Investigation for EPA's Region X, (Febr. 1975). Stack test results were from a telephone conversation with Mark Hooper, Region X, EPA.
4. Conversation with Henry C. Thomas, Control Programs Development Division, Office of Air Quality Planning and Standards, Environmental Protection Agency, May 6, 1976.
5. From information supplied by Tom G. Pace, Control Programs Operations Branch, Control Programs Development Division, OAQPS, EPA. These data are from a study being conducted for EPA by GCA Technology Division: National Assessment of the Particulate Problem, Draft Final Report, April 1976.
6. "Priorities and Procedures for the Development of Standards of Performance for New Stationary Sources of Atmospheric Emissions," report prepared under contract by Argonne National Laboratory, EPA Contract No. EPA-IAG-D4-0463, Project No. 2, April 1975.
7. Telephone conversation with Louis Paley, DSSE, EPA, May 5, 1976.
8. Conversation with Carl Edlund, DSSE, EPA, February 3, 1976.
9. Amendment No. 1597, proposed by Senator Randolph to S.3219, April 13, 1976.

Session II:

MEASUREMENT

James A. Dorsey
Session Chairman

A GUIDELINE FOR THE MEASUREMENT OF AIR-BORNE
FUGITIVE EMISSIONS FROM INDUSTRIAL SOURCES

by

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ABSTRACT:

The paper presents a guide for the selection of the most effective program for the measurement of air-borne fugitive emissions from an industrial source.

The quasi-stack, roof monitor and upwind-downwind techniques presently utilized for sampling a wide variety of air-borne pollutants are described.

General criteria for the selection of the most effective sampling program, relative to characteristics of the site, process and emissions are discussed.

Baseline estimates of manpower, time and cost requirements for typical measurement programs for each technique are provided.

1.0 INTRODUCTION

The term "fugitive emissions" may be applied to any gaseous or particulate pollutant entering the ambient atmosphere without first passing through a stack, duct or other device designed to direct or control its flow.

Industrial fugitive emission sources are, in general, complex combinations of a process or operation and a physical location or arrangement, and are so varied as to make each source essentially unique. All fugitive emission sources share one common characteristic in that standard measurement techniques such as stack sampling are useless for determining the amount or nature of pollutant materials being transmitted into the surrounding atmosphere.

This paper describes the three methods recognized as effective measurement techniques for identifying and quantifying pollutants from almost any industrial source. The methods, identified by their basic sampling schemes, are the quasi-stack, roof monitor, and upwind-downwind techniques. General criteria for consideration in the selection of the most effective technique relative to characteristics of the process, its site, and its emissions are presented. Baseline estimates of manpower requirements and costs for the performance of typical measurement programs for each method are included.

2.0 MEASUREMENT TECHNIQUES

Measurements of air borne industrial fugitive emissions may be made at the source, before the pollutants begin to diffuse into the ambient air; in the air immediately surrounding the source, where the diffusion is limited to a relatively small volume of air; or in the ambient air,

where the diffusion is essentially complete. The respective measurement methods are the quasi-stack, roof monitor, and upwind-downwind sampling techniques. Each is described in general terms below.

2.1 Quasi-Stack Sampling Technique

This technique captures the emissions at their source in a temporarily installed hood and transmits them, by means of an exhaust blower, through a duct of regular cross-sectional area where standard sampling techniques are employed to measure the emission concentration and the flow rate of the emission carrying air stream. (A simplified sampling system is shown in Figure 2-1). The source strength of any pollutant or combination of pollutants may then be determined as the product of these two measured values.

The quasi-stack method is the most accurate of the air borne fugitive emissions sampling methods in that it captures virtually all of the emissions from a given source and, as a result of careful system design, conveys them to their measurement locations with a minimum of dilution by transport air. It is also the least applicable of the methods since its use must necessarily be restricted to those sources of emissions that can be physically and operationally isolated and are arranged to permit the installation of the capture and measurement system in a manner that will not interfere with normal plant operations or alter the character of the emissions or their generating process.

2.2 Roof Monitor Sampling Technique

This technique is used to sample the emissions from processes or operations taking place within buildings with a small number of openings

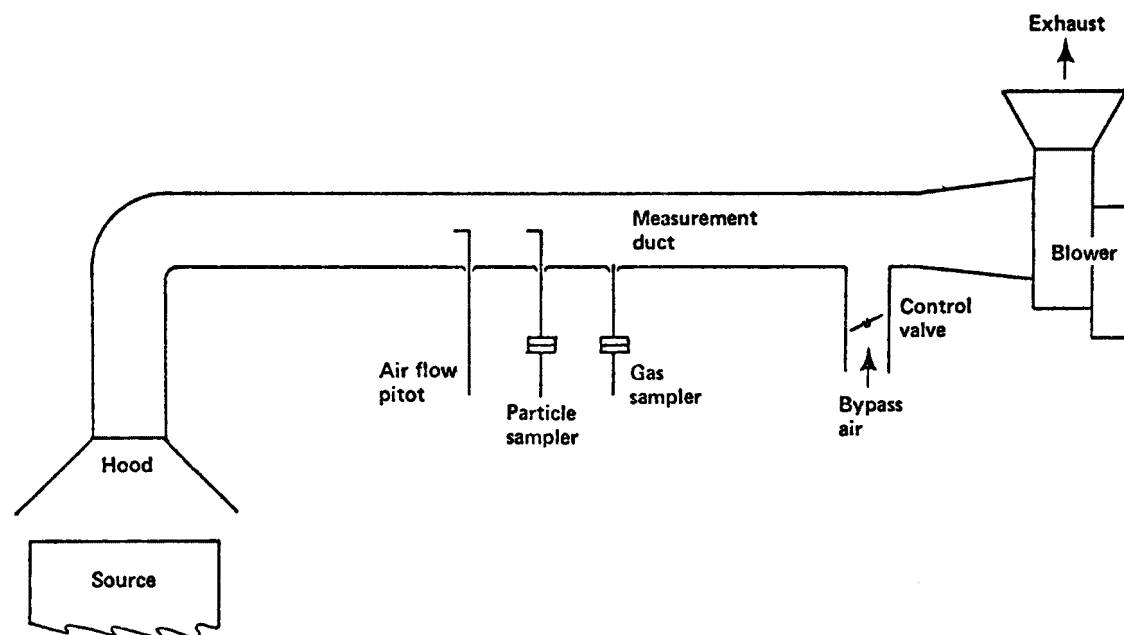


Figure 2-1: Simplified Quasi-Stack Sampling System

to the ambient atmosphere. Such a structure acts as a large hood, confining the emissions to a finite volume of air before transmitting them through one of its openings, such as a roof monitor, an exhaust fan, or a door or window, to the outside air.

Samples are taken at the opening to determine the concentration of the emissions in the transport air flowing to the ambient atmosphere, and the flow rate through the opening is measured. (A simplified sampling arrangement is shown in Figure 2-2). The combined source strengths of all the sources then producing emissions inside the enclosure is then determined as the product of the measured concentration and flow.

The roof monitor method is not as accurate as the quasi-stack method since a significant portion of the emissions may escape through other openings before reaching the measurement point and since a much higher degree of dilution with transport air occurs before measurement. It is a generally more applicable method in that it can usually be utilized to measure any indoor source, either for the composite of all processes within a building, or for a specific combination of sources where processes or operations may be selectively scheduled.

The roof monitor method requires instrumentation and trained personnel capable of making measurements of usually very low air velocities through a relatively large opening and mass balances of very small quantities of materials. It usually does not interfere with operations or schedules since the required equipment and measurement activities are set up and conducted away from the production floor.

2.3 Upwind-Downwind Sampling Technique

This technique is used to determine the emissions generation rate

Figure 2-2: Roof Monitor Sampling System

of sources that cannot be effectively hooded for the quasi-stack technique and are not enclosed in a structure permitting the application of the roof monitor technique. The emission concentration is determined in samples taken from the ambient air approaching (upwind) and leaving (downwind) the industrial site. The site contribution at the upwind location is calculated as the upwind concentration minus the downwind concentration. This contribution is then used in proved diffusion equations, along with measured wind speed and direction, to back-calculate the source strength.

The upwind-downwind method is the least accurate of the three methods described, owing to the very small portion of the emissions that are captured for measurement and the extreme degree of dilution in the transporting ambient air. It is the most universally applicable method, capable of identifying and quantifying emissions from sources indoors or out, large or small, under any operating conditions or schedules. It is sensitive to adverse weather conditions, changes in wind direction and such other outside influences as wet or snow-covered ground, extended dry periods and the like.

The upwind-downwind method can usually be utilized without even considering its affect on plant operations or schedules since the entire operation is so remote from the emissions site.

3.0 SAMPLING METHOD SELECTION

The selection of the most effective method for the measurement of fugitive emissions at an industrial site is influenced by a number of factors relative to the emissions, the process or operation involved, and the source location and arrangement. The degree of influence of

each of these factors in the large number of possible combinations of factors at any given site is so variable that no simple selection process can be developed to cover more than a few specific cases. Each site must be considered as a unique situation and a selection made on the basis of the criteria uniquely or most rigorously affecting that site.

3.1 Selection Criteria

The general selection criteria described below are grouped into three classifications common to all air borne fugitive emissions measurement methods, providing representative examples of the influencing factors to be considered. Other factors will present themselves at any specific site. Some degree of judgment will be required to determine the relative importance any factor may carry.

SITE CRITERIA - factors influenced by geometry, physical layout and location of the facility or source.

Source Isolability - can the emissions be measured separately from other emissions? Can the source be enclosed?

Source Location - is the source indoors or out? Does the location permit the installation of sampling equipment?

Meteorological Conditions - what are typical and critical situations? Will wind or precipitation interfere with measurements? Will wet or snow-covered ground alter emission rates or characteristics?

PROCESS CRITERIA - factors influenced by the nature and extent of the process producing the emissions.

Number and Size of Sources - are emissions from a single location or many scattered locations? Is a single source small enough to hood?

Homogeneity of Emissions - are the emissions of the same character throughout the process? Are reactive effects between emissions involved?

Process Continuity - will emissions be produced steadily for sufficient time to obtain measurable samples? Must sampling be performed over a number of process cycles?

Measurement Effects - are special procedures required to prevent the making of measurements from altering the process or interfering with production?

POLLUTANT CRITERIA - factors influenced by the nature or concentration of the emissions.

Types of Emissions - are measurements of particulates, gases or aerosols required? Are the emissions hazardous?

Emission Generation Rate - is the rate of emission release sufficient to provide measurable samples in a reasonable time period?

Emission Dilution - will transport air reduce emission concentrations below detectable limits before the sampling point? Are baseline measurements of transport air required to identify possible masking effects?

3.2 Criteria Application

The application of the general selection criteria described above to each of the air borne fugitive emissions sampling methods is described briefly and in general terms below. In practice, the application procedure would be addressed to the measurement of specific emissions at a specific site rather than to any measurement method.

Quasi-Stack Method

Effective use of the quasi-stack method requires that the source

of emissions be isolable and that an enclosure can be installed capable of capturing emissions without interference with plant operations. The location of the source alone is not normally a factor. Meteorological conditions usually need be considered only if they directly affect the sampling.

The quasi-stack method is usually restricted to a single source and must be limited to two or three small sources that can be effectively enclosed to duct their total emissions to a single sampling point. Cyclic processes should provide measurable pollutant quantities during a single cycle to avoid sample dilution. The possible effects of the measurement on the process or emissions is of special significance in this method. In many cases, enclosing a portion of a process in order to capture its emissions can alter that portion of the process by changing its temperature profile or affecting flow rates. Emissions may be similarly altered by reaction with components of the ambient air drawn into the sampling ducts. While these effects are not necessarily limiting in the selection of the method, they must be considered in designing the test program and could influence the method selection by increasing complexity and costs.

The quasi-stack method is useful for virtually all types of emissions. It will provide measurable samples in generally short sampling times since it captures essentially all of the emissions. Dilution of the pollutants of concern is of little consequence since it can usually be controlled in the design of the sampling system.

Roof Monitor Method

Practical utilization of the roof monitor method demands that the source of emissions be enclosed in a structure with a limited number of

openings to the atmosphere. Measurements may usually be made only of the total of all emissions sources within the structure. Meteorological conditions normally need not be considered in selecting this method unless they have a direct effect on the flow of emissions through the enclosure opening.

The number of sources and the mixture of emissions is relatively unimportant since the measurements usually include only the total emissions. The processes involved may be discontinuous as long as a representative combination of the typical or critical groupings may be included in a sampling. Measurements will normally have no effect on the processes or emissions.

The roof monitor method, usually dependent on or at least influenced by gravity in the transmission of emissions, may not be useful for the measurement of larger particulates which may settle within the enclosure being sampled. Emission generation rates must be high enough to provide pollutant concentrations of measurable magnitude after dilution in the enclosed volume of the structure.

Upwind-Downwind Method

The upwind-downwind method, generally utilized where neither of the other methods may be successfully employed, is not influenced by the number or location of the emission sources except as they influence the locating of sampling devices. In most cases, only the total contribution to the ambient atmosphere of all sources within a sampling area may be measured. The method is strongly influenced by meteorological conditions, requiring a wind consistent in direction and velocity throughout the sampling period as well as conditions of temperature, humidity, and ground moisture representative of normal ambient conditions.

cise identification and quantification of specific emission constituents that a survey level system or similar reliable data indicates may be present in concentrations of concern. They utilize the latest state-of-the-art measurement instrumentation and procedures in carefully designed sampling programs to provide data accuracy of ± 10 to $\pm 100\%$ of actual emissions, often over a range of process operating or ambient meteorological conditions.

The differences in complexity of the two levels of sampling as applied to the measurement of emissions from a foundry pouring operation are illustrated in Figure 4-1.

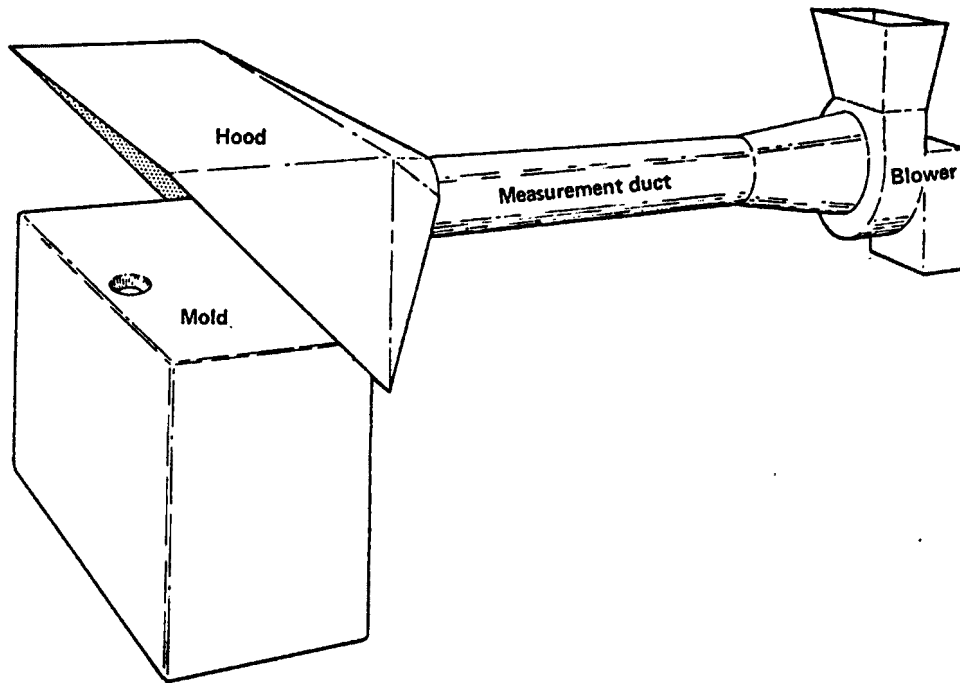
5.0 SAMPLING PROGRAMS COSTS AND TIME REQUIREMENTS

In order to prepare estimates of manpower, cost and time requirements that will permit valid comparisons among the three measurement methods for air borne fugitive emissions, it is necessary to first define a factor or characteristic common to all methods as a basis for comparison. The methodologies, techniques, and equipment requirements are too diverse among the methods to provide that basis. The most significant common factor among the methods is one that is within control of the test program designer, the overall accuracy of the measurements made. The accuracy is relatively easily manipulated over a generally wide range by such means as instrument selection and test replication.

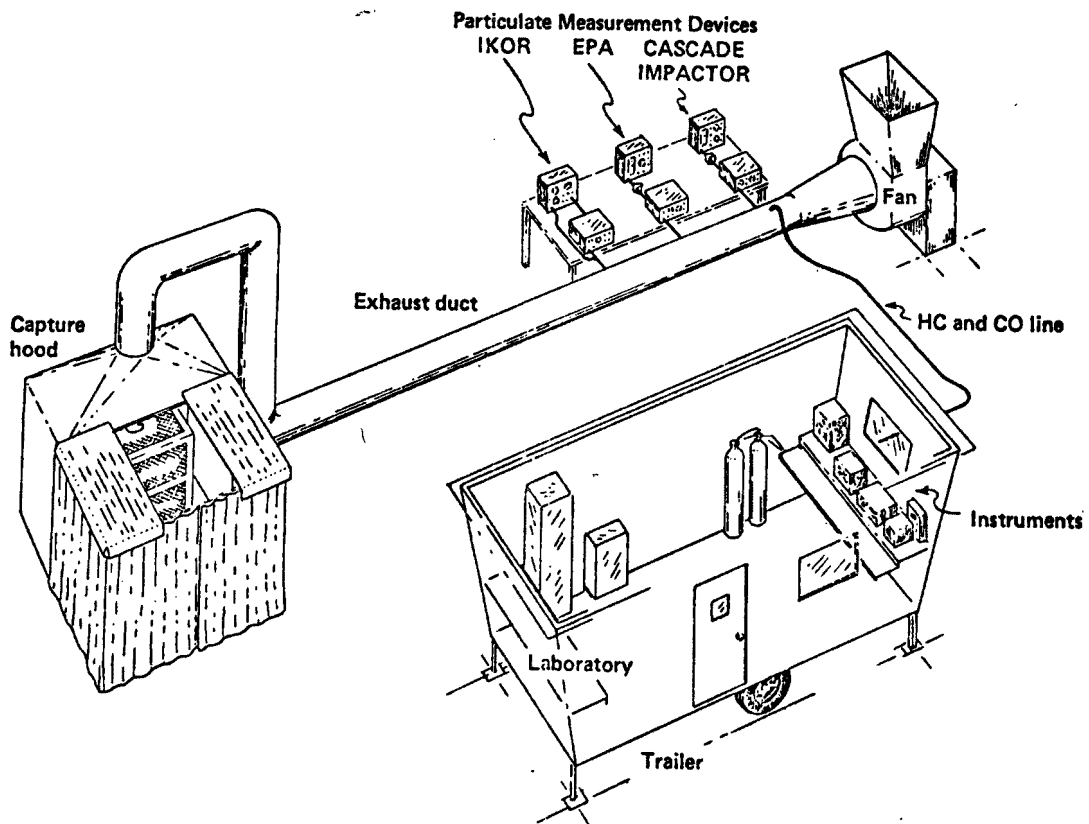
In preparing the estimates presented in Table 5-1 for overall accuracies of $\pm 500\%$, 200%, 100% and 50% for both survey and detailed measurement systems in each of the three methods, the following assumptions were made:

- o Site accessibility equal for all programs.
- o Emission source accessibility and isolability as required for each program.

Figure 4-1: Survey and Detailed Systems for Foundry Mold-Pouring Measurements



Survey System



Detailed System

TABLE 5-1

FUGITIVE EMISSIONS SAMPLING PROGRAMS
COST AND TIME REQUIREMENT ESTIMATES

Measurement Technique	Accuracy Level-%	Manpowers- Man-hours	Field Study Costs-\$	Equipment Costs-\$	Total Cost-\$	Duration, Weeks
Quasi-Stack (Survey)	±500	480	1,000	2,600	18,000	12
	±200	940	2,000	2,800	33,000	16
Quasi-Stack (detailed)	±100	1,400	4,000	12,000	58,000	24
	± 50	2,600	7,000	19,000	105,000	36
Roof Monitor (survey)	±500	380	800	1,800	14,000	12
	±200	750	1,700	2,800	27,000	16
Roof Monitor (detailed)	±100	1,200	3,400	5,600	45,000	20
	± 50	1,800	6,200	19,800	80,000	28
Upwind-downwind (survey)	±500	480	1,100	4,500	20,000	12
	±200	1,020	2,600	8,800	42,000	16
Upwind-downwind (detailed)	±100	2,300	5,000	34,000	100,000	26
	± 50	4,200	10,000	64,000	200,000	40

- o No interference due to meteorological conditions.
- o Emissions homogeneous and continuous for all programs.
- o No process or emission alteration by measurement program.
- o Particulates and gases measured in all programs.

The table presents comparative estimates of total manpower requirements, field study transportation and subsistence costs, equipment purchase costs, total time duration from test planning through report preparation, and total costs. Total costs include manpower at \$30 per hour, field study costs and equipment purchase costs. The total cost for each measurement program is plotted versus % accuracy in Figure 5-1 for easier comparison.

More detailed information relative to the selection, design and application of the three air borne fugitive emissions measurement programs is contained in the Environmental Protection Agency Technical Reports EPA-600/2-76-089a (b,c); entitled "Technical Manual for Measurement of Fugitive Emissions: Upwind/Downwind (Roof Monitor, Quasi-Stack) Sampling Method for Industrial Emissions."

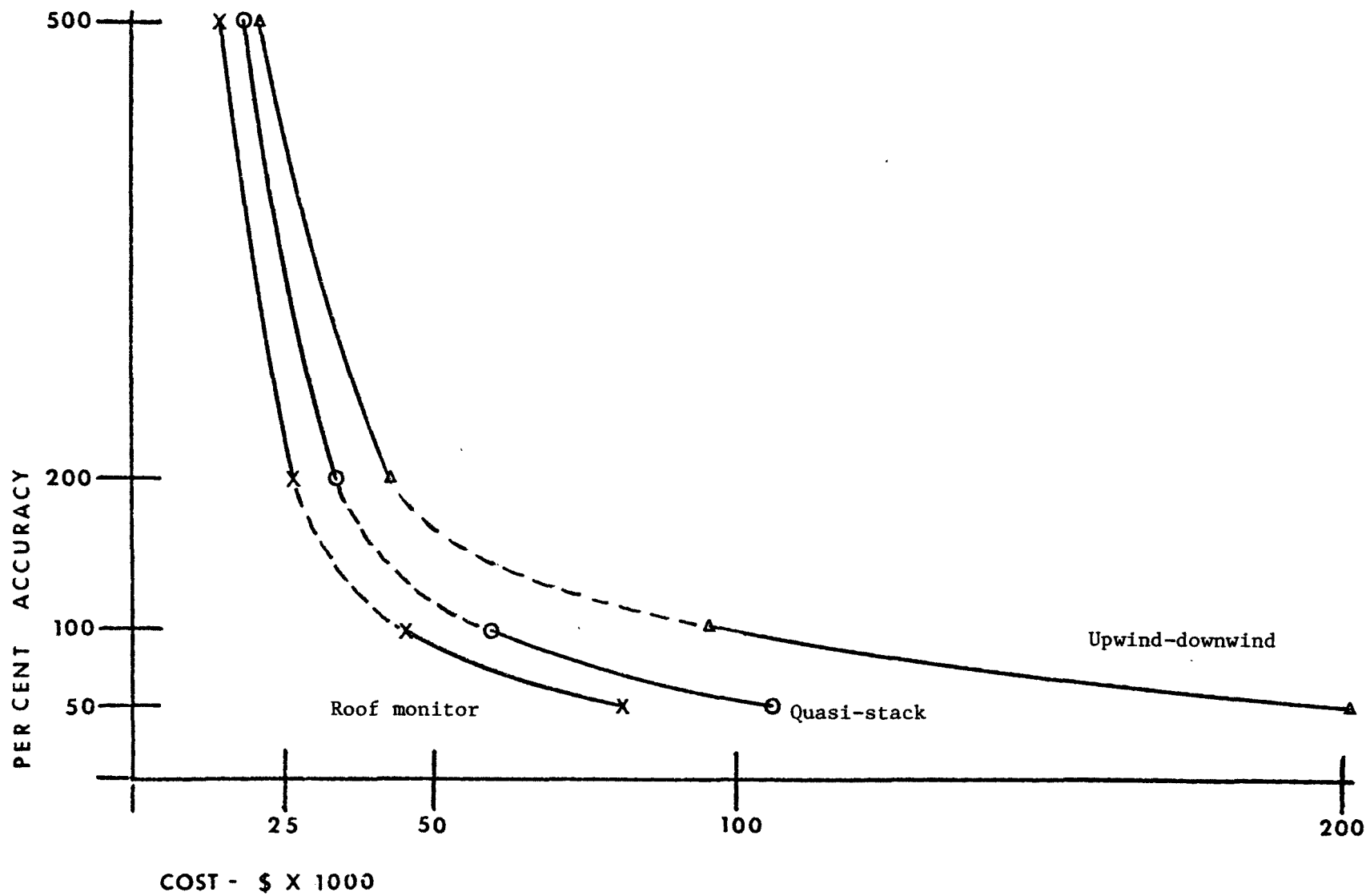


Figure 5-1: Measurement Program Costs as a Function of Overall Accuracy

COKE OVEN EMISSION MEASUREMENTS DURING PUSHING

Robert B. Jacko, Ph.D., Purdue University

I came here today to discuss the fugitive emission problem from coke oven pushing. Purdue University's Civil Engineering Department has been involved with a National Science Foundation funded program since 1972. The purpose of the program was to characterize atmospheric emissions including trace metals from a number of point sources. Some of those point sources included an open hearth steel making furnace, a municipal refuse incinerator, a zinc smelter coker, vertical retort and sinter plant, and a coal fired power plant. One of the problems we identified back in '72 was the coke oven pushing emission problem. At that time we didn't think about it in terms of a fugitive emission -- indeed, the word was not yet coined. So we began to discuss various sampling techniques; in order to quantify the coke oven pushing problem.

A mobile laboratory was constructed, the top of which contains necessary hardware to enable us to sample at the coke ovens during pushing without the constraints of a hood. In other words, sampling could be achieved in the actual push plume in the free atmosphere with no restraints of a hood. A description of the coke ovens is in the following text.

There were 65 ovens in the particular battery that was sampled. Refer to Figure 1. Dimensions of the ovens are 18' wide, 12' high, and 40' in length. Typical charge was about 15 tons of coal -- and obviously that varies, however, this is a typical value. These ovens are approximately 20 years old.

Some of the problems that we encountered while designing the sampling methodology was, first of all, should we position our sampling equipment off the top of the battery and attempt to get into the plume that way. One of the problems was that the Larry car is operated with large DC bus bars and they're

located right in this area on the ovens. Therefore, we didn't want to position any hardware off the side of the ovens which could possibly come in contact with those DC bus bars. It was decided to go with a mobile approach, which provided complete independence from the batteries. In other words, we don't have to rely on, necessarily, perfect synchronization between the operators and our sampling program -- we could operate independently. We wanted to be there when they pushed the coke and we knew what the schedules were, so we worked it out that way. But as many of you know, when you work in a large steel mill complex, there are interdepartmental lines to work across -- the research people work with operations people and they have their own schedules to meet and you don't want to interfere with those schedules any more than you have to. So we felt that going with a mobile sampling arrangement such as this would provide us with the flexibility that was needed.

The general arrangement of the coke ovens, quench car and mobile sampling laboratory is seen in Figure 2. At the completion of the coking cycle (18-20 hrs), the coke oven door is removed and the coke is pushed at a predetermined constant rate by the ram into the quench car. Any remaining volatiles in the carbonized coal ignited in the oxygen-rich atmosphere and incomplete combustion results in the release of particulate matter to the atmosphere. The sampling is initiated when the visible emissions start emanating from the coke quench car. Blowers located on a boom approximately 30' above grade are then activated. Samples would be withdrawn at as close to an isokinetic rate as we could do, as we could set up, and we would draw a sample at about 35 CFM. Now, we needed the high flow rate because, as you know, during pushing, a typical push time is anywhere from 40 seconds to 50 seconds. The 35 CFM enabled extraction of a relatively large sample volume over the push time so that the sample would represent as closely as possible the contents of the actual plume. The samples were taken isokinetically by monitoring a wind anemometer that we

used on the end of the boom and plume temperature was also measured with two thermocouples at the end of the boom.

Figure 3 shows the location of our two cameras which we use to characterize the shape of the plume. The methodology we decided to use was to measure the plume particulate concentration, velocity and cross-sectional area, then compute the mass emission rate. That was the basic approach. The problem then becomes one of trying to characterize the plume shape. At first, we thought it would be just about impossible to do that on motion picture film, but we went ahead, anyway, and looked at some of the results, and I think we're getting reasonable values.

Figure 3 shows a quench car in a quasi-schematic form, with the development, drawn in arbitrarily, of the push plume. We locate one camera, shown here as "A", so that it views through a plane parallel with the front of the coke ovens; and the other camera at location "B". We had two students, each manning a camera -- one at "A" and one at "B". At the onset of pushing, we would commence filming. 16 mm. motion picture films were taken from cameras that were calibrated so that we knew that we were taking pictures at an exact frame rate, so later on we could go back to cross check plume velocities against the anemometer. In this fashion, we attempted to characterize what the shape of the plume was through a plane taken at the sampling heads.

After analyzing many, many feet of film, a ratio of length A to B was roughly 1.5 to 1.0. In other words, some of the data that I'll show you assumes a circular plane cross section. That's an idealization, not necessarily true. Analysis of the film shows that on the average, the plume shape, 30' above grade, for both clean and green pushes, the aspect ratio, if you will -- was 1.5 to 1. We expected greater deviation than that and certainly there was more deviation on individual pushes. But if you give me the liberty of taking the mean value, we found it to be 1.5 to 1. We plan to try other cross sectional

shapes and estimate the resulting effect on the mass emission rate. For example, elliptic shape, possibly a hexagonal shape, and other shapes, but we'll probably stay close to the circular or some modified circular shape. So that was the idea -- to characterize the concentration in the plume at that same elevation and try to characterize the cross sectional area. Knowing the flow velocity up through that plane, we computed the plume volumetric flow rate and, ultimately, the particulate mass emission rate.

Figure 4 is a schematic, looking down on top of the entire sampling apparatus. Note the 16' van truck, outfitted with the instrumentation inside the truck. We have two 10-foot sections of off-the-shelf TV tripod tower supporting the boom off the top of the truck. Since the truck sits about 10' off grade, we are about 30' above grade to the boom. The boom is roughly 25' long and is outfitted with high volume blowers. These blowers at free delivery will develop about 60 to 80 CFM. Two blowers were located quite a distance from the plume for environmental considerations. We didn't want to burn the armatures on the motors. We ran aluminum tubes -- 2" aluminum tubes -- up to three sampling heads, in which were mounted 8 x 10 glass fiber filters, the same filters you use on the standard high volume samplers. At first, we wanted to use a Stausscheibe pitot tube. However, the mean vertical velocity of the plume was so low that we'd be operating down near its resolution points, so we decided to use an anemometer mounted in a horizontal direction. We also mounted two thermocouples on the end of the boom. So this was the general arrangement of our sampling hardware. To measure the flow velocity of the sample, we used orifices at the outlet of the blower and calibrated that orifice with respect to the flow rates that we'd be using so that we could set up a known flow.

Figure 5 is a schematic showing the instrumentation and data handling system. We bring our sample in through a stainless steel sampling snout, which

is about 2½' long. The isokinetic sample, or the variation in the isokinetic rate that we expected, was taken care of by welding an expanding cone on the front side of the snout which could be shortened on site, just prior to sampling to put us into an approximate isokinetic rate. The blowers were variable speed to allow setting up isokinetic rates during sampling. The snout came up inside the transition for the filter holder. We found on some of our earlier samples that much of the material was extremely large, and after we shut down the blowers after the push was completed, we lost material back out the snout because of buildup. So we then modified the sampling snout to bring it up inside the transition so that it would act like a quasi-settling chamber and the big particles would not then fall back out the sampling snout. This worked quite well. The sample then passes through an 8 x 10 glass fiber filter, the same variety used in the high vol paths, and then via 2" diameter aluminum tubes to the blowers. Each blower had an orifice plate installed at its' outlet which was calibrated, and the resulting pressure was read out on a magnehlic gauge in the truck. The anemometer was calibrated prior to use and that, also, read out to a digital recording volt meter in the truck. By the way, we were using an Esterline Angus D 2020 Digital Volt Meter to record all sensor signals.

Comments at this point would be that we had expected problems due to flame burning up our filter pads, and of the first 15 samples that we extracted (my presentation deals with those first 15 -- we now have about 66 samples from pushes at the coke oven, the same battery), we've only had flame problems on two of the samples that we withdrew, which resulted in a fusion of the glass pad. But the 2½' sampling snout appears to act as an adequate flame-arrester. We had considered using other elaborate systems of flame arrest in the sampling nozzle; but found that not to be necessary. Also, on the initial design of the horizontal boom, we thought that it was going to be necessary to install

water-cooling on the boom, but, again, had no major problem in that respect. After our first few preliminary tests, we stripped off the water-cooling arrangement that we had on the boom and on the structural members and we haven't had any problems due to temperature. However, the plume temperatures were not as high as expected in the plume. Maybe the fact that we were sampling a couple of days before Christmas last year and it was -18 degrees Centigrade helped us.

The 16mm film record is a very important part of the data analysis, because we have to correct the sample based on the time period that we were in the plume, since we were not in the plume 100% of the time. The film was analyzed with stopwatches to determine the net time that the sample heads were in the visible portion of the plume. The total sample volume was then corrected for the amount of time that we were not in the plume. We made the assumption that when we were not in the plume and we couldn't see a visible emission, that the concentration of particulates that we were sampling at that point in time would be relatively low compared to the concentration of particulates when it was, indeed, immersed in the visible portion of the plume. That's an idealization and it has problems, but that's what we've done.

Now that we've taken the sample, we wish to retrieve the sample from the hardware. The procedure here is to release the two guy wires from the back of the truck that were connected up near the blower end, allowing the boom to swing on a hinge plate arrangement that we designed. The sampling heads are then lowered down into the vicinity of the top of the truck where the people can get to the heads and retrieve the entire bottom half, including the glass fiber filter. This assembly is taken into the laboratory environment within the truck, where the filter is retrieved and the sampling head backwashed with acetone.

How about summarizing, now, what we've done. We've sampled 15 coke oven

pushes. This was on a 3-day period just before Christmas 1975 with relatively low ambient temperatures of minus 18 C. We categorize clean and green pushes by visual observation. We had 8 clean pushes and 7 green pushes. Refer to Figure 6. Average plume temperatures in degrees centigrade were found to be 32 for the clean and 70 for the green pushes. Now, this may not be the true temperature of the plume -- it's the temperature that we measured with our two radiation shielded thermocouples, located as best we could in the plume. The plume does swing so this probably is an average of upper plume temperatures and ambient temperatures. Average plume velocity was found in clean pushes to be 5 meters per second; in green pushes, 4.4. These were taken with the mounting of the anemometer in the horizontal plane.

Figure 7 contains the particulate concentrations and mass emission rates from the 15 pushes that were sampled. Note that the clean and green push particulate concentration is 1.5 and 2.4 grams/cubic meter respectively. The mass emission rate of the green pushes was found to be 407 grams/sec as compared to 147 for the clean pushes.

The average emission factors, based on a 50 sec push, were calculated as 1.6 lb.-part./ton-coke for the 8 clean pushes and 4.5 lb.-part./ton-coke for the green pushes. In terms of tons of coal charged to the ovens, these values are 1.0 and 3.0 for clean and green pushes respectively. The average is 2.0 lb.-part./ton-coal and compares to 0.4 lb.-part./ton-coal as published in AP 42. Note the coefficient of variation (CV) on the mass emission rate. The CV is defined as one standard deviation from the mean value, divided by the mean value. As you can see, the variability is quite high. In clean pushes, the standard deviation was as high as 74% of the mean value; in green, it was 92% of the mean value, and the overall value was 110.

It appears as though our concentration/ photographic technique, is perhaps viable; that you can utilize it with some degree, hopefully, of representativeness

of what is in that plume. Secondly, we found that the circular plume cross section that we used for computation of a mass emission rate is at least a good approximation, based on the film analysis and the aspect ratio of 1.5 to 1, and this number of 1.5 to 1 comes from an observation of many, many frames of motion picture film, literally thousands.

Conclusions

So what you've seen here is an attempt by us to quantify a fugitive emission from a coke oven pushing operation. We designed the hardware and the sampling methodology much before the time that fugitive emissions were discussed. Our main objectives at that time were to quantify the flow of trace metal emissions into the atmosphere, as well as looking at the total particulates. Work that we have for the future includes not only the mass emission rate of the total particulates, but we've modified our boom and we're going to instrument it for hydrocarbons, for particle size distribution, using inertial impactors -- we'll be mounting two inertial impactors at the boom so we can get Anderson aerodynamic particle size distribution.

COKE OVEN DATA

MANUFACTURER: Koppers

NUMBER IN BATTERY: 65

DIMENSIONS: Width 18 1/4", Height 12', Length 40'

CAPACITY: 15 Tons Coal, Typical Charge

Figure 1 Coke Oven Description

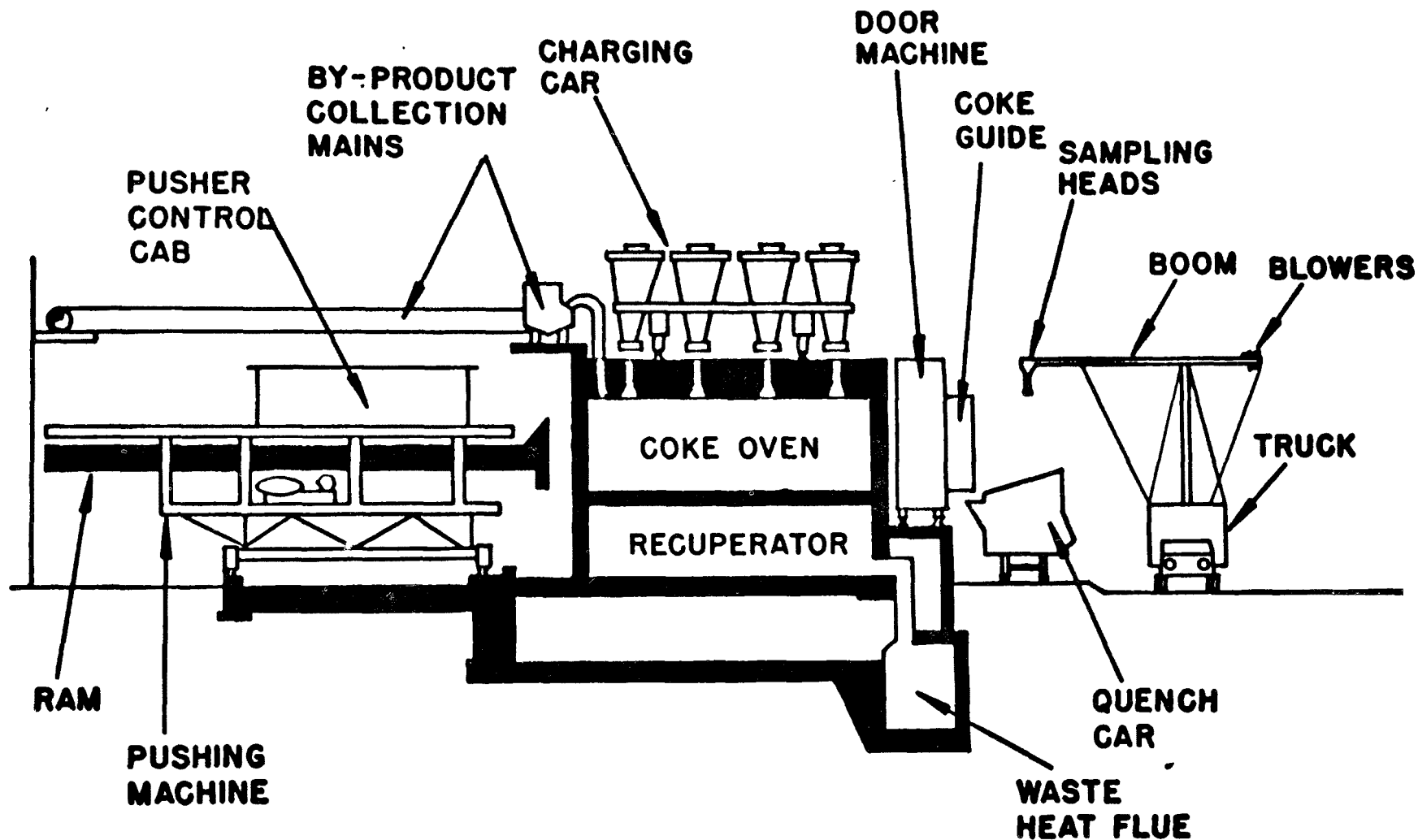


Figure 2 Cross section of coke oven battery, including sampling vehicle

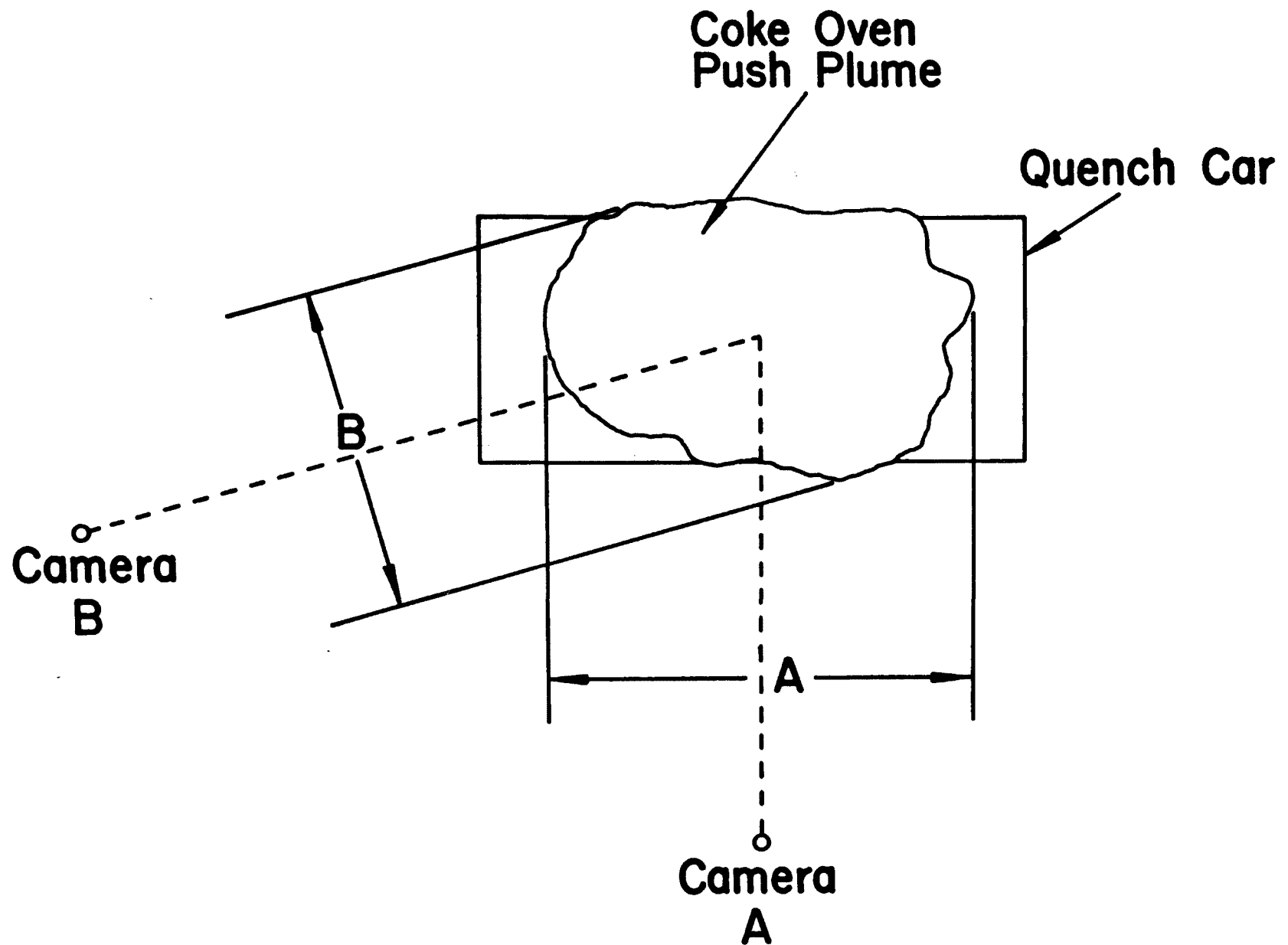


Figure 3 Location of Two 16mm Cameras

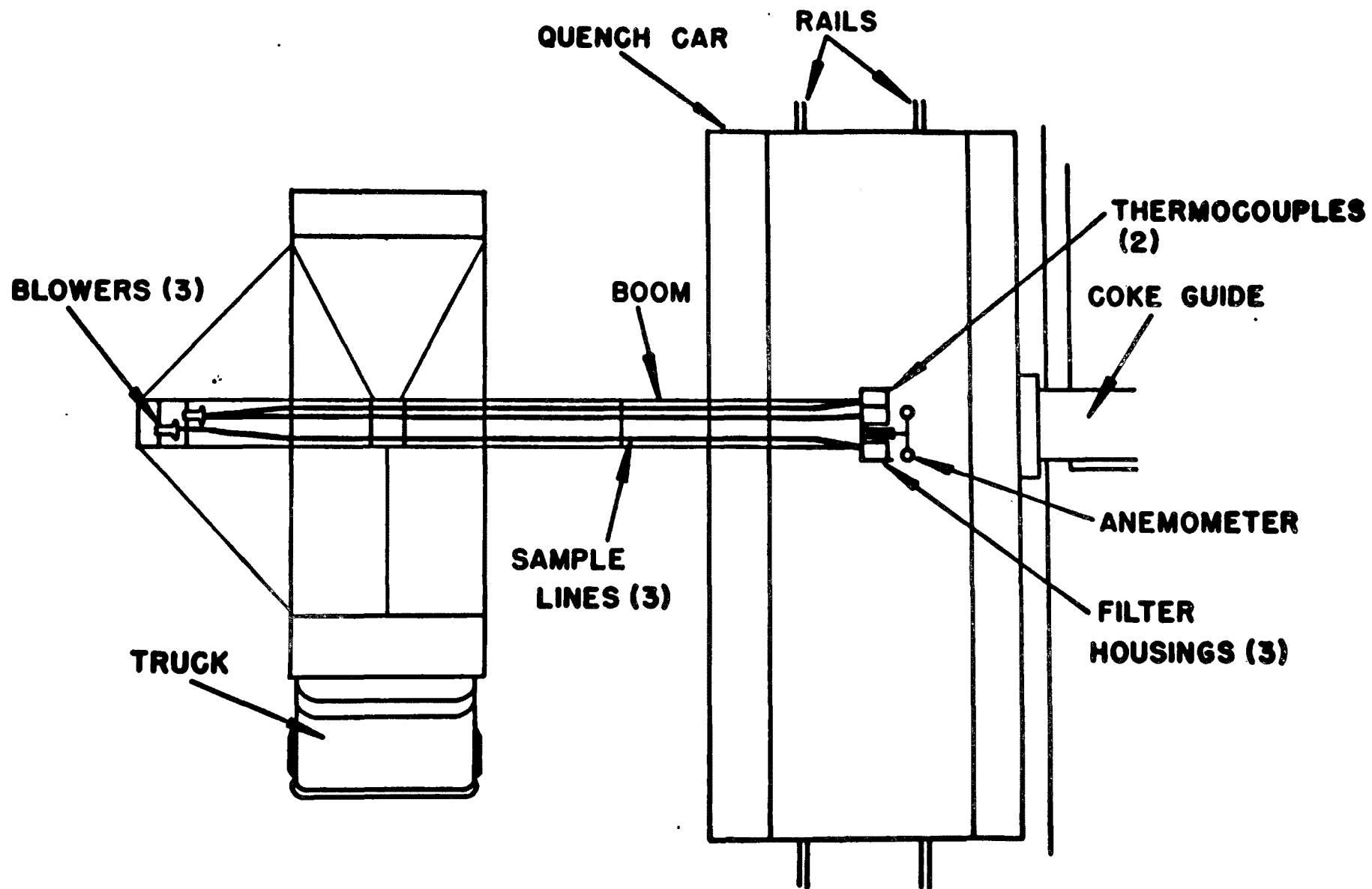


Figure 4. Top view of sampling vehicle and coke quench car

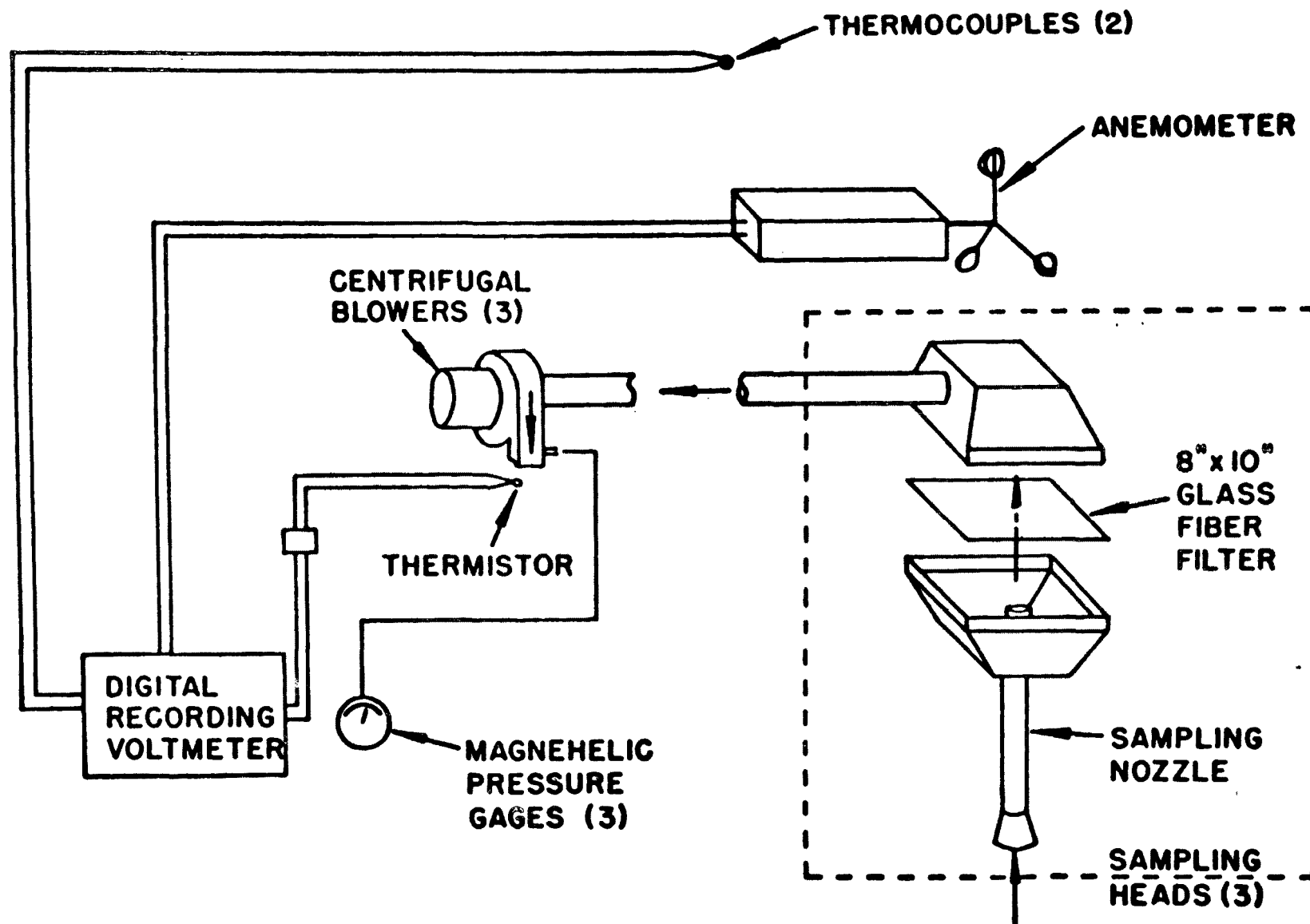


Figure 5 Schematic diagram of sampling heads and instruments used on coke oven sampling boom

TOTAL NO. OF SAMPLES: 15

CLEAN PUSHES: 8

GREEN PUSHES: 7

AMBIENT TEMPERATURE: -18 C

AVERAGE PLUME TEMP., C

CLEAN PUSHES: 32

GREEN PUSHES: 70

OVERALL: 49

AVERAGE PLUME VELOCITY, M/SEC

CLEAN PUSHES: 5.0

GREEN PUSHES: 4.4

OVERALL: 4.7

Figure 6 General Summary of Coke Oven Push Sampling

PARTICULATE RESULTS

GRAIN LOADING,	GR/SCF	G/M ³
CLEAN PUSHES:	0.66	1.5
GREEN PUSHES:	1.1	2.4
OVERALL:	0.83	1.9
MASS EMISSION RATE,	G/SEC	CV(%)*
CLEAN PUSHES:	147	74
GREEN PUSHES:	407	92
OVERALL:	268	110

***CV= COEFFICIENT OF VARIATION
= 100 x STD. DEV. / MEAN**

Figure 7 Coke Oven Pushing Particulate Emissions

PROBLEMS IN MEASURING FUGITIVE
EMISSIONS FROM WASTE DISPOSAL PONDS

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PROBLEMS IN MEASURING GASEOUS FUGITIVE EMISSIONS FROM WASTE DISPOSAL PONDS

William R. King

Abstract

This paper discusses a method of measuring the emission rate of pollutants with finite vapor pressures from contaminated-liquid storage ponds. The techniques described were developed to quantify the emission of fluorine compounds from wastewater disposal ponds connected with wet-process phosphoric acid manufacturing plants. However, they are applicable to any water soluble chemical compound with a finite vapor pressure, such as hydrogen sulfide, phenol or hydrocarbons up to their limit of solubility in water.

Review of Available Methods

Pond emission measurement methods can be divided into two general groups:

- (1) Field measurements conducted at the actual emission source or
- (2) Laboratory measurements conducted on models.

Laboratory modeling usually utilizes a wind tunnel to simulate the pond and the surrounding atmosphere. The experimenter is concerned with evaluating Equations (1) and (2).

$$N_B = K_B (p_B^* - p_B) \quad (1)$$

$$N_B A = C_B u \quad (2)$$

where

N_B = molar mass transfer rate per unit pond surface
of component B from the pond to the atmosphere

K_B = Overall gas-side mass transfer coefficient for
component B

p_B^* = Partial pressure of B in equilibrium with
pond water

p_B = Partial pressure of B in atmosphere above
a pond

A_B = Pond surface area

C_B = Average concentration of B in atmosphere at
downwind edge of pond

u = Average wind speed or air velocity

In a wind-tunnel simulation, the average concentration - velocity product-- $C_B u$ --can be measured by placing a mixing box after the wind tunnel and sampling the gas stream coming out of the box. These measurements obviously can be used in Equation (2) to produce good estimates of the mass transfer rate. The wind tunnel data can be further reduced through Equation (1) to a mass transfer coefficient - K_B . This variable - K_B - is the basic parameter which must be translated from the wind tunnel to the real world.

Normally, the mass transfer coefficient is correlated against a measure of the turbulence in the air stream moving over the pond simulation in the wind tunnel. This correlation is applied to real world situations, if the pond could be considered a flat plant. Equation (3) has been empirically demonstrated to describe the mass transfer coefficient.⁽¹⁾

$$\frac{K_B X}{C_{D_{aB}}} = .0365 \left(\frac{XC_u}{\mu} \right)^{4/5} \left(\frac{\mu}{C_{D_{aB}}} \right)^{1/3} \quad (3)$$

where

C = molar concentration of air

D_{aB} = diffusivity of B in air

μ = air viscosity

ρ = air density

X = length of simulated pond in the direction of air flow

As the reader can see, this correlation, like all mass transfer correlations, requires that a charactic length - X - and a charactic wind speed - u - be defined.

(1) Equation (3) carries the explicit assumption that

K_B the average, overall, gas-side mass transfer coefficient is equal to the average gas-side mass transfer coefficient k_B .

The characteristic length that is used to describe mass transfer from a flat plate in a wind tunnel is well developed in the literature. This is the distance the wind travels across the plate. Unfortunately, the problem of characteristic length is considerably less well defined in the atmosphere.

(No Paragraph)

Is it the pond length in the direction of wind or something else? Likewise, the wind speed to be used is not very well defined. In the wind tunnel the bulk average wind speed can be measured easily and is normally used. In the atmosphere the bulk average speed is not defined.

Based on this brief discussion, the reader can see the difficulty in attempting to quantitatively translate wind tunnel experiments into the real world. I don't think the problem has been solved.

(No Paragraph)

Therefore, field measurements seem to be the only way to quantify emissions from ponds.

Three methods to estimate emissions from ponds seem possible:

- (1) In theory, direct calculation of an average pond emission rate from measurements of wind speed and pollutant concentration profiles in the vertical direction should be possible. Equation (4) is the defining equation.

$$N_B^A = \int_{\text{ground}}^{C_B=0} C_B du \quad (4)$$

- (2) Backing through a diffusion model from single point, concentration measurements, Equation (5),

$$N_B = C_B / f(A, X, Y, Z, \sigma_Z, \sigma_Y) \quad (5)$$

where

X, Y, Z = distance

σ_Z, σ_Y = vertical and horizontal dispersion coefficients

will also yield estimates of an average emission rate.

- (3) Finally, Equation (1) might be used to calculate emission estimates, if field verified estimates of the mass transfer coefficient were available.

Methods (1) and (2) directly measure some sort of average emission rate. More than one pattern of emissions from the diffuse source (pond) could satisfy either equation. However, when coupled with a completely defined Equation (1) (the third field measurement technique described above) which rigorously defines the emission pattern, either method can satisfactorily define diffuse source emissions.

Methods Used In This Study

In this study a combination of methods (3) and a modified method (2) were used to produce emission estimates and to verify them. A correlation for the mass transfer coefficient was developed from field data and used Equation (1) to produce emission estimates. These estimates were used in Equation (5) to predict pollutant ambient air concentrations downwind of the emission source. The predicted values were compared with field measurements of the ambient air concentration of the pollutant. Good agreement between the measured and predicted concentrations demonstrated the accuracy of the emission estimate.

The major difference between this study and other efforts to quantify fugitive emissions is that this study has available a method to estimate rigorously the diffuse source pattern of emissions. The emission estimates can be varied with time and in space in an entirely logical and rigorous manner.

Figure 1 is a work chart detailing the information development necessary to apply this technique. Study of the work chart will show that there are three major tasks. The first task is to develop the emission estimate from the mass transfer equation; the second is to measure ambient air concentrations in the field, and the third is to relate the emission estimates to the measured ambient air concentrations via diffusion modeling.

Calculation of Emission Rate

The right-hand side of Equation (1) is made up of the product of two groups. The first of these is a driving force, $p_B^* - p_B$, which is primarily a function of pond water temperature and the physical nature of the material being emitted by the pond. The vapor pressure, p_B^* can be measured in the laboratory; the atmospheric partial pressure, p_B , is small for "normal" sized ponds and usually can be assumed to be zero. Since this driving force is specific to the pond and to the material being emitted by the pond, it will not be covered in any detail in this report. The second group is the mass transfer coefficient- K_B . This term with straightforward modification, is applicable to any pond. For this reason, its development is discussed in this paper.

As stated previously, the overall average mass transfer coefficient describing material transfer from a flat plate into a flowing stream can be predicted from the Colburn Analogy, Equation (3), when the stream-air mass-is in turbulent flow.

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Since the physical parameters in Equation (3) do not change significantly over the range of conditions found in the atmosphere, they can be grouped with the constant. The result is Equation (6).

$$K_B = a u^{\cdot 8} X^{-\cdot 2} \quad (6)$$

In 1950 and 1951 the U.S. government measured water evaporation rates for Lake Hefner, the water supply reservoir for Oklahoma City. The lake is roughly elliptical in shape--about 2-1/2 by 1-1/2 miles. About 140 of the 500 separate daily, evaporation rate measurements made during the government study were used to develop estimates of the overall, average, mass-transfer coefficient. In addition to the lake data, evaporation rates were measured for four different types of evaporation pans:

1. The class A evaporation pan,
2. The Bureau of Plant Industries evaporation pan,
3. The Colorado evaporation pan and,
4. The screened evaporation pan.

Physical details of these pans are summarized in Table 1.

The measured evaporation rates were transformed into 24-hour-average mass transfer coefficients with Equation (1). Least squares correlations were developed for the logarithmic form of Equation (6)

$$\log_{10} K_b = \log_{10} (aX^{-.2}) + b \log_{10} u_{16} \quad (7)$$

where

u_{16} = wind speed 16 meters above pond surface
for Lake Hefner and each of the evaporation pans.

The resultant correlations were tested statistically; it was demonstrated that the correlations developed for Lake Hefner the Bureau of Plan Industries pan and the Colorado pan were not significantly different. On this basis the three sets of data were combined and a new correlation was developed from this combined data. The statistics for the latter correlation are shown in Table 2.

The slope-b-of the best-fit-linear-least-squares correlation for the screened pan was not significantly different from the slope of the combined data correlation (or the 0.8 slope); however, the intercept- $ax^{-.2}$ was significantly different. This was not too surprising since the screened pan is covered with a relatively small mesh screen that reduces the air turbulence above the water at a given wind speed. The expected effect of the screen is to reduce the mass transfer coefficient at a given wind speed; this was observed.

Both the slope and the intercept of the least squares correlation for the class A pan are significantly different from the combined data correlation. The class A pan sits about 15" above the ground in contrast to the other pans and the lake which sit flush with the ground. This elevation makes the pan look more like a circular cylinder than a flat plate. Wake separation can be expected to occur in the air stream flowing over the surface of the water. There has been no work done specifically to define mass transfer from a cylinder head--the Class A pan's configuration. However, measurements of mass transfer

to fluids flowing past surfaces where wake separation occurs (the curved surface of cylinders, rectangular bars, streamlined cylinders and spheres) demonstrates a wind speed dependency ranging from the 0.5 to 0.65 power. The exponent predicted by the best-fit-least-squares correlation for the Class A pan was 0.58--a reasonable value based on the above discussion.

Since the correlation based on the pooled lake, Colorado pan and BPI pan data shows a dependency upon wind speed not significantly different from $u_{16}^{0.8}$, the empirically well-documented 0.8 velocity exponent was assumed correct. The recommended predictor of the mass transfer coefficient is

$$k_w = .429 u_{16}^{0.8} \quad (8)$$

where

k_w = grams water per sq. meter-millimeter of mercury-hour

U_{16} = wind speed at 16 meters in meters per second

Examination of Equations (3) or (7) indicates that the mass-transfer coefficient should be dependent on pond diameter as well as wind speed. Statistically this difference doesn't exist. But to test further for the length dependency, Equations (3) and (8) were assumed to be the same equation and they were solved simultaneously for length. With this set of assumptions, length-X is equal to 12,000 meters. If the further assumption is made that the equivalent

length of the pans is 12,000 meters and the equivalent length of the lake is 12,000 meters plus two miles, then the ratio of the empirical value of the pan's $X^{-0.2}$ term to the lake's term should be 1.045. In fact, the ratio of the average of the Colorado plus the BPI pan's length-containing value to the lake's is 1.070. Perhaps a length effect does exist, but the effect cannot be separated statistically, and as demonstrated in this paragraph, the value is unimportant in the estimation of mass transfer from moderately sized bodies of water.

In the preceding paragraphs, a mass transfer coefficient describing the evaporation of water into air has been developed. A simple diffusivity adjustment must be made to transform this into a mass-transfer coefficient describing the evaporation of another material into air. Under assumptions previously enumerated the overall, gas-side mass transfer coefficient for water can be transferred into the overall, gas-side mass transfer coefficient for another species by adjusting the water coefficient by the diffusivity ratio to the two species raised to the 2/3 power.

$$K_B = K_W \left(\frac{D_{aB}}{D_{aW}} \right)^{2/3} \quad (9)$$

where:

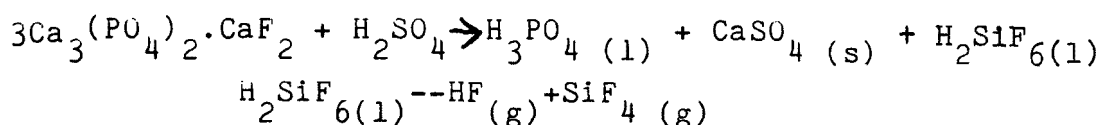
K_B = overall, gas-side, mass transfer coefficient
for species b

D_{aB} = diffusivity of B in air

D_{aW} = diffusivity of water in air

Field Measurements

Two waste disposal ponds connected with wet-process-phosphoric-acid manufacturing plants were studied. The digestion of the phosphate rich minerals with sulfuric acids yield product plus volatile fluorine compounds.



These gaseous fluorine compounds are scrubbed from the plant's air emissions and sent to wastewater ponds.

Over 85% of the fluorine entering the plant reports to the ponds.

The ponds chosen for the ambient air studies are shown in Figures 2 and 3. Pond 10 is a nominal 100-acre, roughly-rectangular, cooling pond. The land beyond the southeast and southwest edges is grass covered for at least 300 meters; it is generally flat and at the same height as the pond water. Pond banks are diked about two meters above the water level except in the area around the two sampling sites. Gypsum piles about 15 meters tall border the northwest and part of the northeast sides of the pond. Gypsum was not being accumulated on the northwest pond during the test period; the gypsum disposal ponds on top of the pile were dry. The northeast pile was

used for gypsum disposal; 50 acres of ponds covered the top of the pile, Pond 20 is a 160 acre-irregularly shaped cooling pond constructed on an old mine site. By-product gypsum was being stacked on the land mass in the center of the pond. The level of the pond was less than 1 meter below the edge of the banks. Small weed trees, 2-3 meters tall, cover the edge of the pond bank.

During the field work at pond 10, winds were usually southerly. Overcast skies and squalls were common; 3(C) and 4(D) stability classifications predominated. The wind speed measured three meters above the ground varied from 1.5-6.3 meters per second.

At pond 20 the wind usually blew from the east although it varied over the day. Sunny skies and 1(A) or 2(B) stability classifications were common during actual sampling period. However, rain squalls frequently occurred in late afternoon and inversions were common in the early morning. Wind speed at 3 meters varied from 0.9 to 3.5 meters per second.

The following data was obtained in the field:

1. One-hour-average, ambient-air fluorine concentrations.
2. Wind speed at three meters above the ground.
3. Wind direction at three meters.

4. Cloud cover estimates, and air stability estimates.
5. Pond water inlet and outlet temperatures.
6. Pond water fluorine concentration.

In the study of pond 10, six upwind samples were taken at station 3, Figure 2. Three of these samples showed no ambient air fluorine content, and three showed an average of about $0.8 \mu\text{g moles/m}^3$ of fluorine not emitted by the pond. The fluorine source was not identified; however, assuming reasonable atmospheric dispersion, it can be demonstrated that the maximum contribution to the measured fluorine concentration at the downwind samplers was about $0.10 \mu\text{g mole/M}^3$ of fluorine--about 5% of the total fluorine measured. This value is the same magnitude as the limit of measurement. Upwind samples from pond 2 were not obtained because of the inaccessibility any upwind site. However, visual inspection of the area revealed no obvious source of fluorine emissions east of the pond closer than about 4 kilometers. Therefore, the effect of other fluorine sources would be similar to those experienced around pond 1; that is, their contribution to the measured concentration would be about the same magnitude as the limit of measurement.

Reconciliation of Data

In review, the paper has so far presented a method to predict emissions from waste ponds. This method depends only on the pollutant being emitted, the pond temperature and the wind speed. The method was used to predict fluorine emissions from wet-acid plant ponds. In addition, the paper describes the measurement of ambient air fluorine concentrations downwind of two wet-acid ponds. How can these two sets of information be compared?

Examination of the diffusion model indicates two possible ways of comparing the two data sets. A back-calculation can be made from the field-measured concentrations through the diffusion model to obtain estimates of the emissions from the pond. These can be compared to the emission rates calculated from the mass-transfer equation. Conversely, the estimated pond emissions can be used in the diffusion model to predict ambient air concentrations at the downwind samplers. These values can be compared with the field measured volumes. Neither method is entirely satisfactory.

Back-calculating emission estimates from ambient-air field measurements requires that the fluorine emission rate from the pond be distributed in some manner. As you remember, at a given wind speed, the emission rate depends upon the fluorine vapor pressure. This in turn is a function of the pond water temperature and the tempera-

ture variation over the pond. The logical way then is to distribute emission rates over the pond surface in proportion to the experimental vapor pressure. Unfortunately the vapor pressure is also in the mass transfer equation. Therefore, both the emission estimate developed from the mass transfer equation and the estimate developed from the measured ambient air concentration utilize the measured vapor pressure of fluorine over pond water. Because both are derived from vapor pressure measurements, neither emission estimate is independent of the other. Statistical comparisons of the two estimates are dangerous. The statistics drawn from the two sets of data may compare vapor pressure with vapor pressure.

This statistical problem can be avoided if ambient air concentrations are compared. In this case, vapor pressure is utilized only in the calculation of ambient air concentration from the diffusion model. In addition, this comparison has an advantage in that all the estimated or measured values--the rate of emission estimate and the diffusion model--are lumped in the calculated concentration. The field-measured concentration is a "pure" number which has not been subjected to any manipulative calculations. The disadvantage of comparing ambient air concentrations is that the emission-rate-calculation method is indirectly tested.

A computer program was developed to calculate ambient air concentration. This program divides the sample time into 10 minute time periods. It divides the pond water studied into a number of isothermal segments. A ten meter wide strip perpendicular to the average wind direction of this isothermal segment is approximately considered to be a finite line source. The program calculates the mass-transfer coefficient per unit length from the strip and assumes that the entire emission comes from a single line at the center of the strip. It calculates the time weighted contribution of this finite line source at the sampler and repeats the calculation for all strips within an isothermal segment and for all segments of the pond. Figures 4 and 5 are graphic comparisons of the calculated and the actual field-measured ambient air concentrations.

(No Paragraph)

The figures also include the 45° , or perfect correlation, line. Tables 3 and 4 summarize the statistical tests which compare the measured and calculated corrections.

As these tables and Figures 4 and 5 show the hypothesis that the least-squares, best-fit lines are not significantly different from perfect correlation line is confirmed. Therefore, the method of predicting ambient air fluorine concentrations developed in this paper are confirmed by field measurement. By extension, the method of predicting pond emissions developed in this paper has also been confirmed.

As the reader knows, when making field measurements in the atmosphere, every now and then a bad experiment occurs. Sometimes these runs can be explained and sometimes they can't. The field studies described in this paper were no different. The experiments discarded in this study are shown in Figures 6, 7, 8 and 9 along with a short explanation on what may have gone wrong. Other than the data shown in Figure 8 for pond 20, the discarded runs could have been correlated with the main body of data with no appreciable effect on the study's conclusion. The same can not be said about the data shown in Figure 8; its inclusion would change the slope of the least squares correlation for pond 20 from $+45^{\circ}$ to -30° .

Conclusion

This paper proposes and demonstrates a method to estimate emission from ponds containing water-soluble, volatile atmospheric pollutants. The method has general applicability. To apply it to a specific pond and pollutant, the only additional information that must be developed is the pollutant's vapor pressure curve.

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BV24

Acknowledgment

This paper is based on "Fluoride Emissions from Phosphoric Acid Plant Gypsum Ponds"--EPA-650/2-74-095, a report prepared for the Office of Research and Development, U. S. EPA, Washington, D.C. EPA project officer was B. N. Murthy, Control Systems Laboratory, NERC, Research Triangle Park, N. C. 27711.

Table 1 Pan structural details

Type	Diameter (Ft.)	Depth (Ft.)	Relation of Water Surface to Ground Surface
Class A	4	0.83	(est) 15-18" above ground
Screened ($\frac{1}{4}$ inch mesh screen over top of pan)	2	3	even with ground
Colorado	3 ft. sq.	1.5	even with ground
Bureau of Plant Industries (BPI)	6	2	even with ground

Table 2 Statistical Summary of Correlation Developed for
Combined Lake Hefner, PBI Pan and Screened Pan
Data Base

$\log_{10}(ax^{-.2})$ (Standard Error)	b (Standard Error)	Number of Observations	F	R ²
-.3487 (.02477)	.7727 (.03497)	319	488.254	.6063

$$^a \log_{10} K = \log_{10}(ax^{-.2}) + b \log_{10} u_{16}$$

$$K = \text{g moles/hr m}^2 \text{ mm Hg}$$

$$u_{16} = 16 \text{ meter velocity in meters/second}$$

Table 3 Least squares regressions of calculated ambient air
fluorine concentrations on measured concentrations

Pond	a^v (Standard Error)	b^v (Standard Error)	Number of Observations	F	R^2
10	.2536 (.16825)	.9122 (.06346)	50	206.58	.81145
20	.1043 (.15425)	.9888 (.12911)	37	58.649	.62626

Table 4 Testing the coefficients of the least squares regressions of the calculated fluorine concentration on the measured fluorine concentration with the student's t test to determine if $a^v = 0$ and $b^v = 1$

Pond	Value of t For a^v	Significant ^a	Value of t For b	Significant ^a
10	1.507	No	1.383	No
20	.67626	No	.0868	No

^aAt 95% confidence level

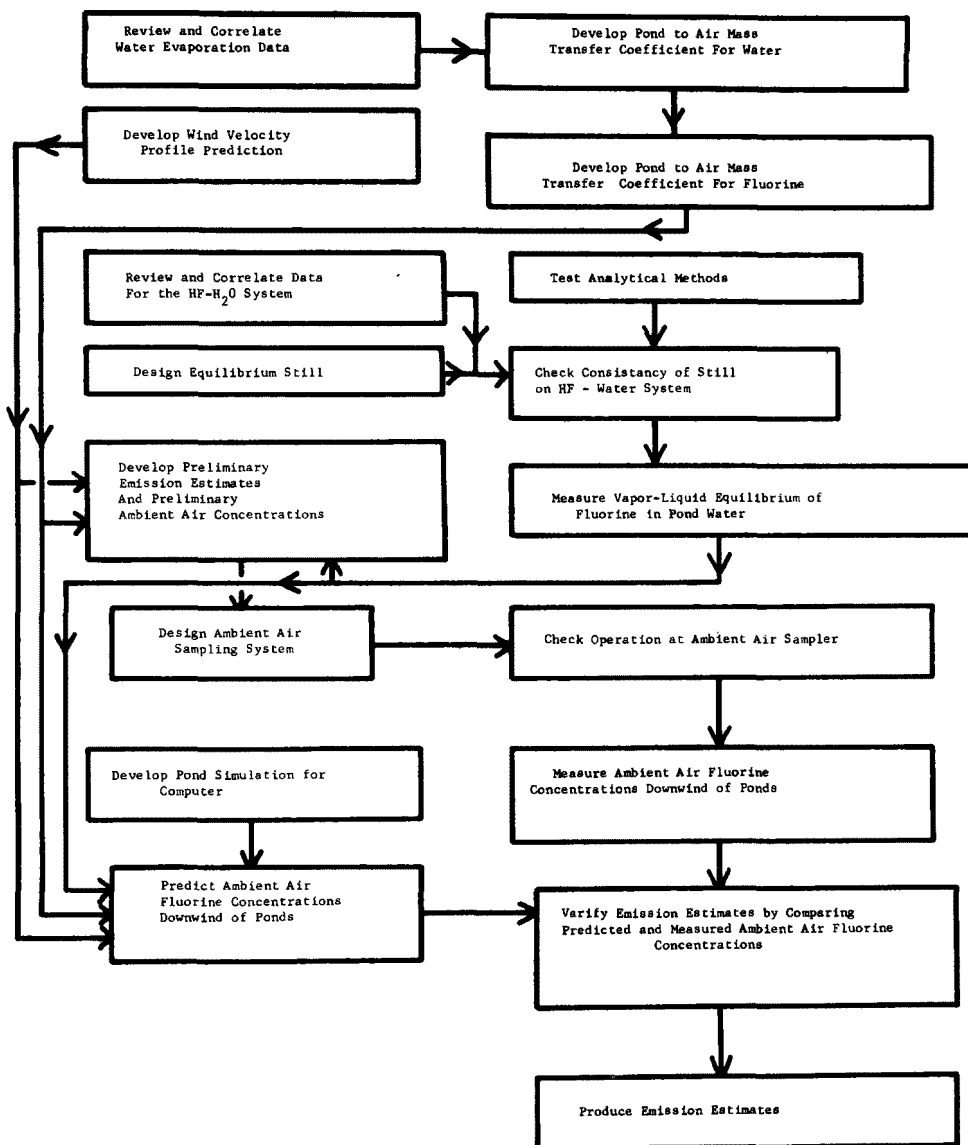


Figure 1 Work flow chart to produce an estimate of fluorine compound emissions from wet process phosphoric acid plant waste ponds

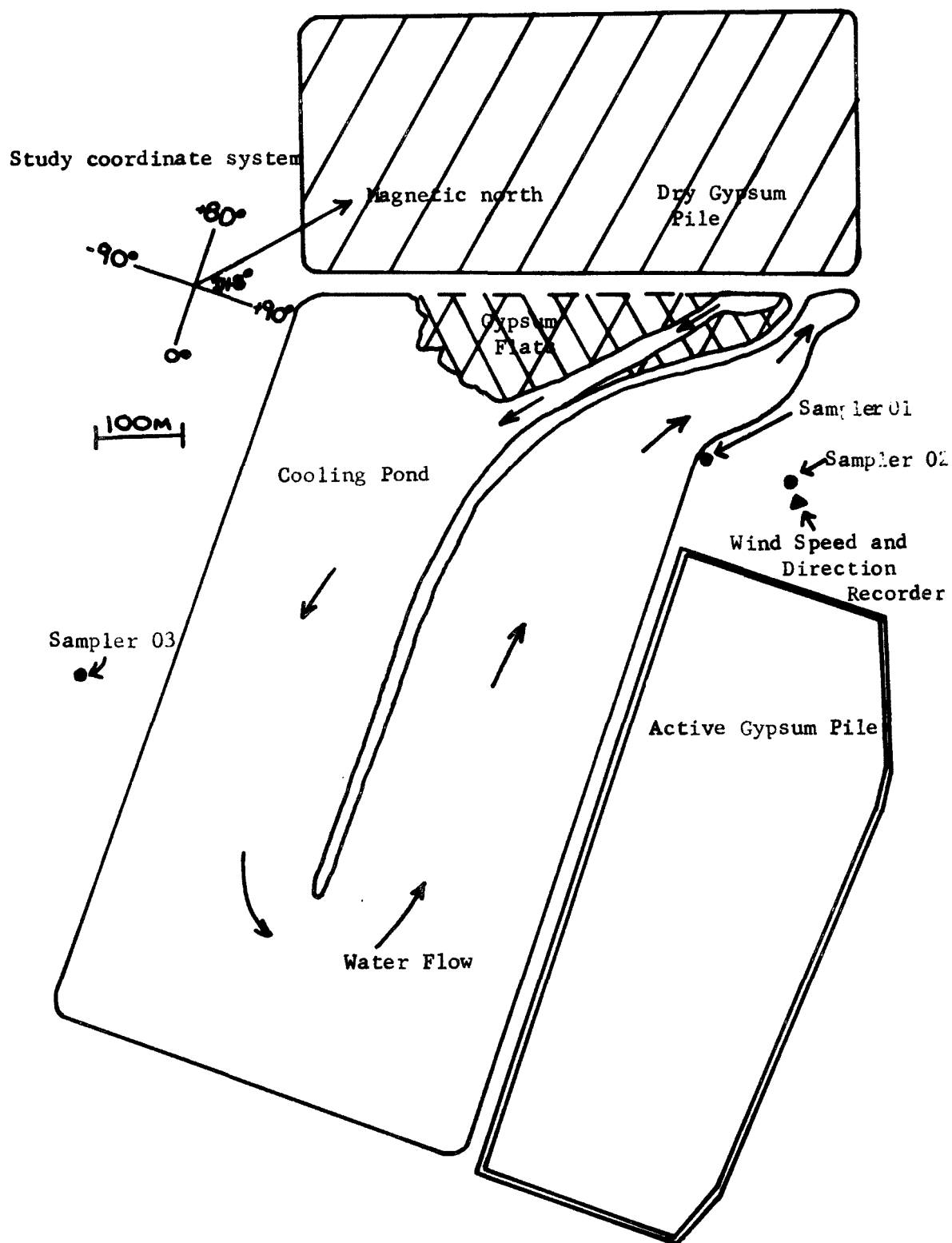


Figure 2 Pond 10 plot sketch

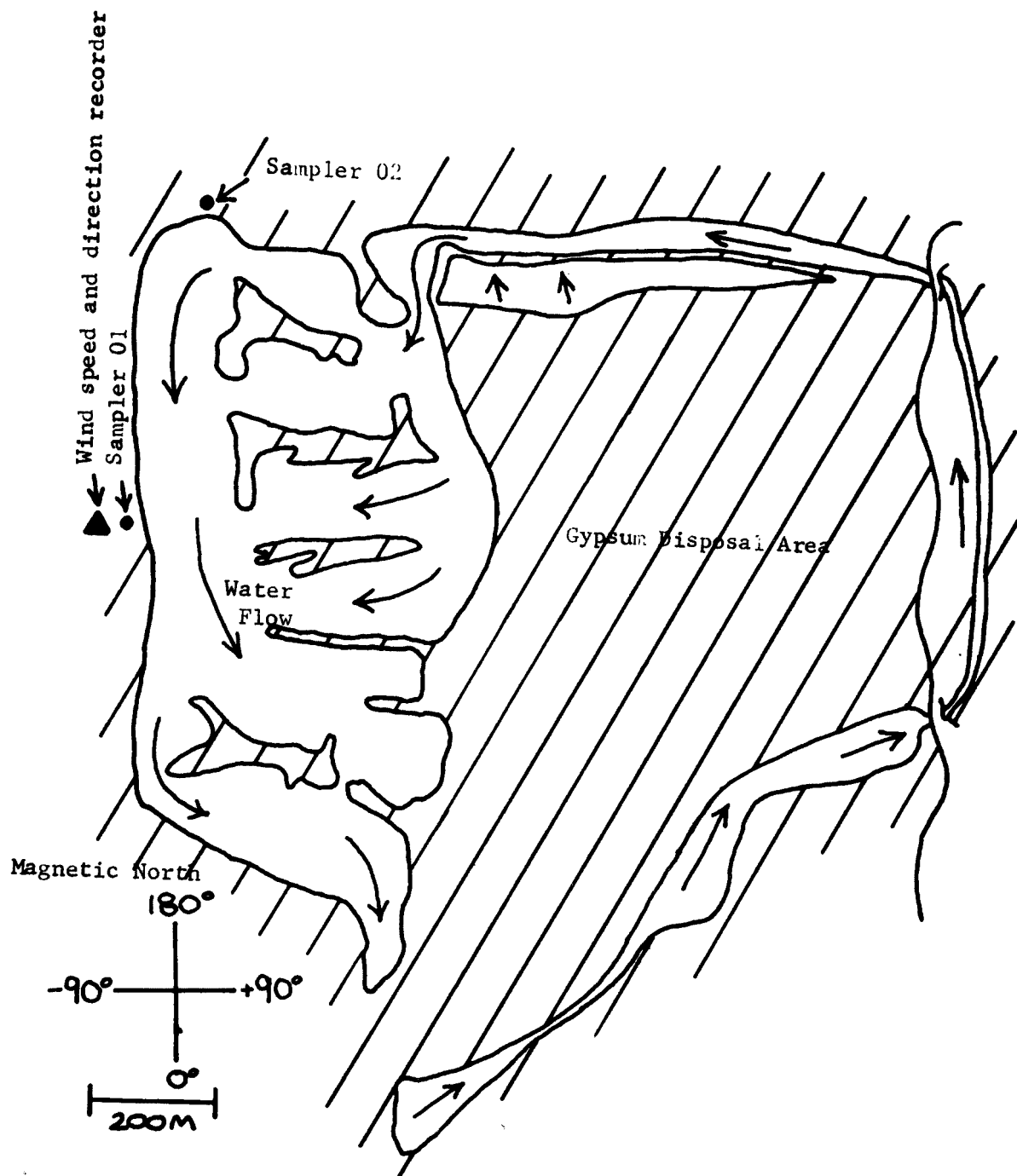


Figure 3 Pond 20 plot sketch

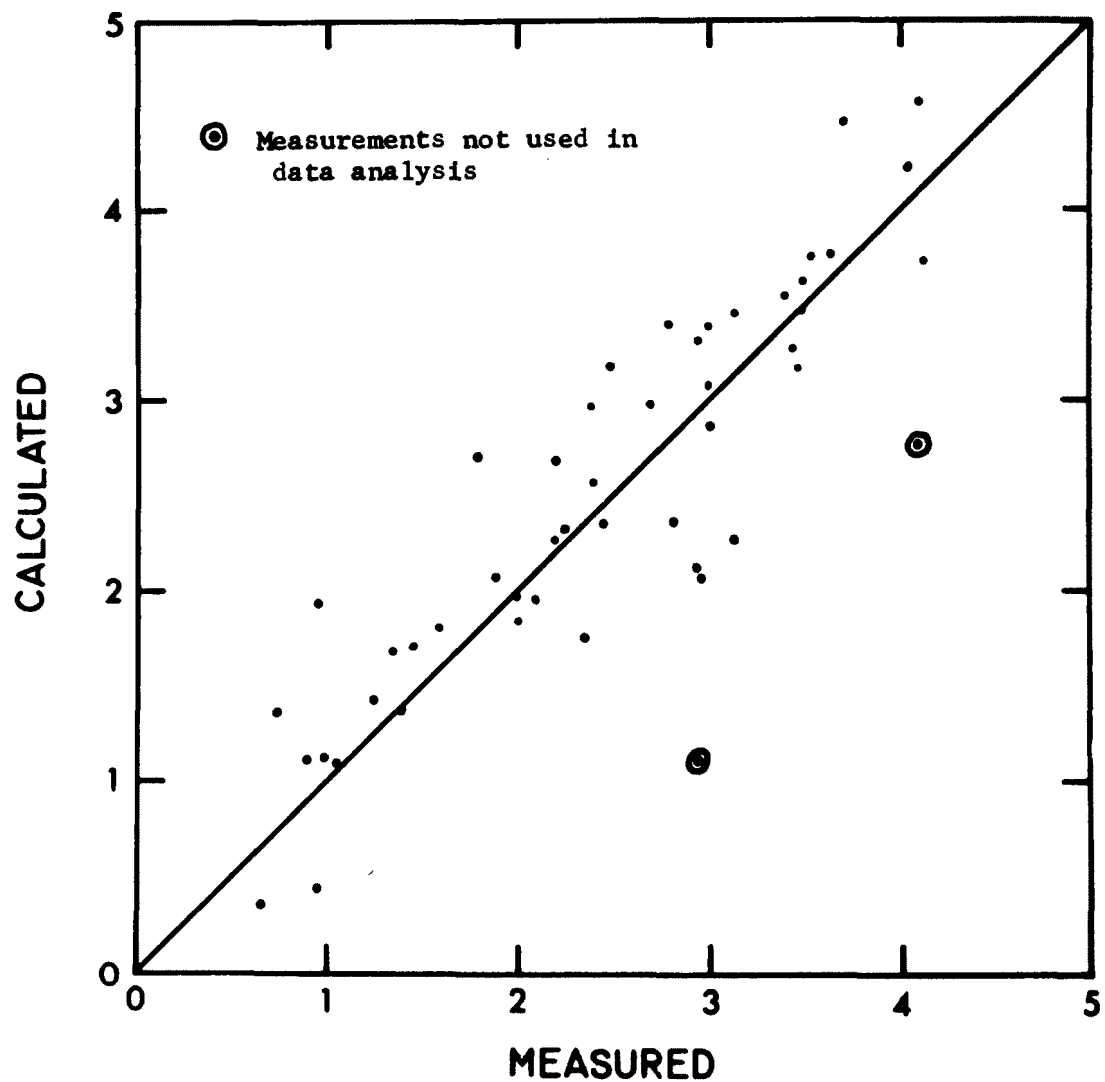


Figure 4 Pond 10 measured versus calculated ambient air concentrations at samplers $\times 10^{-6}$ g moles/ m^3

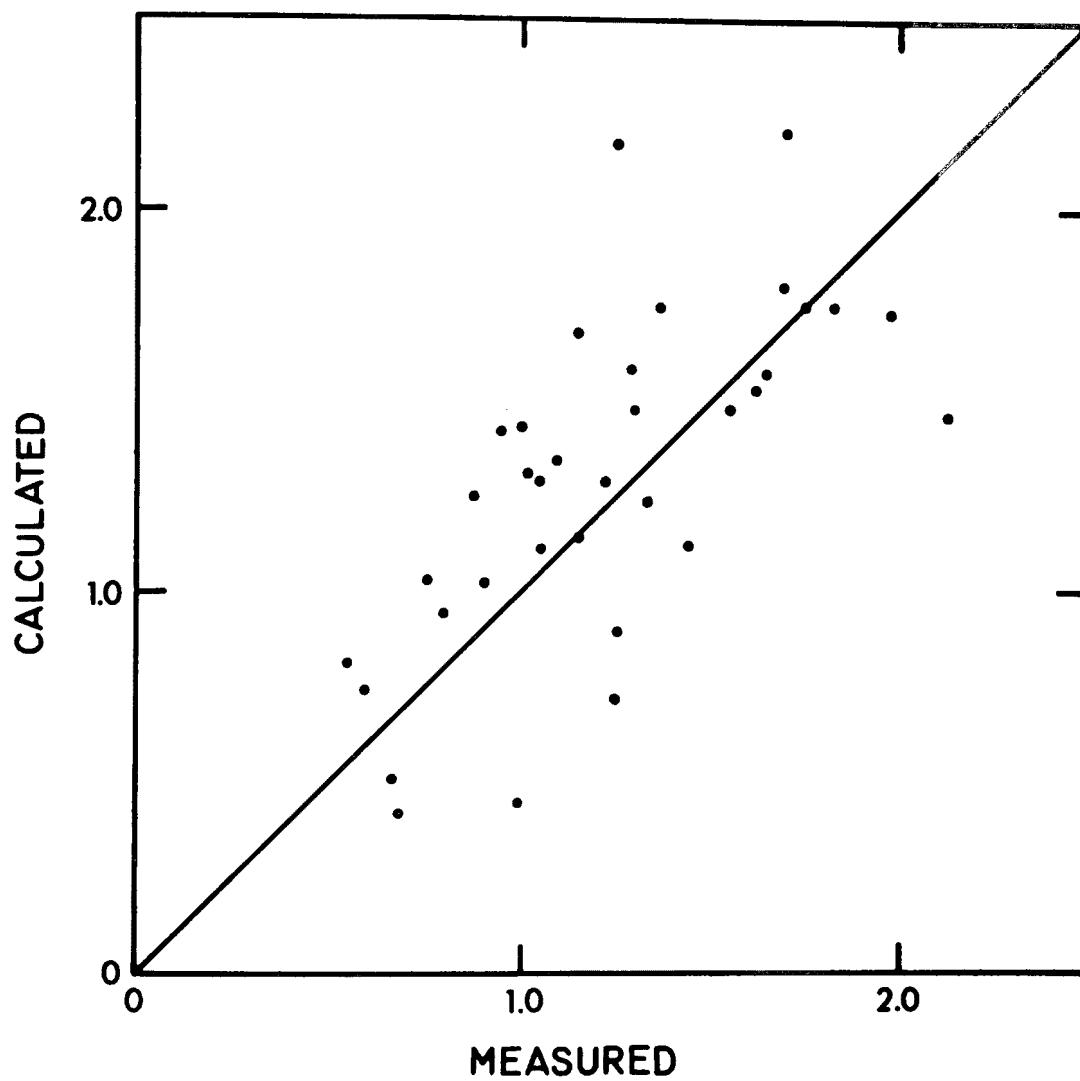


Figure 5 Pond 20 measured versus calculated ambient air concentrations at samplers $\times 10^{-6}$ g moles/ m^3

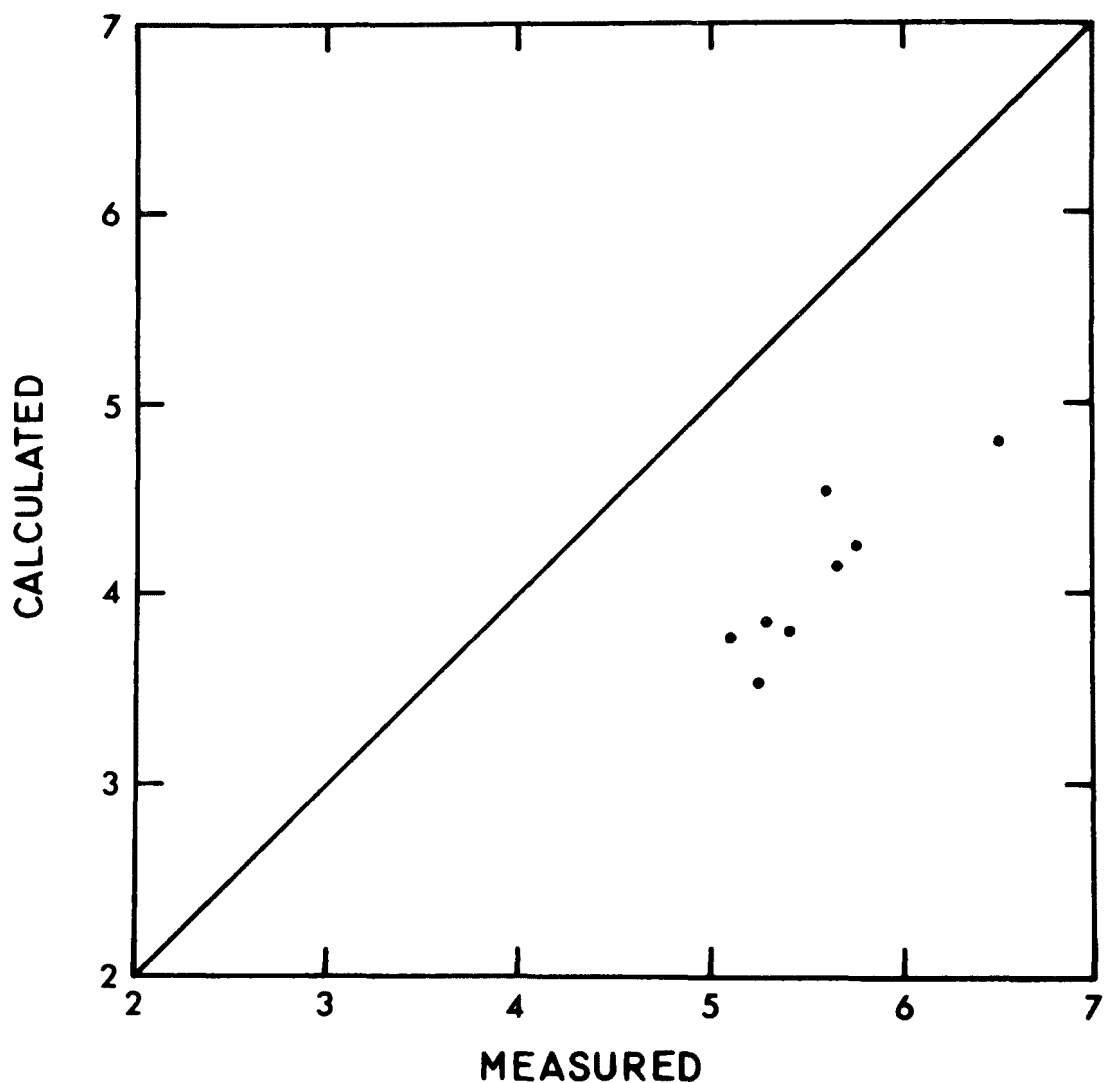


Figure 6 Pond 10 Sampler 01, high wind speed experiments
 $\times 10^{-6}$ g moles/ m^3

Concentrations measured by sampler 01, the sampler nearest the pond, when the wind velocity was over 5 meters/sec are considerably higher than the predicted values. Since measurements made with sampler 02 during the same period did not show this trend and no entrainment was noted, it is likely that the dispersion model cannot handle the combination of high wind speeds and short distances.

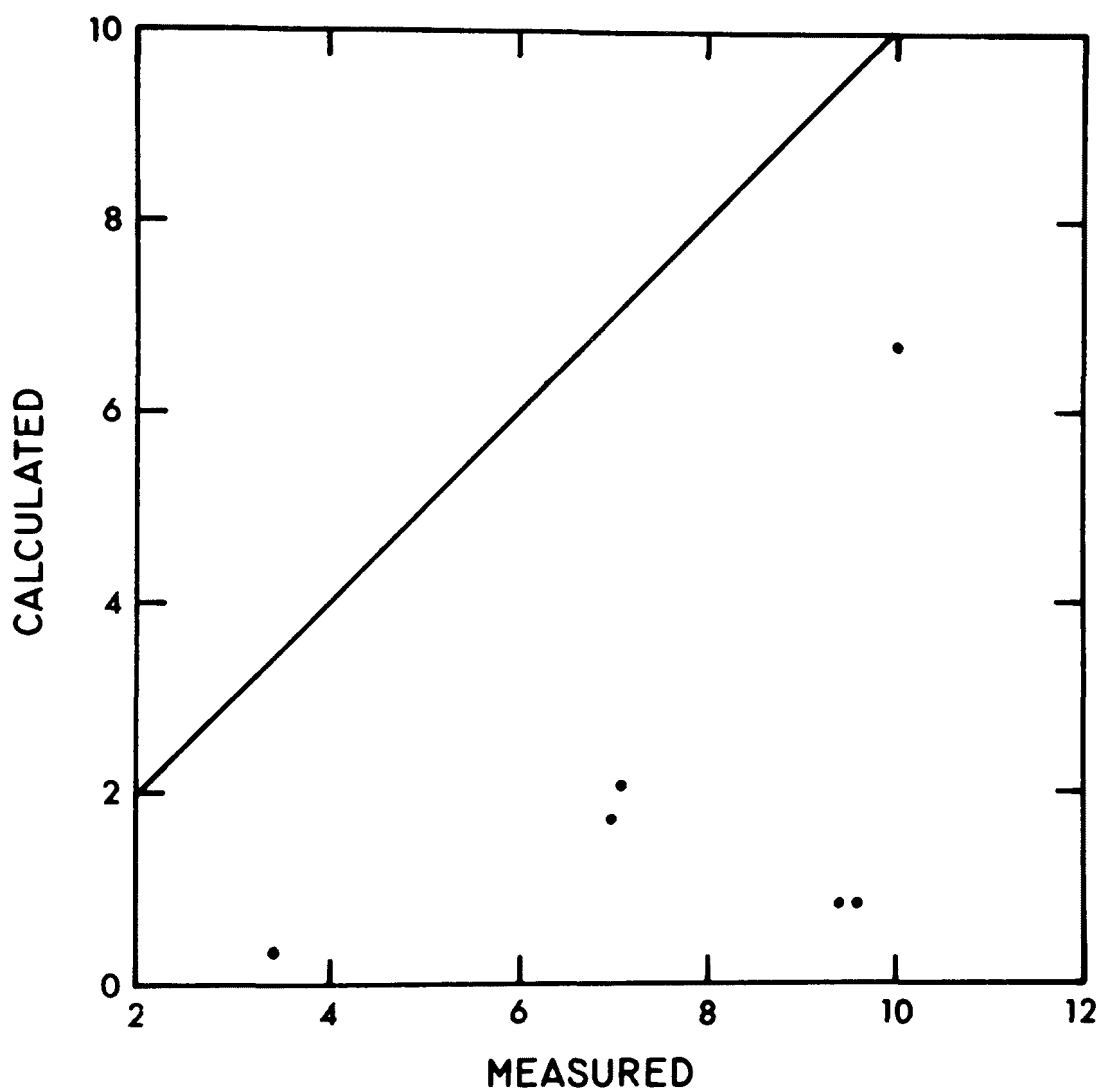


Figure 7 Pond 10 wind flow over active gypsum pile $\times 10^{-6}$ g moles/m³

Measurements were made when the wind was blowing from the active gypsum pile (wind direction 90° and over). The measured values were considerably greater than the predicted values. In this case, the piles caused extremely complex wind patterns - including downwashes that the dispersion model could not handle.

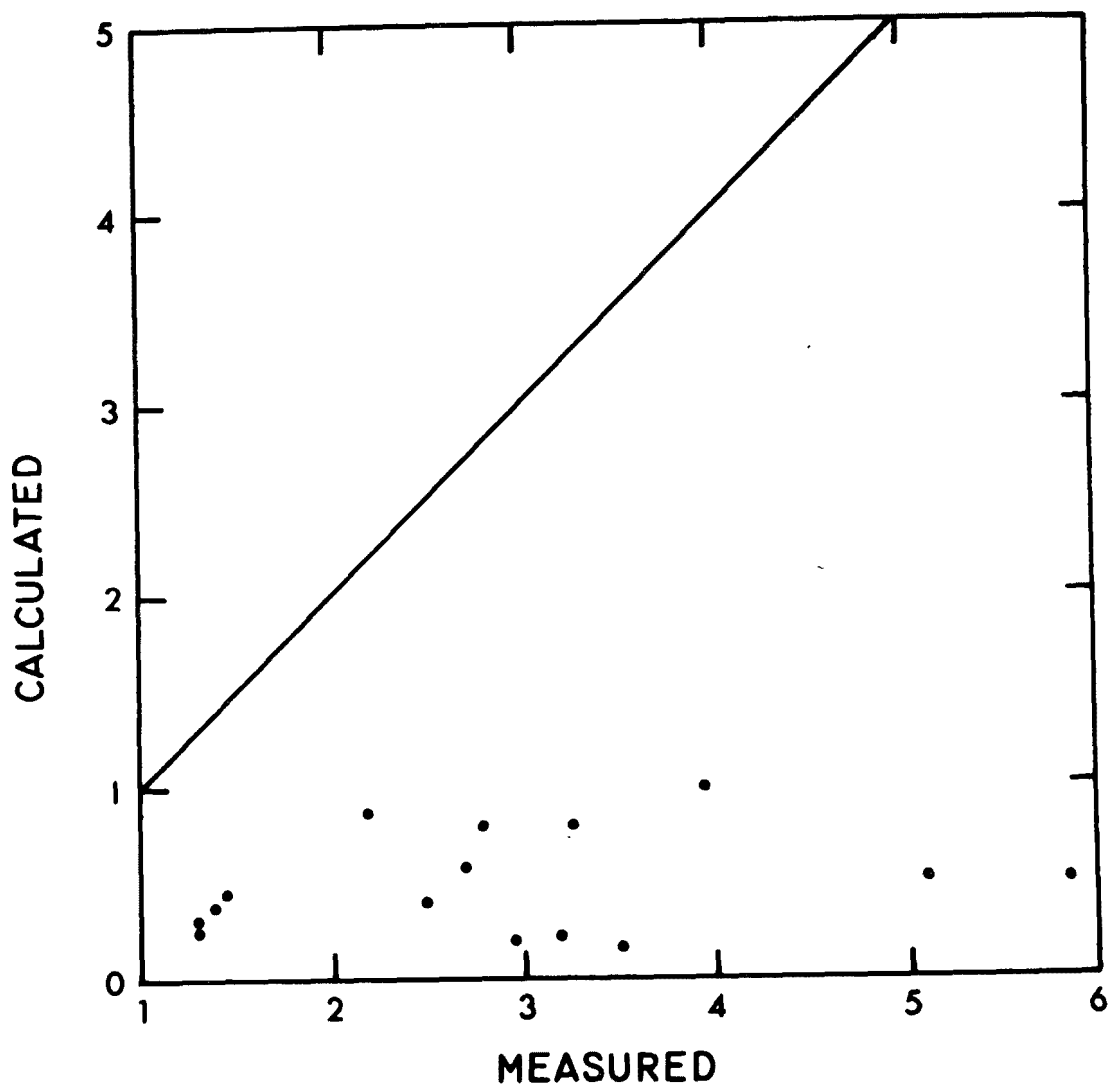


Figure 8 Pond 20 Sampler 02 high fluorine measurements $\times 10^{-6}$
g moles/m³

On a number of days after 10:00 AM, the fluorine concentration measured by sampler 02 increased dramatically. No similar increase was noted in sampler 01. Since the phenomena was not noted on the day after a plant shutdown, a tentative explanation is advanced that some action by the plant in the gypsum disposal area caused the high ambient air concentrations.

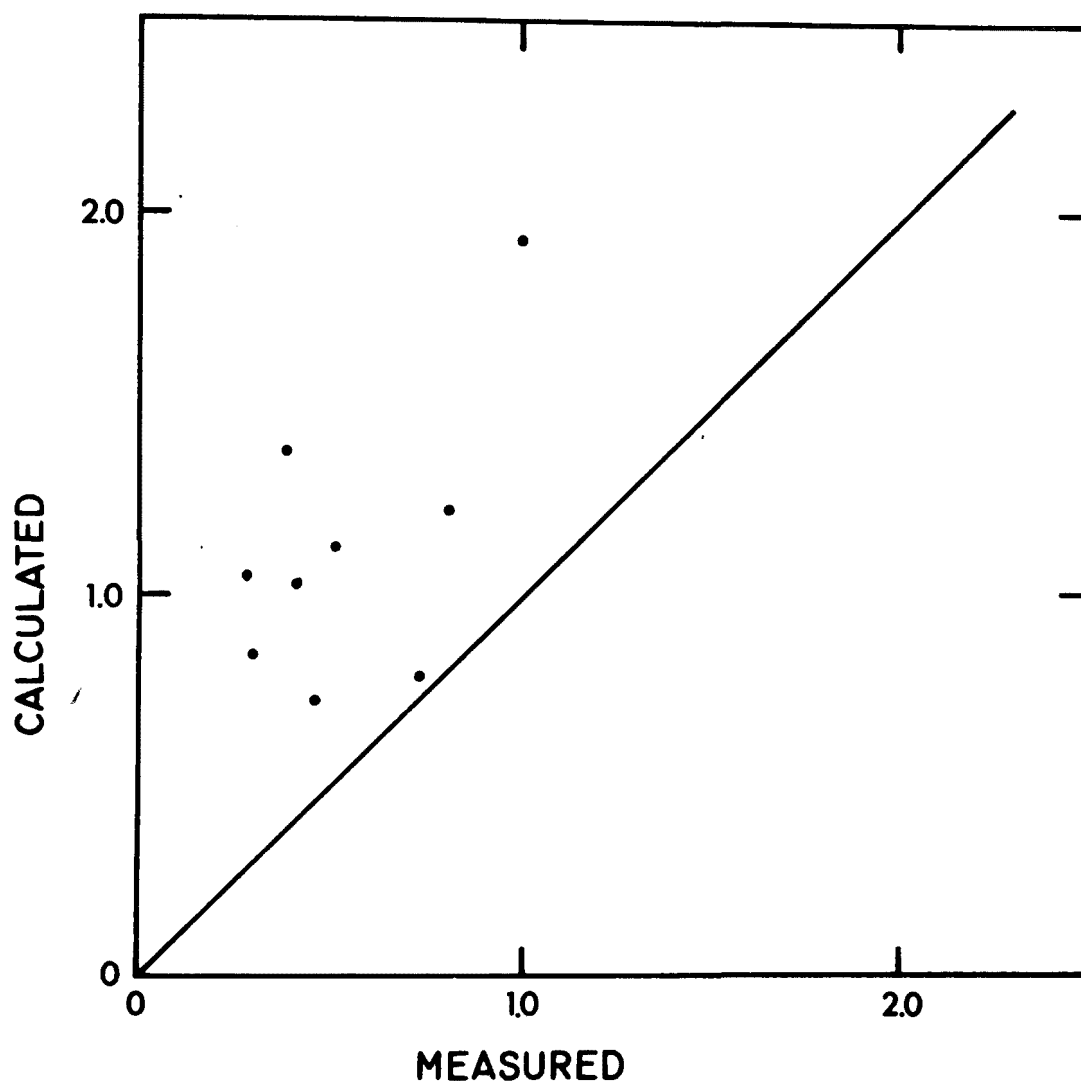


Figure 9 Pond 20 strong solar radiation, low wind speed experiments $\times 10^{-6}$ g moles/ m^3

A combination of low wind speeds (less than 1.3 meters/second) and strong solar radiation (late morning and afternoon) produced a greater degree of dispersion than could be predicted by the dispersion model. Low wind speeds alone (early and mid-morning) could be satisfactorily handled by the model.

CONTINUOUS ROOF MONITOR EMISSION TESTS

Abbas F. Souka, Ph. D., Airco Speer Carbon-Graphite

Single story buildings in certain industries such as metal smelting, graphitizing, silicon carbide manufacturing, glass plants, foundries, and many other manufacturing activities require a substantial amount of ventilation to maintain human comfort and safety. The most efficient and economical method to achieve this purpose is the use of wall louvers and building monitors capable of exhausting large amounts of heated gases at a very rapid rate. The quantities of influent air and effluent gases are dependent on the building design and location, the prevailing climatic conditions and on the activity within the building. The two last factors are variable and greatly influence the amounts of emissions from such structures.

In order to prove compliance with the air pollution regulations, most of the above mentioned industries are required to sample the effluent gases as they exit the building. At first, we at Airco Speer attempted to apply the standard EPA method for particulate sampling of stack effluents only to realize that such technique is not applicable and that an alternate approach is required.

First, I will enumerate the reasons why sampling using the standard EPA method for stack sampling is not applicable to monitor sampling:

1. The velocity of the effluent gases as measured in the building monitor fluctuates within a wide range. This can be readily seen by inspection of figures (1) and (2). They show seven plots of instantaneous monitor exit velocity as a function of time. Four of these plots were recorded on four consecutive days and the remaining three were recorded on the same day. It can be seen that within the course of one minute the exit velocity may fluctuate by more than 600%
2. Roof monitor designs cause turbulence in the gas stream and consequently do not fulfill the isokinetic sampling requirements that sampling and velocity measurement points must be taken at locations away from any disturbances.
3. The variation in activities within the building and the rapid changes in the climatic conditions make it necessary to collect a large sample of the effluent gas over an extended period of time in order to obtain results that are representative of the actual contribution of the manufacturing facility to the ambient air.
4. Pitot tubes are not suitable for measurements of low velocity heads.

Sampling Procedure

The first published¹ application of high volume samplers to sample emissions from roof monitors that we are aware of used suspended high volume samplers which traversed the building monitor, figure (3). Samplers were positioned along the length of the building; each to collect a sample representative of the emission through a known area. Each sampler in each section was hung on a cable so that it can be traversed back and forth across the width of the monitor. The effluent velocity was measured using hot wire anemometers.

The procedure² which has been applied in three separate tests by Airco Speer for measurement of emissions from building monitors also included the use of high volume samplers as described below:

The building monitor was divided into three equal sections. One sampling station equipped with a high volume sampler was located at the center of each section. Figure (4) shows the 3 sampling stations and fig. (5) shows the high volume sampler affixed in its normal position with the filter facing upwards. A goose neck attachment directs the flow into the filter. The sampling velocity was maintained at about 100 fpm; the normal sampling rate used with high volume samplers. This rate is less than the isokinetic velocity and gives emission values biased to values higher than the actual values. Figure (6) shows the deviation between the observed concentration to the actual concentration with departure from isokinetic sampling.³ It is noted that the smaller the particle size the less the error in measurement becomes. Figure (7) shows the sampling probes which consisted of three - 9" diameter nozzles each connected to a U-shaped 4" diameter pipe. Figure (8) shows one of the high volume samplers mounted on a platform outside the monitor. Particulates were collected on glass filter paper retentive to particles larger than 0.3 micron. Before initial and final weighing the filters were dried at room temperature over silica-gel for 24 hours. Weighing was made to the nearest 0.1 mg with a balance having a precision of $\pm .05$ mg.

The volume of air sampled was determined by using calibrated rotameters. Flow meters readings were generally obtained at least once an hour. In order to obtain a sampling velocity between 100-120 fpm, a sampling rate of about $1.4 \text{ m}^3/\text{min}$. was necessary. By observing the rate of decrease in flow meters readings an estimate as to the necessity of a filter change could be made. Generally, the filters were changed when the flow meter indicated a rate of $1.3 \text{ m}^3/\text{min}$.

The average velocity of the air as it exits the monitor was continuously recorded. A Gill Propeller Electric Anemometer Model 27100 was mounted in the monitor at each of the sampling stations to a depth of about 12 in. Figure (9) shows the anemometer mounted in the monitor. The anemometer⁴ is an extremely sensitive air speed measuring instrument employing a foamed polystyrene propeller. The propeller rotates 0.96/revolution for each foot of passing wind for all wind speeds about 4 ft./sec. Its threshold speed is 0.8'/sec.

The propeller drives a miniature D. C. tachometer generator. The propeller anemometer will measure both forward and reverse flow. When the propeller rotation reverses, the generator signal polarity reverses. The propeller responds only to the component of the wind which is parallel with its axis.

The anemometer was purged with filtered air in order to prevent dust or corrosive gases from interfering with the proper function of the instrument. Calibration of the anemometer was done by connecting the anemometer to a synchronous drive with flexible coupling. At 1800 r.p.m. the D. C. generator generates 500 m.v.

The signals from each anemometer, which are directly proportional to wind speed, were recorded in the first two tests by a Leeds and Northrup Speedomax W recorder which printed a reading from each anemometer every 75 seconds. This gave an extensive history of the monitor exit velocity. Approximately 24,000 velocity readings were averaged. Interpretation of the data proved to be a tedious and time consuming operation and prompted us to look for an alternative recording instrument. Subsequently, we purchased a Martek Model EDP Environmental Data Printer, which was used instead of the L&N recorder and which greatly simplified the data logging.

The data printer can be operated in one of three modes; continuous, program or manual mode. In the program mode record, period and length are switch selected. Record period may be set at 4, 8, 16, 32, 64 or 128 minutes. Record length determines the duration of each recording and may be set to either scan or 1/2 to 2 minutes position, recording will start by printing the time and continue for the time selected. After the last active channel has been recorded the instrument reverts to a standby condition and all assemblies except the clock are turned off to reduce the power drain. In the manual mode each channel is advanced by depressing the step switch.

The data collected by the data printer is in the form of a printed tape and a recording on a magnetic tape cassette. The cassette can be read on a Data Reader. A digital printout of the taped information can also be acquired through external data processors.

Emission Data

The testing period for evaluating particulate emissions from our graphitizing plant roof monitor was continuous for one week. Sampling was continuous with the exception of time required to make filter changes and short intervals to make minor equipment adjustment and repair.

The emission rate was calculated as indicated below:

1.
$$\frac{\text{Dust load/filter (mg/m}^3\text{)}}{\text{total volume sampled}} = \text{Weight of particulates collected (mg)/}$$

2. Average monitor velocity (ft/min). Velocity values were recorded by means of L&N Speedomax type W recorder in the first two tests and by a Martek Model EDP Environmental Data Printer in the third test. These values were averaged over the time interval corresponding to the filter paper used.
3. Station emission rate (#/hr) = Station fractional monitor area (ft²)
x average vel. (fpm) x dust load (mg/m³) x 2.2 (#/kg) x 60 min/hr
x 10⁻⁶ (kg/mg)x .0283 (m³/ft³).

Figure (10) shows emission rate vs. time at one of the three sampling stations. Studying of the bar chart shows periods of higher emissions and other periods when emission rates are quite small. These fluctuations reflect activity within the building and can serve as a guide to point out the dusty operations so that corrective measures may be taken.

Safety

Top consideration has to be given to prevention of injury to the sampling team. Installation of railings for high areas is a must. Sampling for possible existence of harmful concentrations of gases must be performed by the industrial hygienist. Sampling personnel must work in pairs. The installation of a telephone for easy communication with the rest of the plant is recommended.

Sampling Cost

The cost of performing this sampling technique can be divided into 3 portions:

1. Cost of Equipment

This is a one time expense which amounts to approximately \$4,000.00 to cover the cost of 3 High Volume Samplers and one Data Logger.

2. Cost of Sampling Station

This is a cost which is incurred every time a new monitor is sampled. It covers the cost of setting up the shelters for the high volume samplers as well as the Central station where the recording equipment is situated. This cost will vary depending upon the monitor being sampled. An average figure of \$10,000 represents a realistic figure.

3. Cost of Conducting Test

This represents the cost of data collection, performing the

necessary calculations and writing the final report. This cost amounts to about \$8,000.

EPA Method for Sampling of Emissions from Building Monitors

A sampling method⁵ for measuring emissions from primary aluminum industry building monitors has been promulgated by the EPA (reference methods 14 and 13A or 13B). In this method a manifold system and connecting duct is permanently installed to draw an air sample from a roof monitor to ground level, figure (11). The system includes eight nozzles each having a diameter of 0.40 to 0.5 meters spaced along the length of the manifold which measures 8% of the monitor length with a minimum of 35 meters. The exhaust fan which is installed at the ground level draws a portion of the exhaust at a rate equal to the isokinetic rate determined by means of electric anemometers installed in the monitor. The monitor exit velocity may vary during sampling as much as $\pm 20\%$ of the previous 24 hours average isokinetic velocity as recorded by the electric anemometers. The sampling duration for fluorides has been set at a minimum of four to eight hours.

Comparison of Monitor Sampling Methods

The first two sampling methods discussed in this presentation follow the approach which uses high volume samplers to filter a large sample of effluent gases over an extended time period in order to determine the particulate concentration which fluctuates with the activity inside the building. This presentation also acknowledges that isokinetic sampling as defined and applied to stack sampling is not applicable to monitor sampling.

The EPA methods 14, 13A or 13B promulgated for the Primary Aluminum Industry can not necessarily be extended to other industries. The limitation which require the average monitor velocity, misnomered isokinetic velocity, on the day of sampling to be within $\pm 20\%$ of the average monitor velocity recorded in the previous 24 hours is very restrictive. Disregarding the changes in work activity within the buildings which, by itself, may result in variation in the internal heat load to a degree producing change in the monitor exit velocity approaching the $\pm 20\%$ allowable range, variations in wind velocity by as little as 2 mph can produce as much as 20% change in the monitor exit velocity.

Isokinetic Sampling

A method has been suggested² in which an approximation of isokinetic sampling may be realized. In this procedure, reproduced hereafter, two probes are equipped with a Model 27100 Gill Electric Propeller Anemometer or equivalent, figure (12). Each anemometer generates a direct current proportional to the speed of rotation of the propeller. The electric

signal produced by virtue of rotation of the comparator anemometer represents the velocity of the effluent gases while that of the sampling anemometer represents the sampling velocity. The net signal is fed into a controller which actuates a motorized butterfly valve towards close or open positions till the flows through the comparator and sampling probes match. Alternatively this net signal may be fed to a control circuit designed to adjust automatically the speed of rotation of the high volume sampler till an isokinetic sampling rate is achieved.

When such a system is used for sampling emissions from building monitors, a more representative sample may be obtained if a sampling chamber which continuously travels from one end of the monitor to the other end is substituted for the stationary sampling stations, figure (13).

Acknowledgment

This work would not have been possible without the participation and valuable contribution of my colleagues at Airco Speer.

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2. A New Approach to Roof Monitor Particulate Sampling
A. Souka, R. Marek & L. Gnan
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3. H. Watson, Amer. Ind.
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Gill Anemometer Brochure
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Performance Standards for New Stationary Sources
January 26, 1976



Fig. (1) Variation of Monitor Exit Velocity With Time

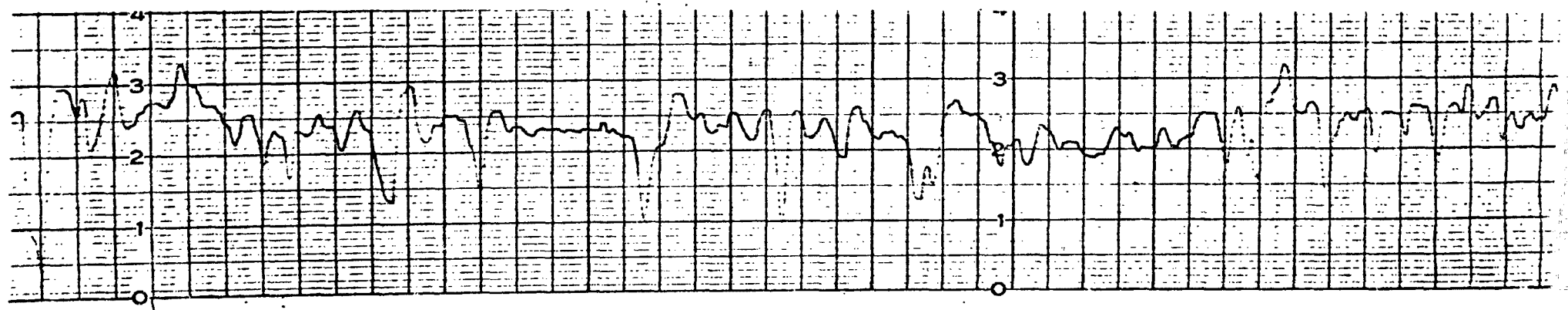
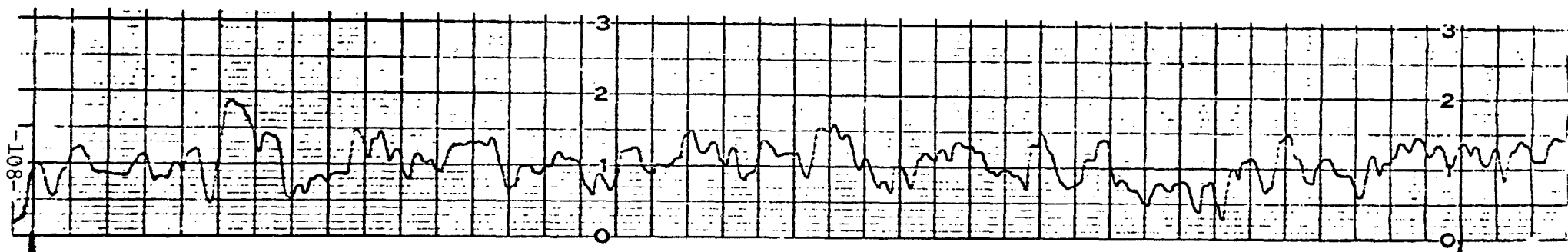
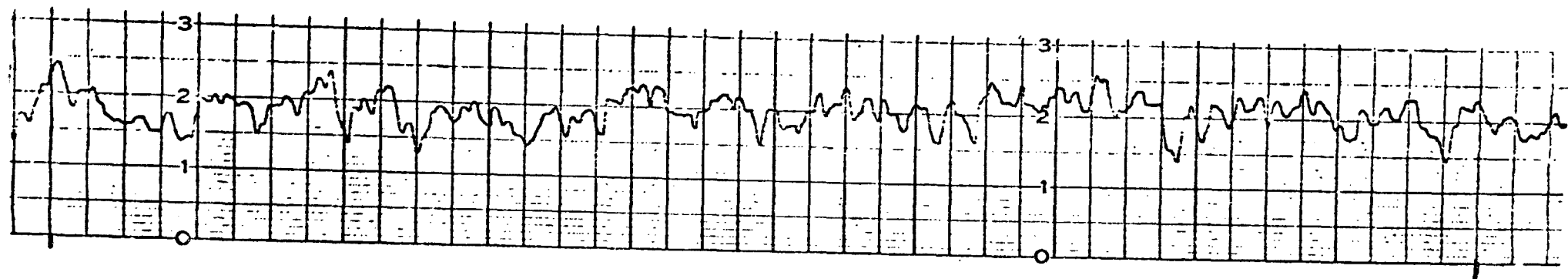
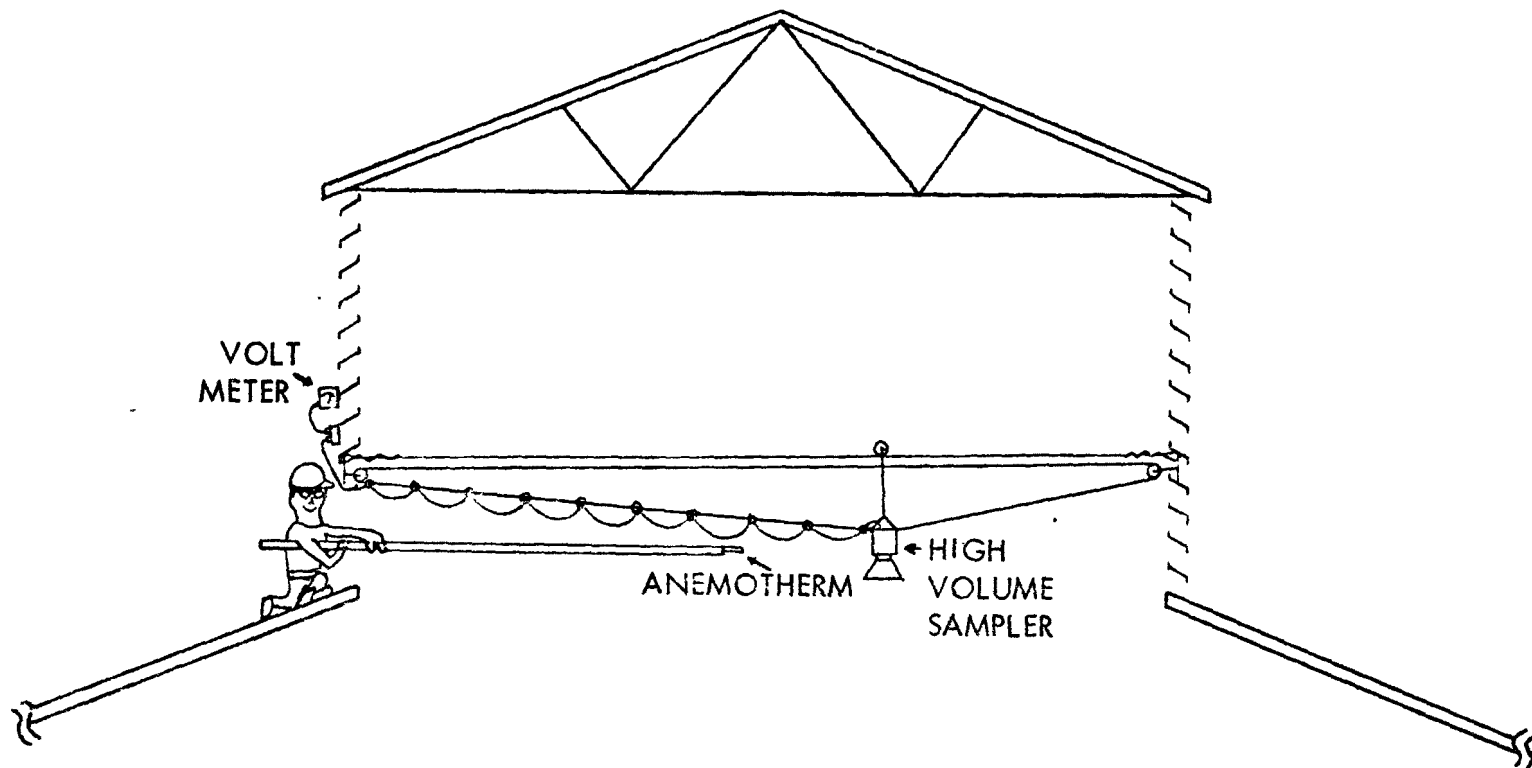


Fig. (2) Variation of Monitor Exit Velocity With Time



FIGURE(3) CROSS SECTION OF ROOF MONITOR SHOWING TEST EQUIPMENT (Ref. 1)

Figure 4
Sampling Stations

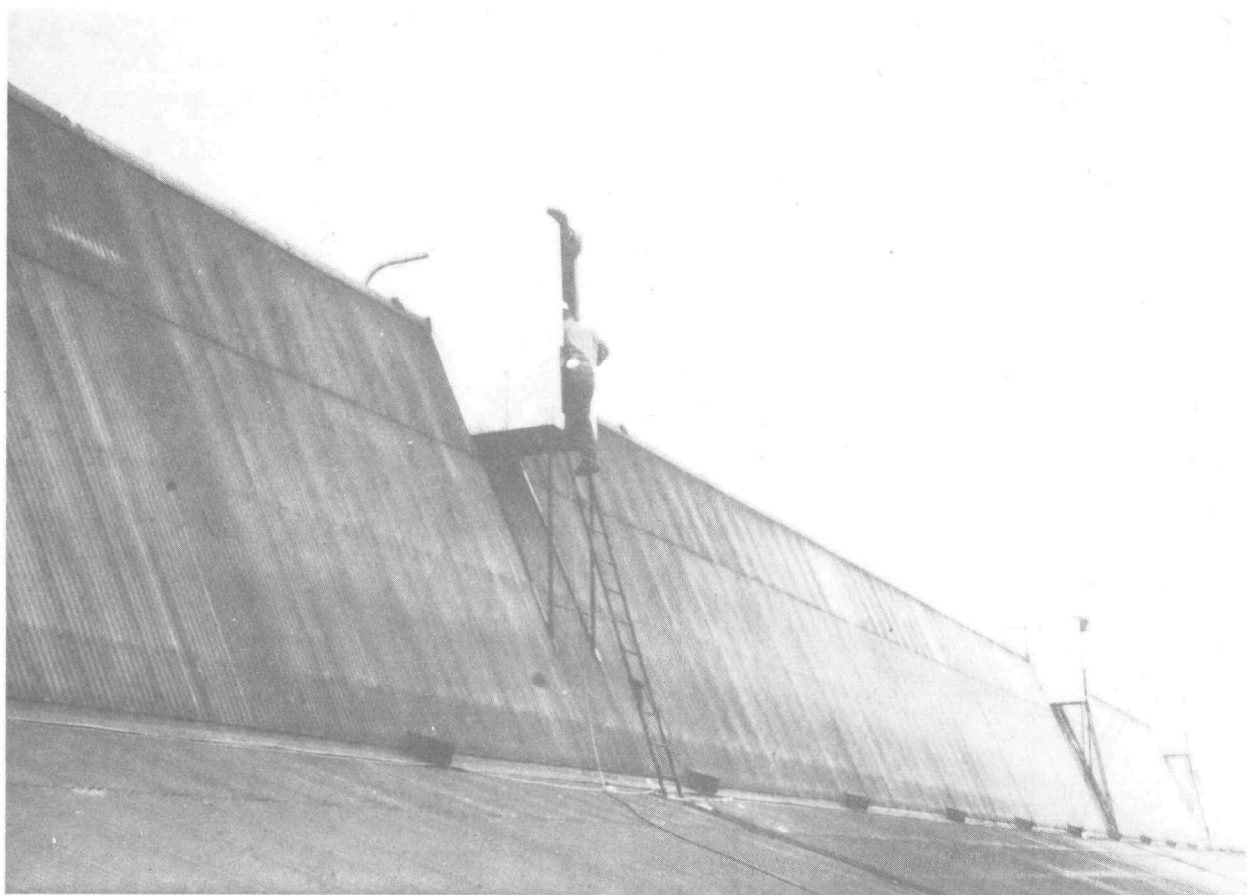


Figure 5
Volume Sampler



Figure 7
Sampling Probe

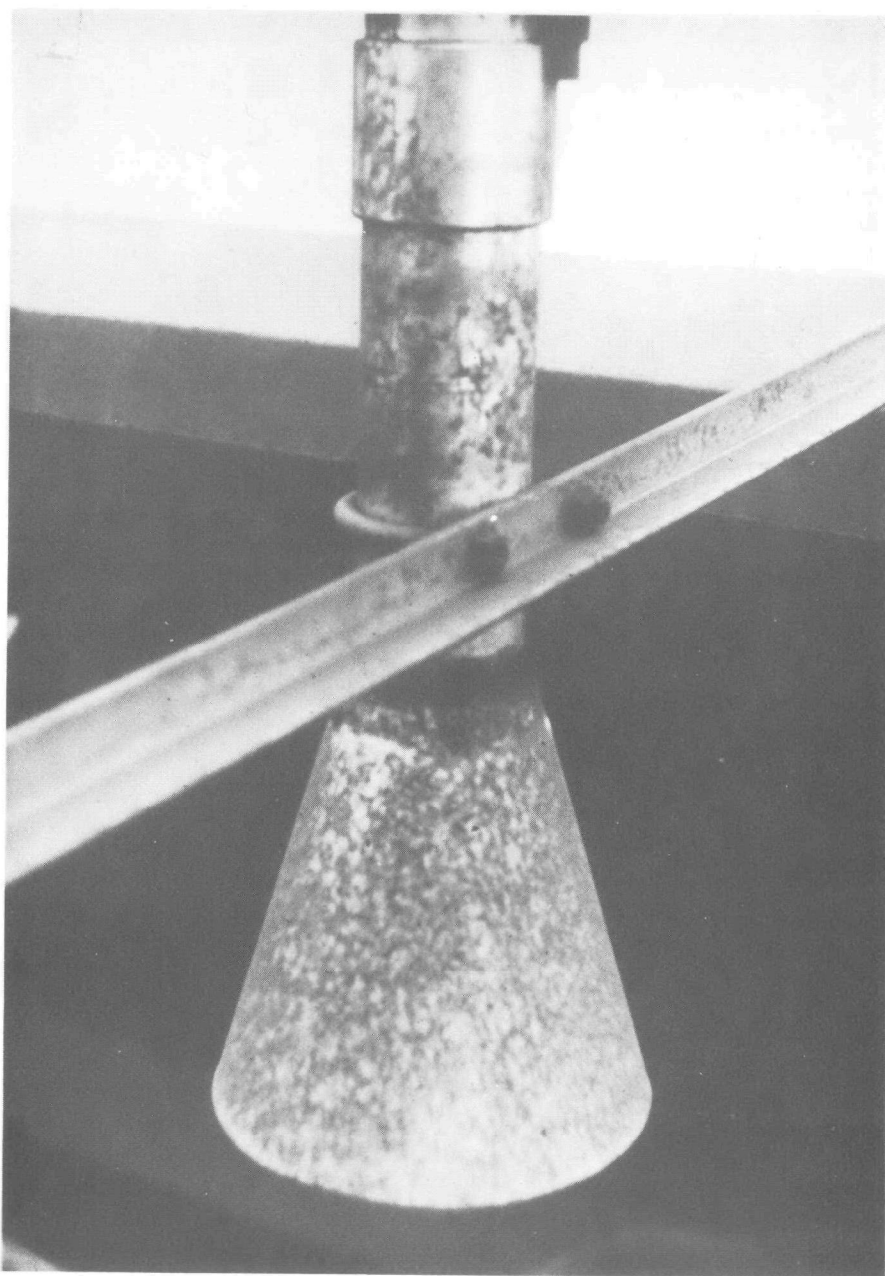
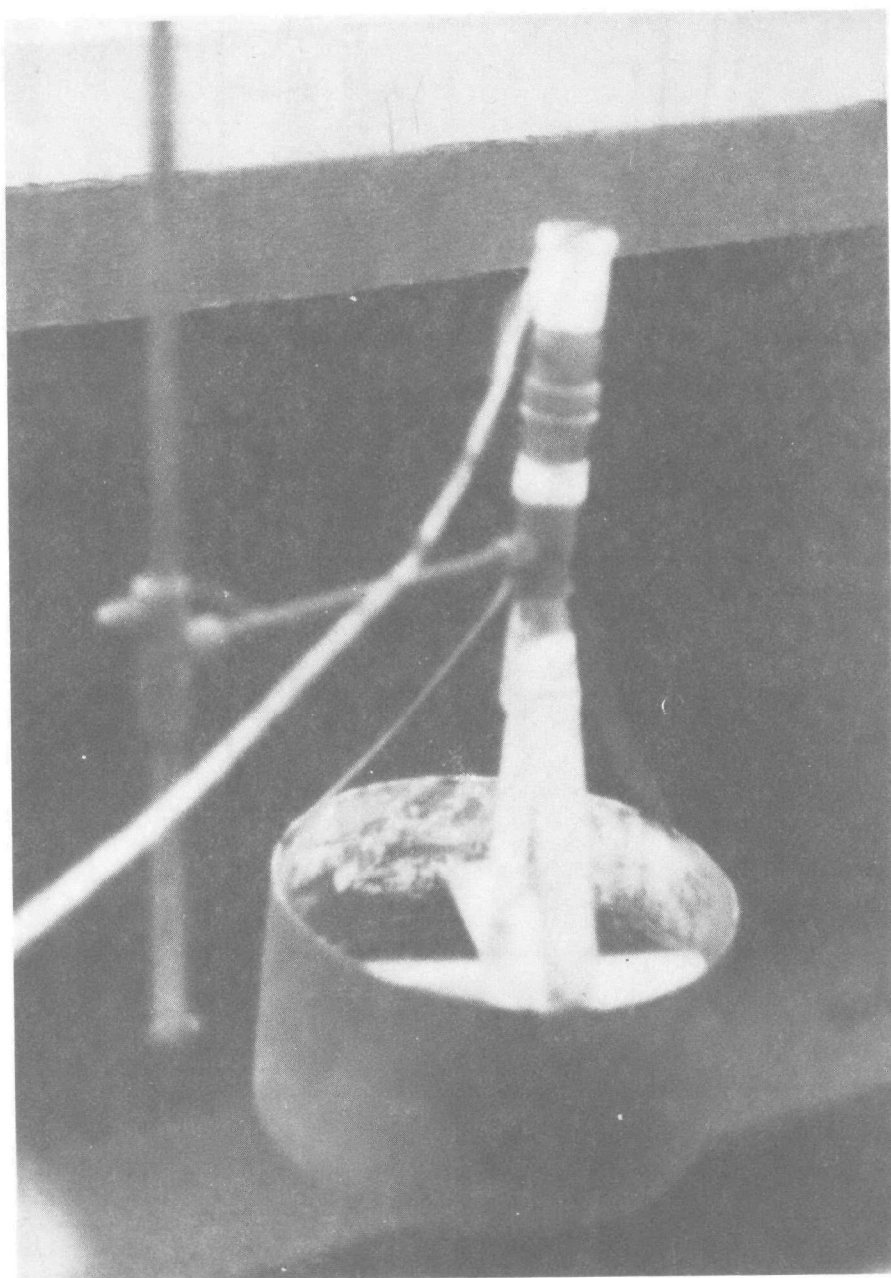


Figure 9

Elec. Anemometer



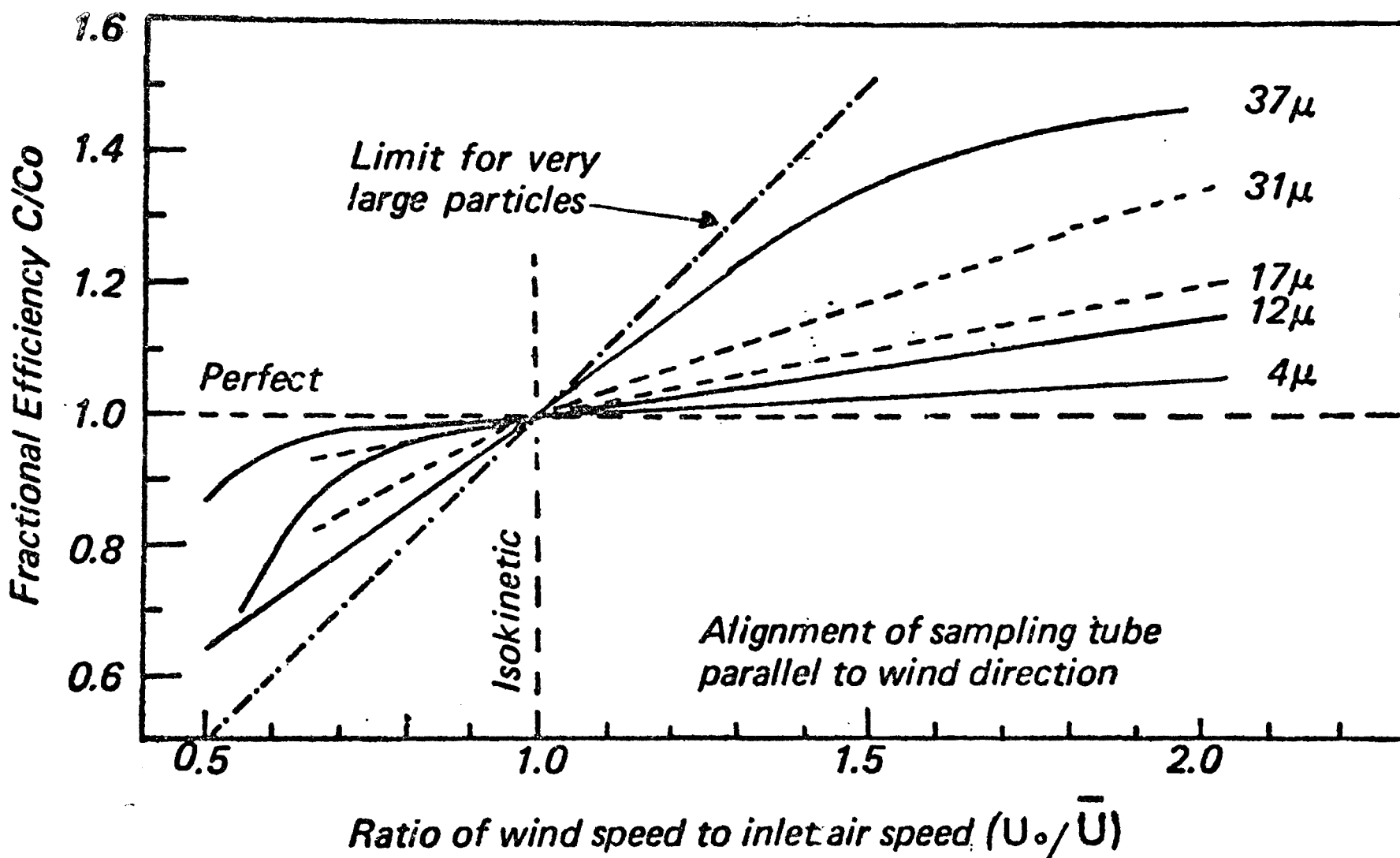


Figure (6)—Diagram showing change in ratio of observed concentration to true concentration with departure from isokinetic conditions. (Ref. 3)

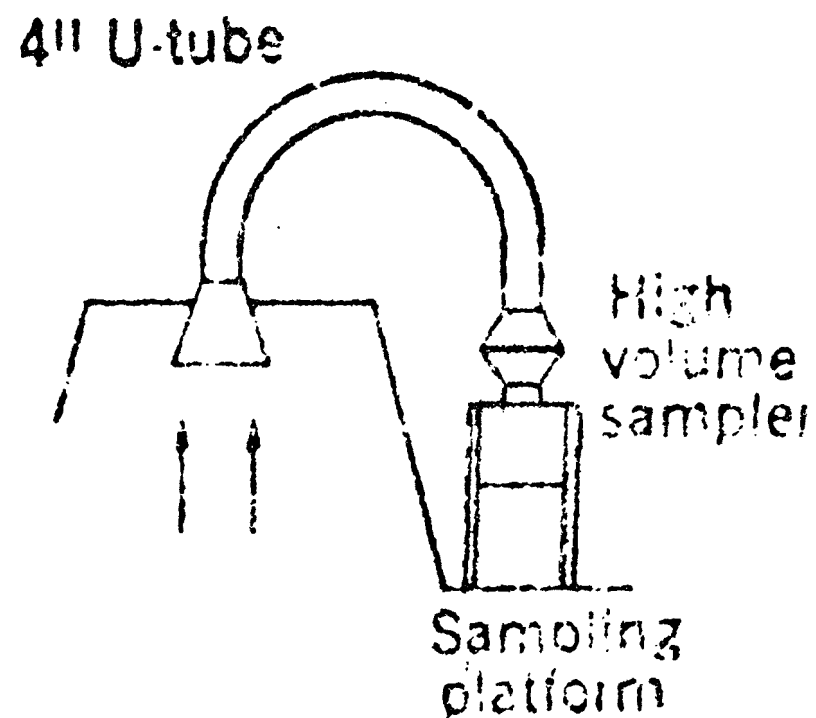
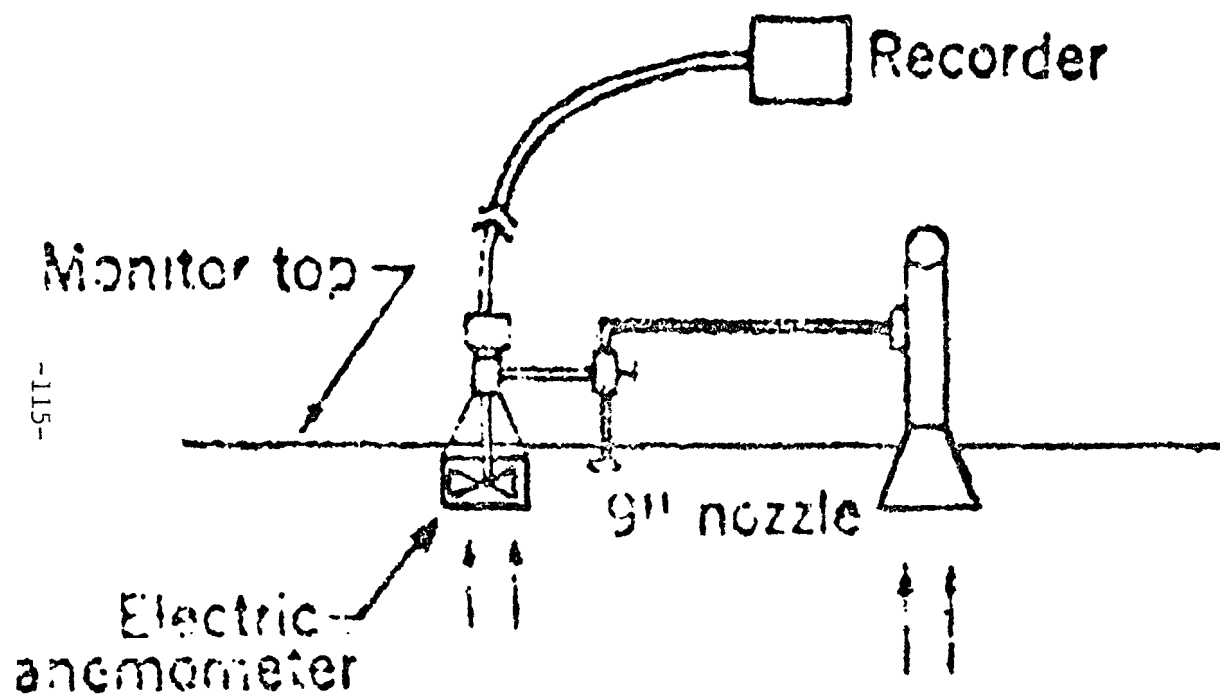
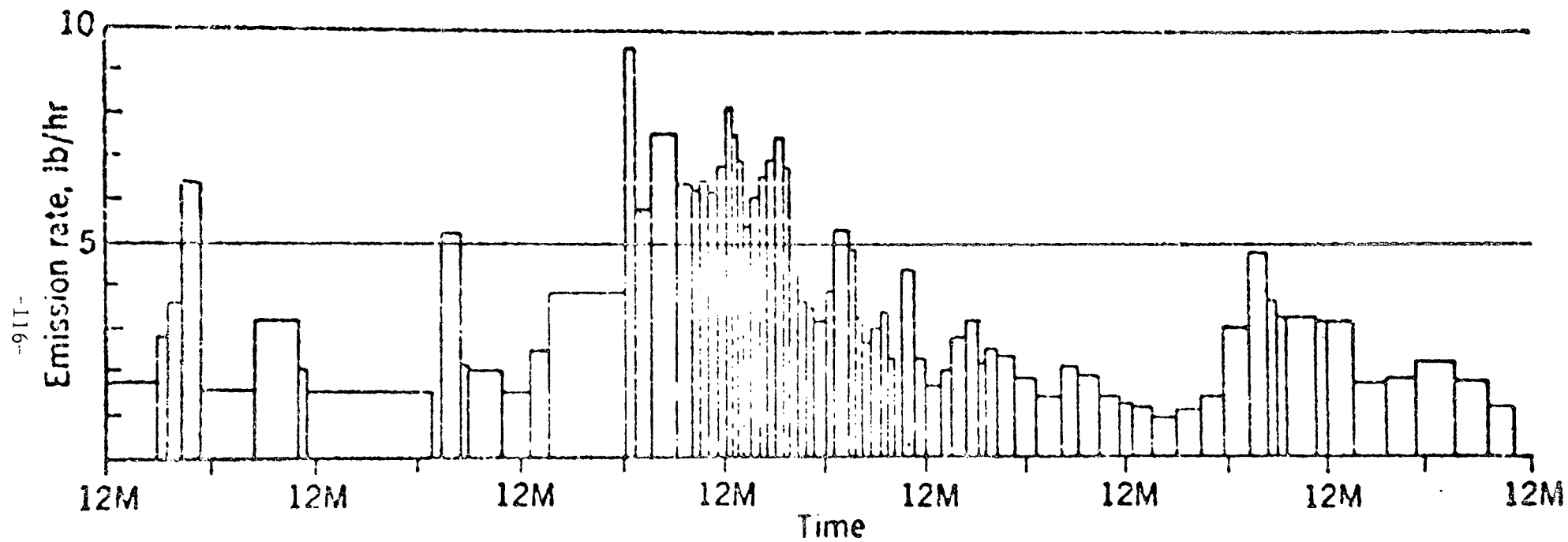


Figure (8) Sampler system.



Figure(10) Emission rate vs. time interval, Central Station.

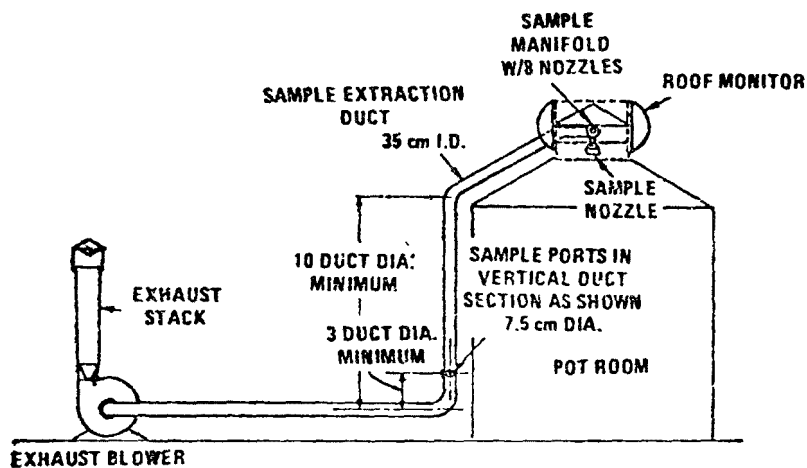


Fig. (11-A) Roof Monitor Sampling System.

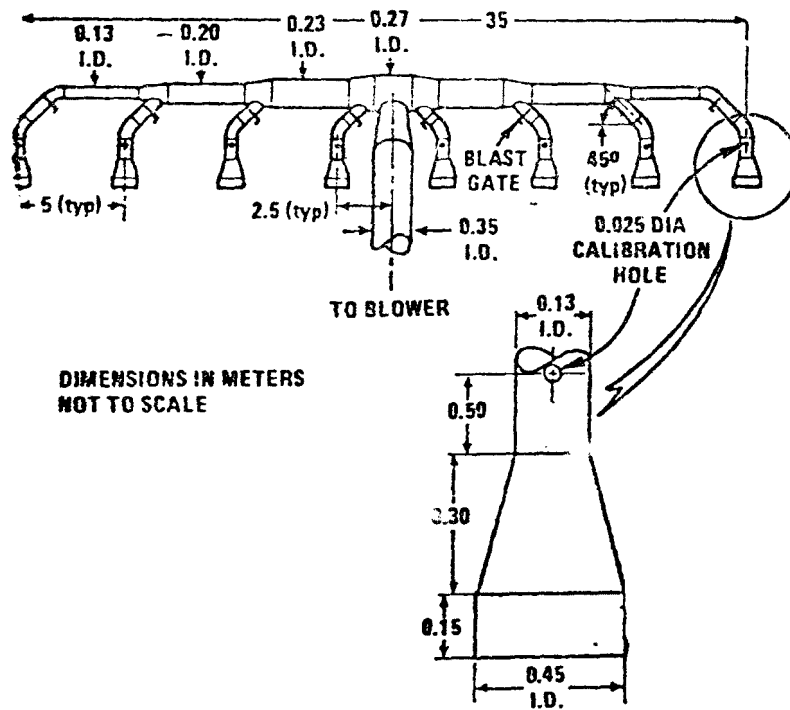


Fig. (11-B) Sampling Manifold and Nozzles.

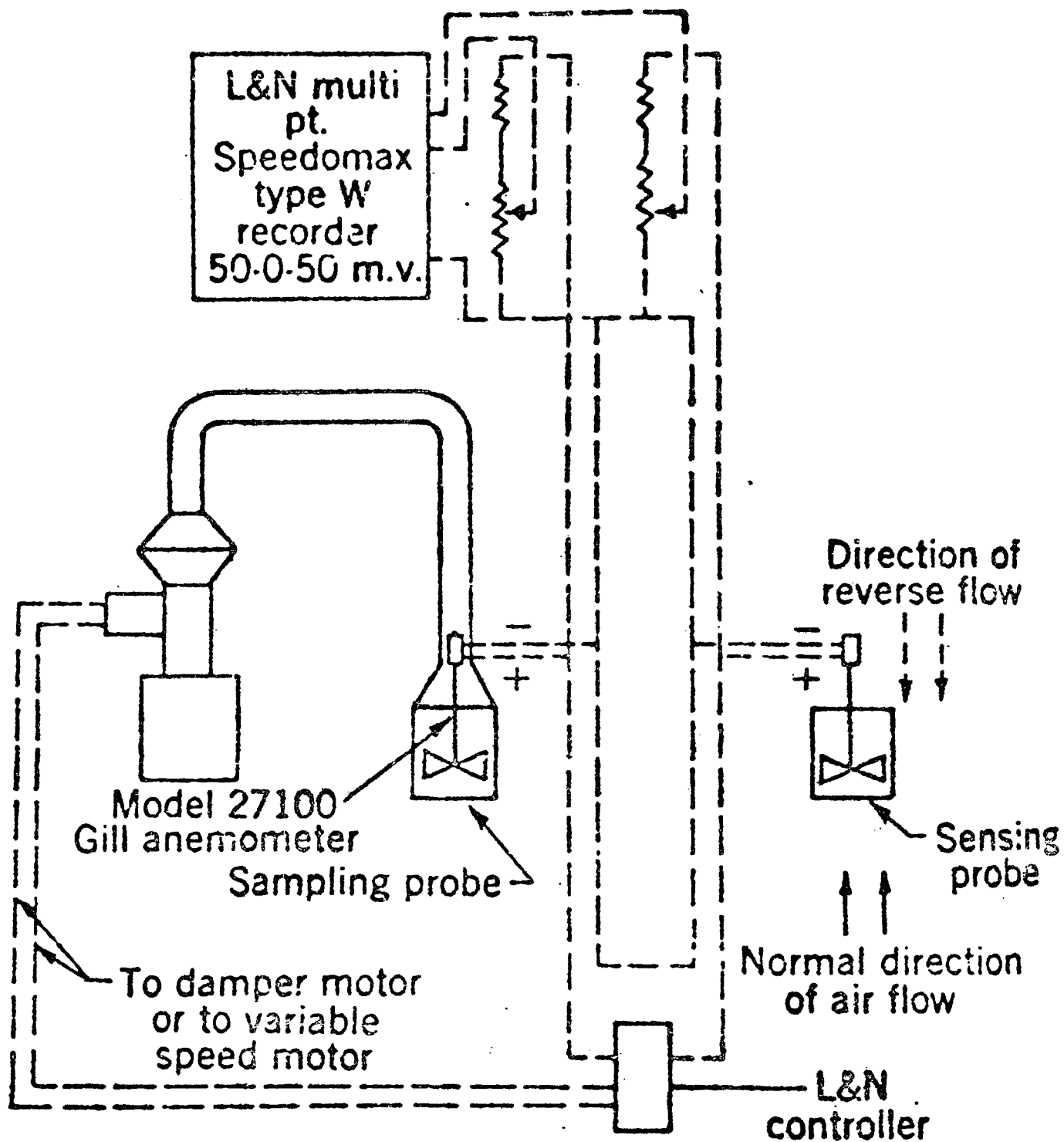


Figure (12) High volume isokinetic sampling system.

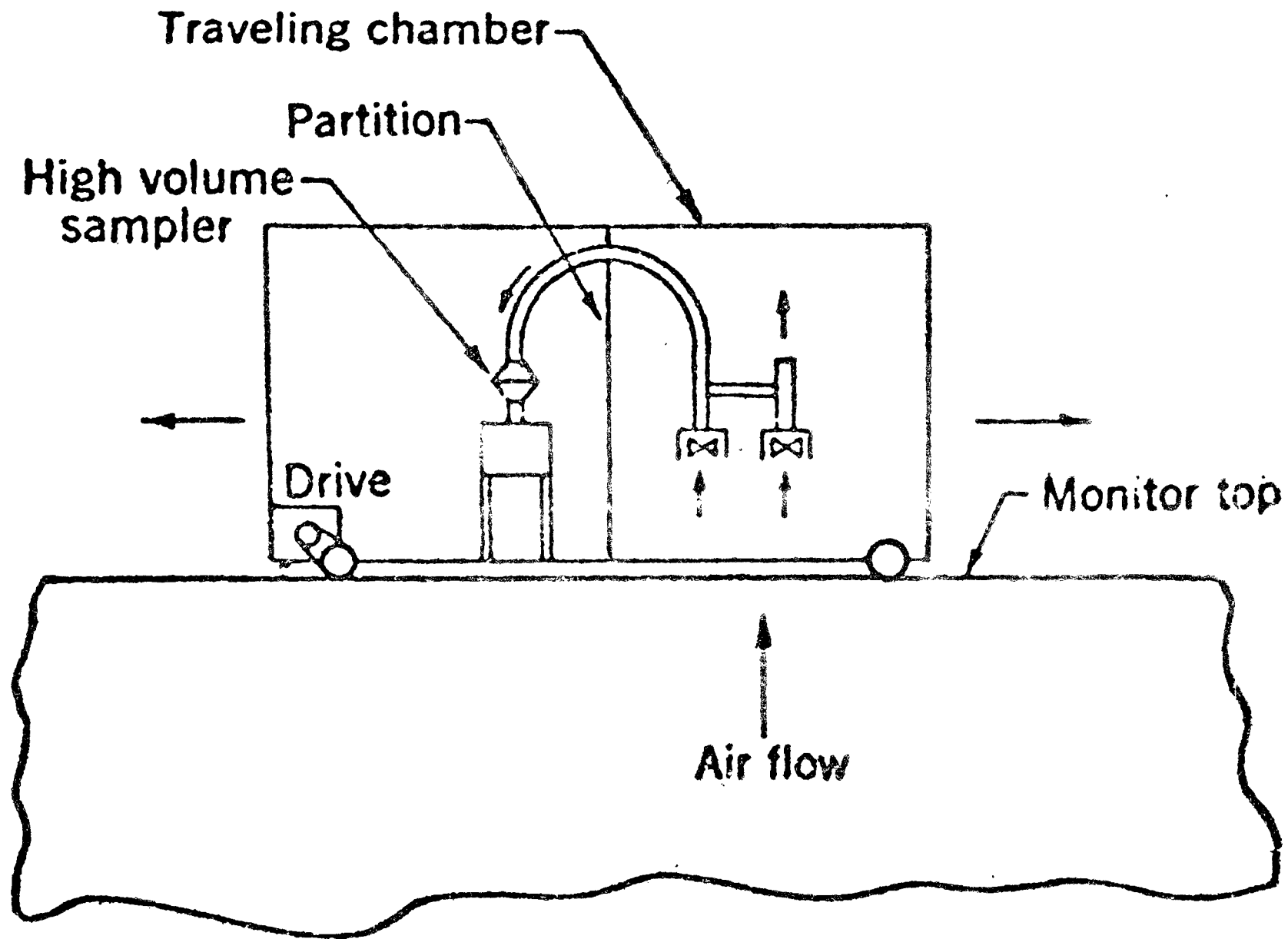


Figure (13) Traveling chamber.

Session III:

IMPACT OF FUGITIVE EMISSIONS

Mary Stinson
Session Chairman

RELATIVE IMPACTS OF OPEN SOURCES OF EMISSIONS

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RELATIVE IMPACTS OF OPEN SOURCES OF EMISSIONS

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Abstract

This paper describes approaches which can be taken for comparing and assessing open sources based on the magnitude and composition of the emissions. An open source is an industry which emits air pollutants in a primarily non-point manner. This includes sources of fugitive gases and dusts. These sources can have a major impact on ambient air quality owing to the persistence of the fine particle fraction and the disperse geographical locations. Also described are source-oriented sampling techniques which are used to determine the emission rate while minimizing the cost of sampling activities.

A methodology for objective source comparison, called source severity, is described which provides a consistent basis for comparing the various emissions generated by a single source as well as for comparing the environmental impact of diverse sources.

Open sources which have been evaluated by MRC include the following:

- Coal Storage
- Sand & Gravel Processing
- Cotton Defoliation
- Cotton Harvesting
- Grain Harvesting
- Grain Elevators
- Crushed Stone Processing
- Surface Coal Mining
- Crushed Granite Processing
- Crushed Quartzite Processing

Emission factors are presented for previously estimated and never-before-sampled open sources. An example of the differences found from previous estimates for crushed stone processing is given.

Introduction

An open source is an industry which emits air pollutants in a primarily non-point manner. Open sources are generally large, diffuse in nature, and are often comprised of several fugitive emission points. Consequently, the source strength of open sources is quite difficult to assess accurately since emission rate is not a simple function of gas flow rate and pollutant concentration.

Emissions of particulates and other pollutants from open sources have drawn increased attention in recent years because fugitive emission problems are frequently encountered. An investigation of approximately 140 open sources is being conducted under EPA Contract No. 68-02-1874 to provide a better understanding of the magnitude and composition of emissions for the purpose of determining needs for developing control technology. An understanding of the extent of emissions from open sources is also necessary to provide a perspective on the benefits to be derived from controlling point sources. If an ambient air quality problem is mostly caused by fugitive and open emissions, there is little to be gained in controlling a few point sources.

Some typical open sources are dusty material transfer, crushing, sizing, screening, landfill and excavation, blasting, ship loading and unloading, unpaved roads, molten metal transfer, beef cattle feedlots, tailings ponds, coal refuse fires, agricultural tilling, agricultural chemical application and volatilization, harvesting activities, storage piles, and erosion of soil.

Consideration

Open sources can be easily observed adding emissions to the atmosphere, but more often than not they are located in sparsely populated and remote areas of the country. Thus, why consider a source which, on the surface, may not affect a large segment of the population? Unfortunately, in many urban areas the ambient air quality standards for total suspended particulates (TSP) are not being met. This can be shown by looking at the yearly average TSP levels across the United States. Although certain industrial areas are hotspots of air quality degradation, the high annual TSP levels in the Western U.S. are difficult to explain or rationalize without a consideration of open sources of particulate emissions. Many of these are naturally or quasi-naturally occurring. Also, a mounting body of data indicates that particulate matter injected into the atmosphere at one location can be deposited

at locations up to several hundred miles away. The implication is obvious; in regions where this deposition occurs, enforcement of strict pollution control procedures on a local basis will have little impact upon air quality.

On a mass emission basis, open sources certainly do account for a disproportionate part of the impact on particulate air quality standards, as illustrated in Table I. Unpaved roads predominate U.S. mass emissions of particulates, but the influence of other open sources such as agricultural tilling, construction activities, grain handling, crushed stone processing, grain harvesting, and wind erosion of dormant soil are not to be discounted. This table lists the top 12 particulate emission sources from an overview matrix which includes 670 sources emitting criteria pollutants.

Measurement

Each open source has to be measured and evaluated on an individual basis. This is important because the method of evaluation depends on how the data and information are collected on the source. Two techniques of field sampling are briefly described below.

Suppose that the airborne drift is to be measured from an airplane applying a pesticide to a fieldcrop. The field can be divided into finite grid elements, and samplers are placed at selected locations upwind and downwind of the site, as shown in Figure 1. As the applicator moves back and forth across the field, samplers at each location can be turned on and off to correspond to the application zone represented by the dot in the figure. Correlation with downwind power law decay and crosswind variations can be accomplished without relocating a sampler site.

In this instance it may be important, both from a safety as well as operational standpoint, that control be maintained at some location where the proper perspective and overview of the application of a toxic material could be observed. This is achieved by utilizing a series of sampling devices which are controlled by radio frequency, as illustrated by the schematic diagram in Figure 2. Each individual unit can be turned on and off at the appropriate times through the remote control point. We have used a pulse modulation multiplex system which is capable of 360 simultaneous manipulations and operations. The operation could also be converted to automatic or computer control through the installation of proximity sensors located throughout the field being evaluated. With this technique a concentration for some time period is measured and then correlated back to a given application rate or area treated.

Table I. Overview matrix for air pollution sources - July 1975.

Source	Mass of Emissions (1,000 kg/yr)	
	Particulates	Percent of Total
Unpaved Roads	99,990,000	73.30
Coal-Fired Steam Electric Utilities	6,059,000	4.44
Agricultural Tilling	5,500,000	4.03
Oil-Fired Industrial/Commercial Boilers	3,633,000	2.66
Construction Activities	3,414,000	2.50
Grain Handling	2,935,000	2.15
Wind Erosion of Soil from Dormant Land	2,139,000	1.57
Crushed Limestone	2,034,000	1.49
Coal-Fired Industrial/Commercial Boilers	1,185,000	0.87
Cement Production	887,000	0.64
Steel Production	526,000	0.39
Grain Harvesting	452,000	0.33
Crushed Stone/Traprock	395,000	0.29
Lime Kilns	312,000	0.23
Crushed Granite	299,000	0.22

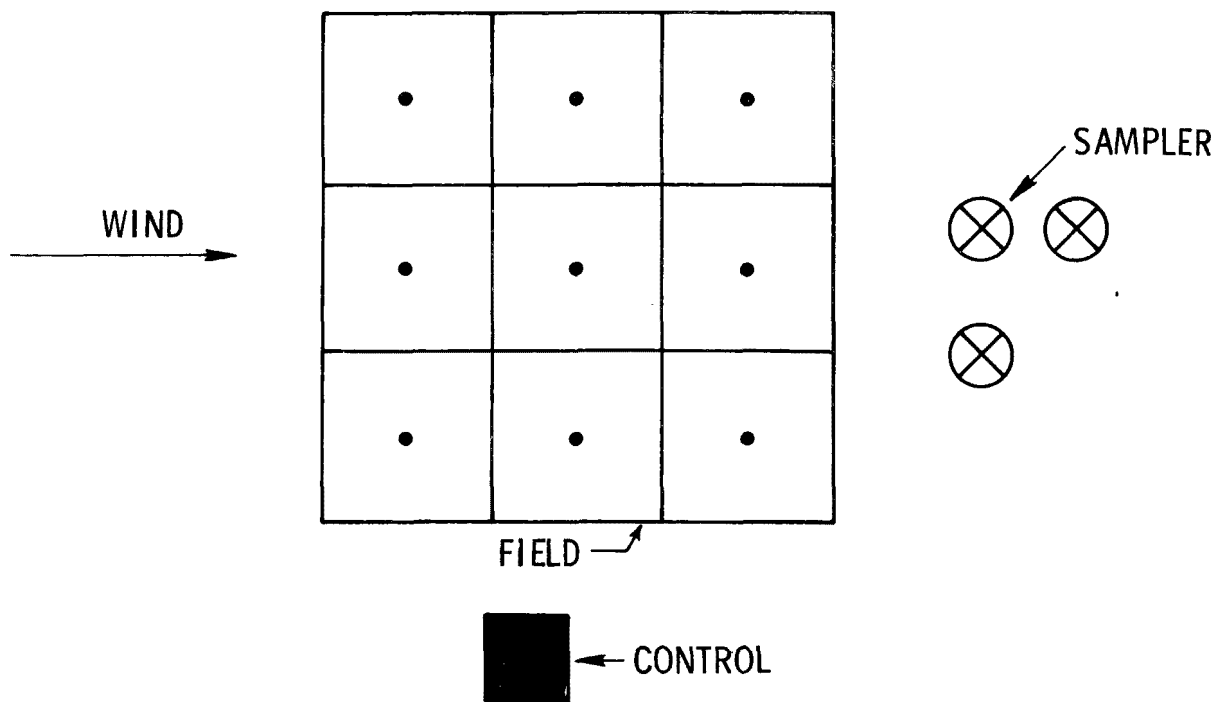


Figure 1. Drift sampling approach

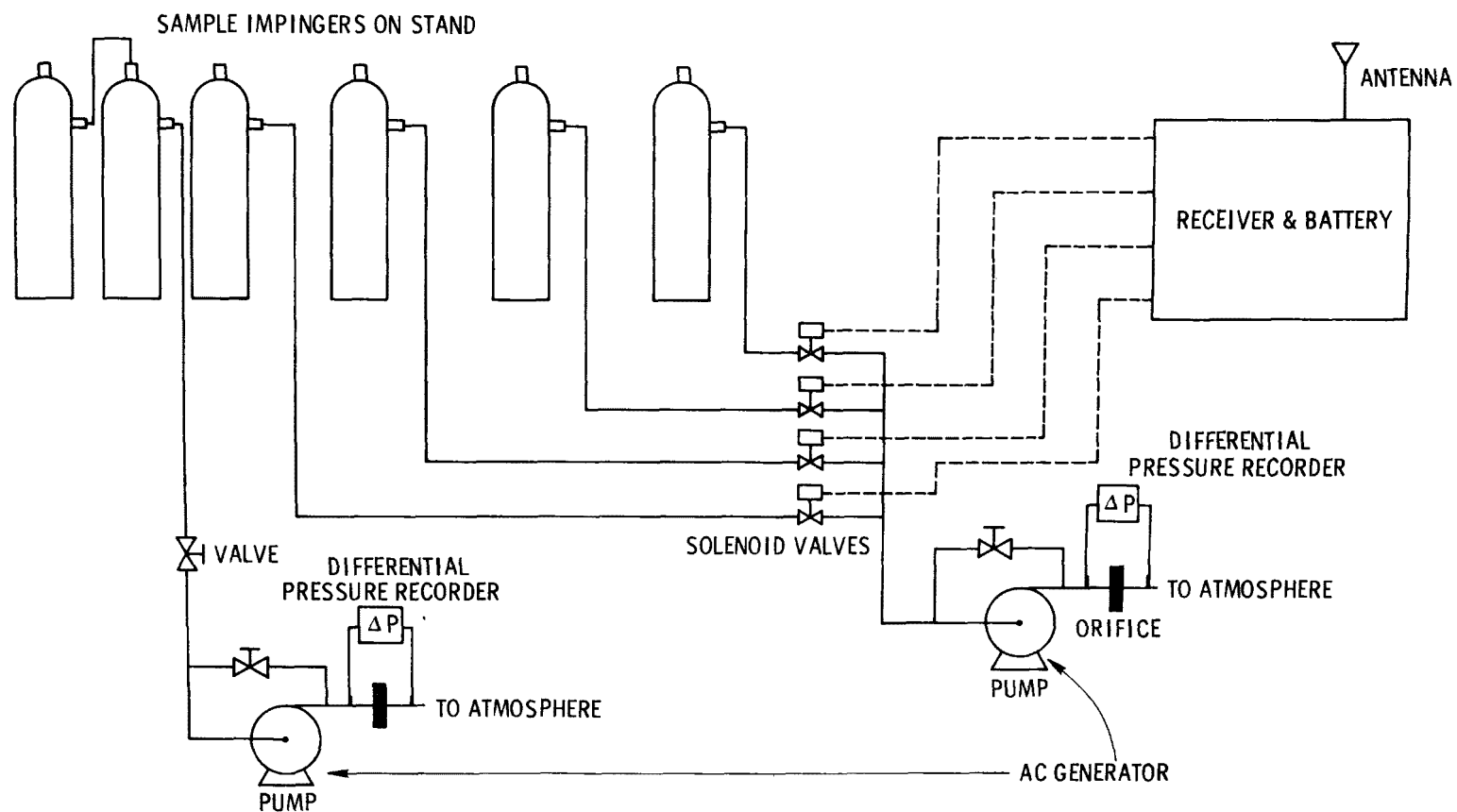


Figure 2. Airborne drift sampling apparatus

Another example of open source sampling techniques for continuously or semi-continuously emitting sources is to obtain many real-time concentration readings within a plume, preferably at several downwind distances along the centerline. Figure 3 shows such a system which is fully portable and consists of a portable anemometer, a beta attenuator dust monitor, and a personnel cassette sampler. Accurate ($\pm 25\%$) concentration readings can be rapidly taken with this system and it reduces field sampling costs by about 75% compared to Hi-volume sampler arrays. Also, particle size separation between the respirable fraction ($< 10 \mu$) and TSP ($< 50 \mu$) can be accomplished.

Source Severity

A methodology has been developed which provides a consistent basis for comparing various emissions generated by a single and for comparing the environmental hazard between different single sources. Called the source severity, this multifaceted approach can compare, in a relative fashion, an open source with an elevated point source, an open source with another open source, one pollutant emission with another pollutant emission, the components of a pollutant with the whole pollutant (e.g., free silica, Pb, Cd), one location of emissions with another location, and the effects of emission parameter changes. As an assessment tool, the source severity approach has proved to be extremely flexible and useful. A discussion of its development and types of use follow.

The air pollution severity of a given source should in some way be proportional to the degree of potential hazard it imposes on a population in its environment. The relative hazard, H, from a specific emission can be defined as being directly proportional to the delivered dose, the probability of dose delivery, and the number of people who receive it, and inversely proportional to the toxicity of the material as follows:

$$S \propto H \propto \frac{NP\psi}{LD_{50}} \quad (1)$$

where

- S = source severity
- H = relative hazard
- N = number of persons
- LD_{50} = lethal dose for 50% of the people exposed
- P = probability of dose delivery
- ψ = delivered dose = $B \cdot R' \cdot \int \chi(t) dt$
- B = average breathing rate
- R' = lung retention factor
- $\chi(t)$ = concentration time history

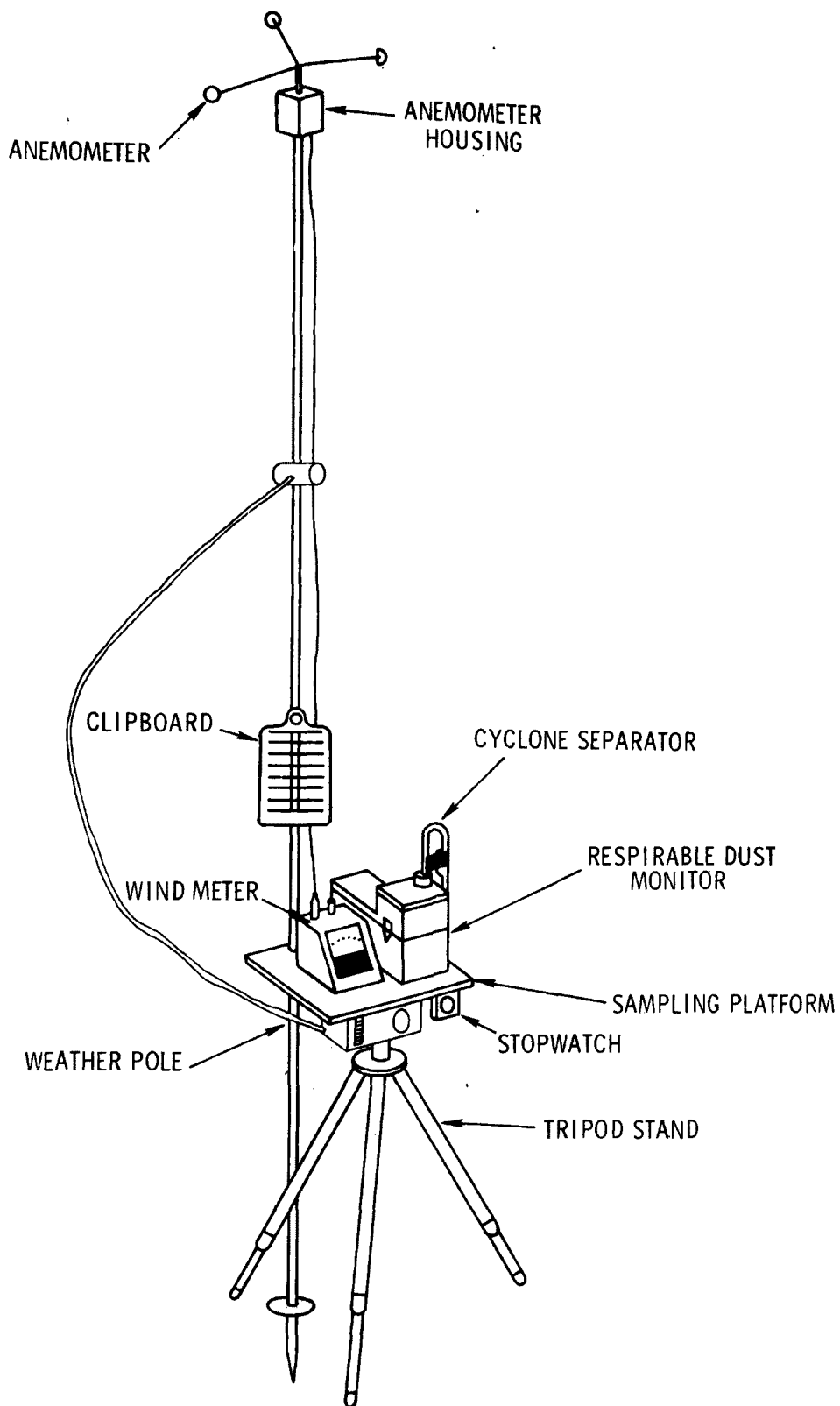


Figure 3. Sampling apparatus

The source severity, S, has been defined as the ratio of the dose of a pollutant delivered to a population relative to some potentially hazardous dose. Since LD₅₀ data are not available for humans as a measure of hazardous dose, another measure was used. The potentially hazardous dose for a given pollutant from a specific point source is defined instead as:

$$\psi_F = NBR' \int_{t_1}^{t_2} TLV(t) K dt \quad (2)$$

where ψ_F = potentially hazardous dose, g
 N = population exposed to a specific source, persons
 B = average breathing rate, m³/s-person
 R' = lung retention factor for the pollutant of interest (dimensionless factor, $0 < R' < 1$)
 K = safety factor = $\frac{8}{24} \cdot \frac{1}{100}$
 t = time
 t_1 = start time, s
 t_2 = finish time, s
 TLV° = threshold limit value, g/m³

The total time of interest, T, is defined as:

$$T = t_2 - t_1 \quad (3)$$

Similarly, a hazard potential factor, F, is defined as:

$$F = TLV \cdot K \quad (4)$$

Since TLV is a constant:

$$\psi_F = N \cdot B \cdot R' \cdot T \cdot F \quad (5)$$

The term, ψ_F , is, thus, the potentially hazardous dose for each pollutant which has a TLV value. In the safety factor, K, the ratio 8/24 adjusts the TLV to a continuous rather than workday exposure, and the factor of 1/100 accounts for the fact that the general population is a higher risk group than healthy workers.

The actual pollutant dose delivered, ψ_A , from a given point source can be calculated as follows:

$$\psi_A = N \cdot B \cdot R' \int_{t_1}^{t_2} \chi(t) dt \quad (6)$$

where $\chi(t)$ = the actual ground level concentration time history of a pollutant of interest emitted by a specific point source, g/m³

The value of $\chi(t)$ is very difficult to obtain and was therefore approximated by an average value, $\bar{\chi}$. The total actual dose delivered for a specific pollutant from a specific source is then:

$$\psi_A = N \cdot B \cdot R' \cdot T \cdot \bar{\chi} \quad (7)$$

Since our measure of source severity was defined as the ratio of the two dosages, then:

$$S = \frac{\psi_A}{\psi_F} = \frac{N \cdot B \cdot R' \cdot T \cdot \bar{\chi}}{N \cdot B \cdot R' \cdot T \cdot F} \quad (8)$$

or

$$S = \frac{\bar{\chi}}{F} \quad (9)$$

The above term for source severity results in a simple ratio of the time-averaged concentration to a potentially hazardous concentration from a single source, although it is based on dosage. Ambient concentration measurements around a source are commonly considered as the primary entity and dosage derived from them. From the experimental point of view, the inverse is true; mostly dosage is observed and, at the price of involved assumptions, a concentration valid for some smoothing or sampling time is reported.

In order to calculate the source severity, $\bar{\chi}/F$, and the affected population, the Gaussian plume dispersion model is employed, which relates the concentration of pollutant occurring at ground level at any given distance from the source of emissions to the rate and height of the emissions and the local wind velocity. The ground level concentration is of primary importance since it is the one to which the human population is exposed. Also, the rate and height of emissions are quantities which can be readily measured and/or compiled.

For elevated point sources the maximum ground level concentration, χ_{\max} (ref. 1), is used to determine the numerator of source severity:

$$\chi_{\max} = \frac{2Q \sigma_z}{\pi e u h^2 \sigma_y} \quad (10)$$

where χ_{\max} = maximum ground level concentration (3-min averaging time), g/m³
 π = 3.14
 e = 2.72
 u = wind speed, m/s
 h = emission height, m
 σ_z = vertical dispersion coefficient, m
 σ_y = horizontal dispersion coefficient, m
 Q = emission rate, g/s

The above equation yields a value for a short-term averaging time (~ 3 min) during which the Gaussian plume dispersion equation is valid. For a continuously emitting source, the average maximum concentration, $\bar{\chi}_{\max}$, is a function of sampling time, t , and it can be related to χ_{\max} as follows (ref. 2):

$$\bar{\chi}_{\max} = \chi_{\max} \left(\frac{t_1}{t_2} \right)^p \quad (11)$$

where t_1 = 3 min
 t_2 = 24 hr
 p = 0.17

We use national average meteorological conditions since they prevail at most locations and it simplifies the comparison process; thus, atmospheric stability is approximately neutral (class C assumed), wind speed is 4.5 m/s, and $\sigma_y \cong \sigma_z$ (ref. 2) or:

$$\frac{\sigma_z}{\sigma_y} \cong 1.0 \quad (12)$$

The severity equation becomes:

$$S = \frac{\bar{\chi}}{F} = \frac{\chi_{\max} \left(\frac{t_1}{t_2} \right)^p}{\text{TLV} \cdot \frac{8}{24} \cdot \frac{1}{100}} \quad (13)$$

or

$$S = \frac{(2) (105) Q \sigma_z}{\pi e u h^2 \sigma_y (\text{TLV})} = \frac{5.5 Q}{(\text{TLV}) h^2} \quad (14)$$

Since the criteria pollutants (particulates, SO_x , NO_x , CO, and HC) have established ambient air quality standards, the appropriate standard (in g/m^3) is substituted for the potential hazard factor, F. Severity equations for each of the five criteria pollutants have also been derived, and the only data needed then to calculate source severity, S, are the emission rate, Q, and the emission height, h.

For most open sources, which are ground level sources ($h = 0$), x_{\max} occurs by definition at the nearest plant boundary or public access. Since this also occurs when $y = 0$, the appropriate equation to calculate the "maximum" concentration is (ref. 2):

$$\chi = \frac{Q}{\pi \sigma_y \sigma_z u} \quad (15)$$

By letting D equal the distance to the occurrence of x_{\max} for ground level sources (ref. 3):

$$\sigma_y = 0.209D^{0.903} \quad (16)$$

$$\sigma_z = 0.113D^{0.911} \quad (17)$$

Thus, x_{\max} is determined as follows:

$$\chi_{\max} = \frac{42.36 Q}{\pi u D^{1.614}} \quad (18)$$

It will be noted that, under average meteorological conditions, the x_{\max} equations are identical with the algebraic substitution of:

$$h^2 = 0.01737D^{1.814} \quad (19)$$

If a ground level source is a line or area source rather than a point source, it is treated in a similar fashion to calculate source severity.

Once source severities have been determined, one of the uses is to compare locationally a source's impact on the population. Affected population designates the number of persons exposed to high concentrations, e.g., those for which $\chi_{\max}/F > 1.0$. This quantity is useful in characterization because a given source may have a high severity, yet, if it is located in a sparsely populated area it may have only a small effect on human health.

Another form of the plume dispersion equation is needed to calculate the affected population since the population is assumed to be distributed uniformly around the source. If the wind directions are taken to 16 points and it is assumed that the wind directions within each sector are distributed randomly over a period of a month or season, it can be assumed that the effluent is uniformly distributed in the horizontal within the sector. The appropriate equation for the average concentration as a function of distance, $\bar{\chi}(X)$, is (ref. 2):

$$\bar{\chi}(X) = \frac{2.03 Q}{\sigma_z u X} \exp \left[-\frac{1}{2} \left(\frac{h}{\sigma_z} \right)^2 \right] \quad (20)$$

where

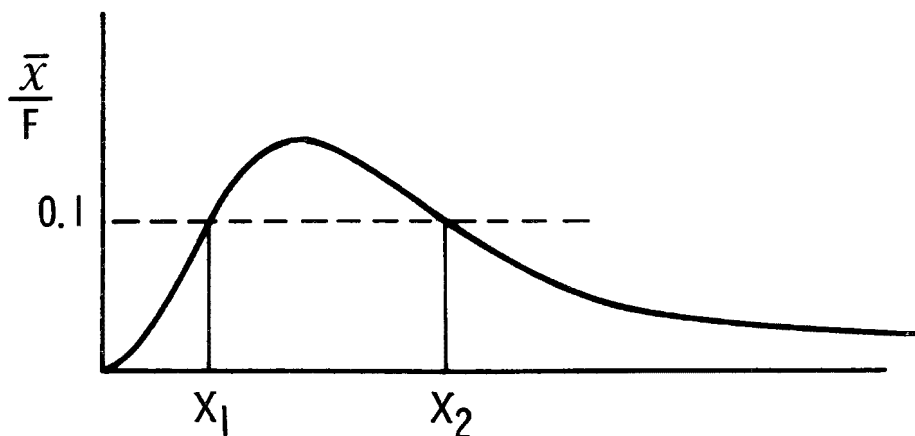
Q = emission rate, g/s
 h = effective emission height, m
 X = downwind distance from source, m
 u = 4.5 m/s
 $\sigma_z = 0.113 X^{0.911}$ = vertical dispersion coefficient, m

To find the distances at which $\bar{\chi}(X)/F > 1.0$, roots are determined for the following equation:

$$0 = \left\{ \frac{2.03 Q}{F \sigma_z u X} \exp \left[-\frac{1}{2} \left(\frac{h}{\sigma_z} \right)^2 \right] \right\} - 1.0 \quad (21)$$

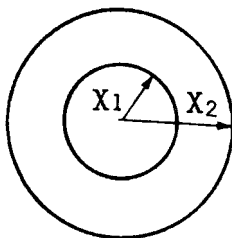
Since Equation 21 is a transcendental equation the roots are found by an iterative technique using the computer.

For a specified emission from a typical source, $\bar{\chi}/F$ as a function of distance might look as follows:



DISTANCE FROM SOURCE

with X_1 and X_2 being the distances where $S = 1.0$. If those distances are swept through 360° , an annulus is obtained containing the affected population:



The affected area in km^2 is then computed as:

$$A = \pi(X_2^2 - X_1^2) \quad (22)$$

where X_1 and X_2 are the two roots of Equation 21.

The population density, D_p , of the area of concern is determined and the product $A \cdot D_p$ is designated as the affected population and reported as number of persons.

Experimental Results

In order to apply source severity methodology in an assessment of open source types, extensive field sampling was conducted so that emission rates could be compiled. The simplest way to take a "first look" at many open sources is to sample a source thought to be representative of the whole and estimate the emission factor. Tables II through V, present the emission factors determined for open coal mining and storage, agricultural activities, fugitive emission sources at grain elevators, and crushed stone quarrying and processing, respectively. The emphasis was given to determining the fine particle, or respirable ($<7 \mu\text{m}$), fraction where dust emissions were concerned. It is this fraction which will impact human health and is likely to disperse extensively beyond plant boundaries.

Comparison of Emission Factors with Previous Studies

Table VI compares the emission factors as determined by downwind ambient sampling with those estimated in the Compilation of Air Pollutant Emission Factors (ref. 4). As can be seen, the emission factors for crushed stone processing as determined in our work are two orders of magnitude lower than previous estimates.

Table II. Coal emission factors.

Source Type	Emission Factor, lb/ton (resp)
Surface Coal Mining	0.025
Drilling	3.2×10^{-3}
Coal Loading	3.4×10^{-3}
Transport & Unloading	9.7×10^{-3}
Blasting	8.3×10^{-3}
Augering	1.6×10^{-3}
Coal Storage	0.013

Table III. Agricultural emission factors.

Source Type	Emission Factor
Cotton Defoliation (w/Arsenic Acid)	12.2 lb/ton
Cotton Harvesting	
Picking	2.63 lb/mi ² (resp)
Stripper	24.6 lb/mi ² (resp)
Grain Harvesting	2.34 lb/mi ² (resp)

Table IV. Grain elevator emission factors.

Unit Operation	Emission Factor, lb/ton (resp)
Truck Unloading	
Uncontrolled	0.272
Controlled	0.0028
Truck Loading	0.0011
Hopper Railcar Loading	0.0031
Ship Loading	0.0013

Table V. Emission factors for aggregate unit operations.

Unit Operation	Emission Factor for TSP in lb/ton of Material Processed Through Primary Crusher			
	Granite	Traprock	Limestone	Quartzite
Drilling	-	-	-	0.060
Blasting	0.16	-	-	-
Loading at Quarry and Vehicular Transport to Plant	-	0.02	0.004	0.34
Unloading and Primary Crushing and Screening	-	0.026	0.002	-
Secondary Crushing and Screening	0.044	0.002	-	0.024
Conveying	-	0.004	-	-
Unpaved Road Traffic Between Finished Stockpile and Nearest Paved Highway	0.048	0.002	-	-
TOTAL	0.25	0.056	0.006	0.42
Fraction Respirable, by Weight	6%	11%	17%	6%

Table VI. Comparison of the emission factors at traprock plants for AP-42 and MRC sampling.

Operation	AP-42			MRC (Sampling at Traprock Plants)		
	Uncontrolled Total Emissions, g/metric ton	Percent Settling in Plant	Suspended Emissions, g/metric ton	Total Particulate Emissions, g/metric ton	Percent Respirable Emissions	Respirable Emissions, g/metric ton
Primary Crushing	250(0.5) ^a	80	50(0.1)	13 (0.026)	10	1.3(2.6x10 ⁻³)
Secondary Crushing and Screening	750(1.5)	60	300(0.6)	1 (0.002)	55	0.6(1.2x10 ⁻³)
Tertiary Crushing and Screening	3,000(6.0)	40	1,800(3.6)	0.4(0.0008)	18	0.1(2.0x10 ⁻⁴)
Fines Milling	3,000(6.0)	25	2,250(4.5)	0.1(0.0002)	16	0.2(4.0x10 ⁻⁴)
Recrushing and Screening	2,500(5.0)	50	1,250(2.5)			
TOTAL	9,500(19.0)	41	5,605(11.3)	14.5(0.03)	15	2.2(4.4x10 ⁻³)

Note: Blanks indicate no data reported.

^a: Numbers in parentheses are in English units, i.e., pounds per ton.

There are several possible explanations for these differences. Our emission factors were determined by measuring ambient air concentrations around a source and then calculating the emission rate using a dispersion equation. The samplers were placed about 30 m to 40 m away from the source and, thus, did not measure particles that settled between the source and the samplers. Based on a particle count on high volume filters, the samplers did not collect particles more than 40 μm in diameter. Thus, the emission factor for total particulates includes only particulates less than 40 μm in diameter.

On the other hand, the previous emission factor estimates were based on the results of sampling the dust loading at the inlet of a baghouse used to control dust emissions from crushing and screening operations at a crushed traprock plant, and on the assumption that about 41% of the emitted particulates settle within the plant. High air flows encountered in baghouse ducts cause the entrainment of very large particles (up to about 350 μm in diameter), so the emission factor for total particulates based on the dust loading at the baghouse inlet may be an inflated value.

One of the purposes of determining an emission factor is to estimate the ambient air concentrations due to a source. The sampling methods outlined in this report allow a more realistic estimate of source emissions than previously reported since they are based on measurements of air concentrations around the contributing source of interest. Based on this, the emission factors in Tables II through V will predict, using the same dispersion methodology, real-life ambient air contributions of sources.

Future plans call for a more extensive evaluation of the major sources of emission, overall mass emissions, respirable mass emissions and composition of some of these sources, and a more complete analysis in order to best judge relative severity and emission burden.

Acknowledgment

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THE IMPACT OF FUGITIVE EMISSIONS
OF FINE PARTICLES

by

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THE IMPACT OF FUGITIVE EMISSIONS OF FINE PARTICLES

Introduction

Failure to attain national ambient air quality standards for total suspended particulates (TSP) in both urban and rural areas has spurred a detailed reexamination of the national TSP problem. As a result of this assessment, it has become evident that fugitive emissions sources contribute substantially to TSP levels in many of the nonattainment areas. The regional impact of fugitive sources of particulate matter is the result of emissions of fine particles which have the potential for atmospheric transport over substantial distances from the source.

Analysis of the impact of fugitive emissions of particulate matter must take into account the ranges of particle size associated with the adverse effects of particulate air pollution. As shown in Figure 1, most of the recognized adverse effects are attributable to fine particles, i.e., particles smaller than about 10 μm in equivalent aerodynamic diameter, which may be transported over distances of regional scale. By way of contrast, soiling phenomena which result from the rapid settling of coarse particles, tends to be confined to localized areas in the vicinity of the source.

This paper focuses on two topics critical to the assessment of the impact of fugitive emissions of fine particles: (a) methods for fugitive emissions quantification, and (b) analysis of the potential for atmospheric transport of particulate matter.

Methods for Fugitive Emissions Quantification

The quantification of particulate emissions from fugitive sources is necessary to the rational assessment of the environmental impact created by these sources and to the development of effective control technology. In addition to mass rates of emission, information is needed on particle size distribution and the presence of toxic constituents within specific size ranges.

In large part, proven methods for quantifying fugitive emissions have not been fully developed. Atypical quantification problems are presented by the diffuse and variable nature of fugitive sources. Standard source testing methods, as written, strictly apply only to well defined, constrained flow fields with velocities above about 2 m/sec.

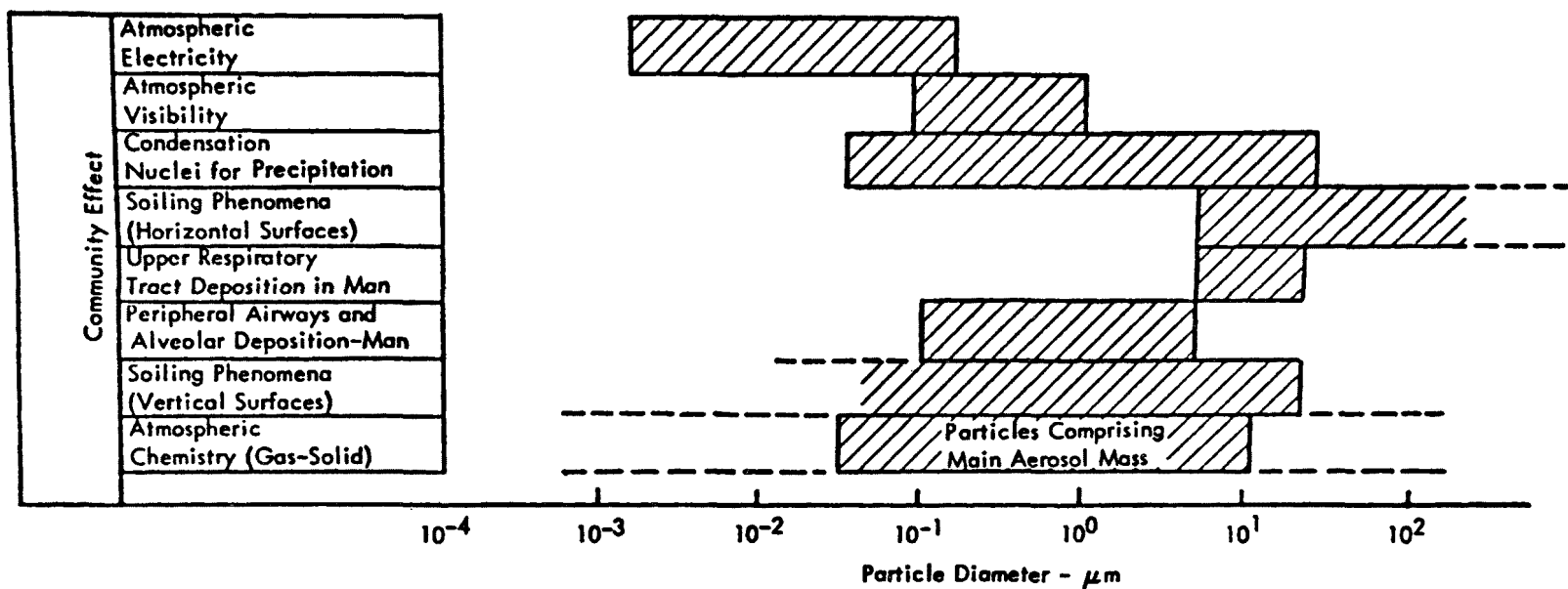


Figure 1 - Effects of Particulate Air Pollution versus Particle Size ^{4/}

MRI has employed two basic sampling strategies to quantify emissions from open dust sources: exposure profiling and dispersion model calculation. Each of these techniques is discussed below.

Exposure profiling: Particulate emissions from an open source can be determined directly by measuring the total passage of airborne particulate matter (after subtraction of background) at some distance downwind of the source. This method is feasible (a) for area sources smaller than about 0.1 km in diameter or (b) for line (elongated area) sources of width smaller than 0.1 km.

The passage of airborne particulates can be obtained by spatial integration (over the effective cross-section of the plume) of distributed measurements of exposure (mass/area). The exposure is the point value of the flux (mass/area-time) of airborne particulates integrated over the time of measurement. Mathematically stated, the total mass emission rate is given by

$$R = \frac{1}{t} \iint_A \frac{m(h,w)}{a} dh dw \quad (1)$$

where m = particulate catch by exposure sampler after subtraction of background

a = effective intake area of sampler

t = sampling time

h = vertical distance coordinate

w = lateral distance coordinate

A = effective cross-sectional area of plume

In order to obtain an accurate measurement of airborne particulate exposure, sampling must be conducted isokinetically, i.e., flow streamlines enter the sampler rectilinearly. This means that the sampling intake must be aimed directly into the wind and, to the extent possible, the sampling velocity must equal the local wind speed. The first condition is by far the more critical.

A variation in the exposure profiling method has recently been developed by MRI for application to buoyant plumes emitted by pyrometallurgical processes. This method utilizes a two-dimensional grid of thermocouples, horizontally

distributed above the source, for temporal and spatial characterization of the plume. Laboratory experiments have shown that plume temperature and concentration profiles are linearly related.

Dispersion model calculations: Atmospheric dispersion models can also be used to calculate rates of emission from area sources based on ground-level measurement of upwind and downwind air quality under known meteorological conditions. This method of emissions quantification is required when the extent of vertical dispersion at the downwind edge of the source is too large to allow for ground-based characterization of the vertical exposure profile.

Atmospheric Transport of Fugitive Dust

This section presents an assessment of factors which determine the drift distances of fugitive dust particles in the atmosphere. Drift distance is defined as the horizontal displacement from the point of particulate injection to the point of particulate removal by ground-level deposition.

Factors to be considered in this assessment may be grouped into two categories:

1. Meteorological factors - properties of the atmosphere which affect contaminant advection and turbulent diffusion over surfaces of varying roughness scales.
2. Source factors - height of injection and particulate properties which affect gravitational settling and vertical mixing.

This assessment does not treat atmospheric washout of particulate matter.

Meteorological factors: Fugitive dust particles are typically injected into the lower portion of the "surface layer" region of the atmosphere which extends from ground level to a height of about 100 m. In this region the profile of the wind and its turbulence characteristics are strongly dependent on surface roughness properties.

For neutral atmospheric stability, the vertical profile of mean wind speed, $u(z)$, in the surface layer is described by a logarithmic relationship:

$$u(z) = \frac{u_*}{k} \ln \left(\frac{z}{z_0} \right) \quad (2)$$

where u_* = friction velocity

k = von Karman's constant (0.4 for clear fluids)

z_0 = surface roughness height

Neutral stability occurs with wind speed exceeding 12 mph or with overcast conditions regardless of wind speed.

The friction velocity, u_* , is related to the rate of momentum exchange at the surface:

$$u_* = (\tau_0 / \rho_a)^{1/2} \quad (3)$$

where τ_0 = surface shear stress

ρ_a = density of air

Within the surface layer, the vertical flux of momentum (and hence u_*) is known to be roughly constant and the eddy diffusivity is given by

$$e(z) = k u_* z \quad (4)$$

Aerodynamic roughness height, z_0 , is related to the size, shape and spatial density of the roughness elements. Based on similarity concepts Lettau^{2/} has derived the following expression for evenly spaced elements:

$$z_0 = \frac{H a}{2A} \quad (5)$$

where H = effective height of roughness elements

a = silhouette area normal to the wind

A = total ground area per element

$1/2$ = average drag coefficient.

Figure 2 gives roughness heights for various natural and man-made roughness features.

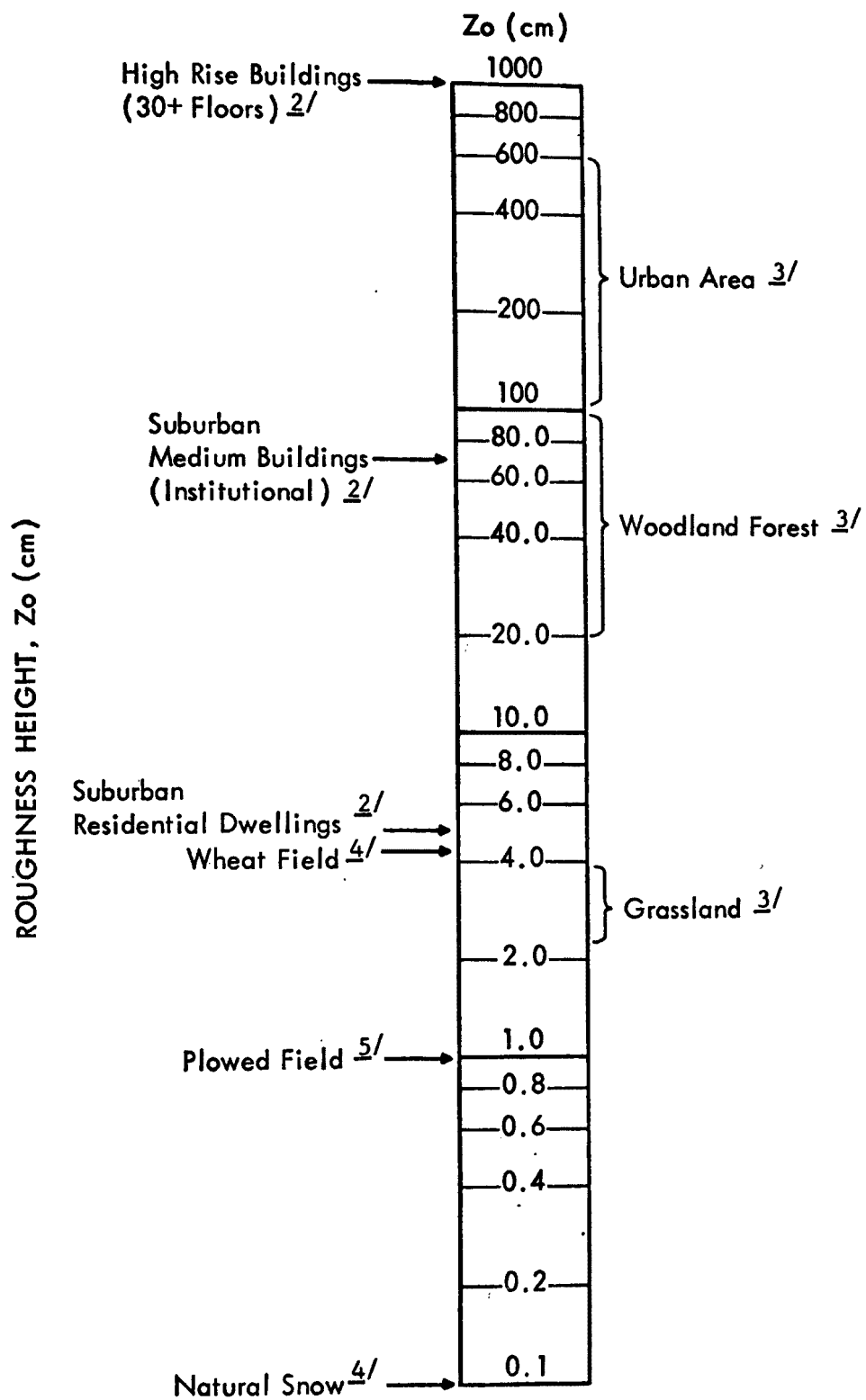


Figure 2 - Roughness Heights for Various Surfaces

Source factors: The primary source factors which affect the drift distance of a fugitive dust particle are injection height, h , and particle settling velocity, V_s , which may be approximated by the Stoke's relationship:

$$V_s = 0.00301 \rho_p D^2 \quad (6)$$

where V_s = terminal settling velocity (cm/sec)

ρ_p = density of particle (g/cm^3)

D = particle diameter (μm)

Fugitive dust particles typically have a mineral composition with a density of about 2.5 g/cm^3 .

Calculation of drift distance: In the past, most analyses of the atmospheric dispersion of particles with appreciable settling tendencies have focused on the distribution of settling rate, $S(x)$, expressed as:

$$S(x) = V_s C_o(x) \quad (7)$$

where C_o = the ground-level concentration of particulate with settling velocity V_s

x = downwind distance from the source

Accordingly, an Eulerian approach to the problem has been taken.

However, analysis of particle drift with no net effect of atmospheric turbulence, is most conveniently treated by a Lagrangian approach. This is illustrated in the following section.

Case 1: Monodisperse particles, single injection height, negligible turbulence effect.

Consider the case of a steady stream of monodisperse particles released from a continuous crosswind line source at height h . It is assumed that each particle during its lifetime in the atmosphere is subjected to a balanced set of vertical turbulent velocity fluctuations with the result that the particle does not deviate appreciably from the trajectory it would have in the absence of turbulence.

The vertical position, z_p , of the particle as a function of time is given by

$$z_p(t) = h - V_s t \quad (8)$$

Substitution of Eq. (8) into Eq. (2) gives the following expression for the horizontal speed of the particle:

$$u_p = \frac{u_*}{k} \ln \left(\frac{h - V_s t}{z_o} \right) \quad (9)$$

The particle drift distance, x_p , is given by:

$$x_p = \int_0^{\frac{h-z_o}{V_s}} u_p dt \quad (10)$$

where the upper limit of integration is the lifetime of the particle in the atmosphere. Integration of Eq. (10) yields

$$x_p = \frac{u_* h}{k V_s} \left[\ln \left(\frac{h}{z_o} \right) - 1 \right] + \frac{u_* z_o}{k V_s} \quad (11)$$

To determine the effect of injection height and roughness height on the drift distance of particles of given aerodynamic sizes, the wind speed at $z = 100$ m was fixed at 6.9 m/s (15.4 mph) and friction velocities were determined from Eq. (2). The results are shown in Table 1 for injection heights of 1, 3 and 10 m and for roughness heights spanning the range given in Table 1. Figure 3 shows the variations of x_p for $h = 3$ m, measured above z_o .

As expected, for particles of a given size, drift distance increases with injection height and decreases with roughness height. The latter effect is a direct result of the decrease in wind velocity near the surface caused by obstacles to the flow.

Case 2: Monodisperse particles, single injection height, turbulent atmosphere.

Table 1. PARTICLE DRIFT DISTANCES CALCULATED FROM EQ. (11)

Injection height, ^{a/} h (m)	Roughness height, z _o (m)	Friction velocity, u* (cm/sec)	Drift distance, x _p , by particle size				
			30 μm	20 μm	10 μm	5 μm	1 μm
1	0.01	30.0	40.6 m	91.2 m	366 m	1,460 m	36.6 km
	0.05	36.4	29.5	66.4	266	1,060	26.7
	0.10	40.0	24.2	54.4	218	871	21.8
	0.50	52.2	12.5	28.1	113	450	11.3
3	0.01	30.0	157.1 m	353 m	1,418 m	5.66 km	141.8 km
	0.05	36.4	128.2	288	1,157	4.62	115.7
	0.10	40.0	112.9	254	1,019	4.07	101.9
	0.50	52.2	73.5	165	663	2.65	66.3
	1.00	60.0	56.4	127	509	2.03	50.9
10	0.01	30.0	655 m	1,474 m	5.92 km	23.6 km	592 km
	0.05	36.4	582	1,309	5.25	21.0	525
	0.10	40.0	541	1,216	4.88	19.5	488
	0.50	52.2	423	952	3.82	15.3	382
	1.00	60.0	363	816	3.28	13.1	328

^{a/} Injection height measured above roughness height.

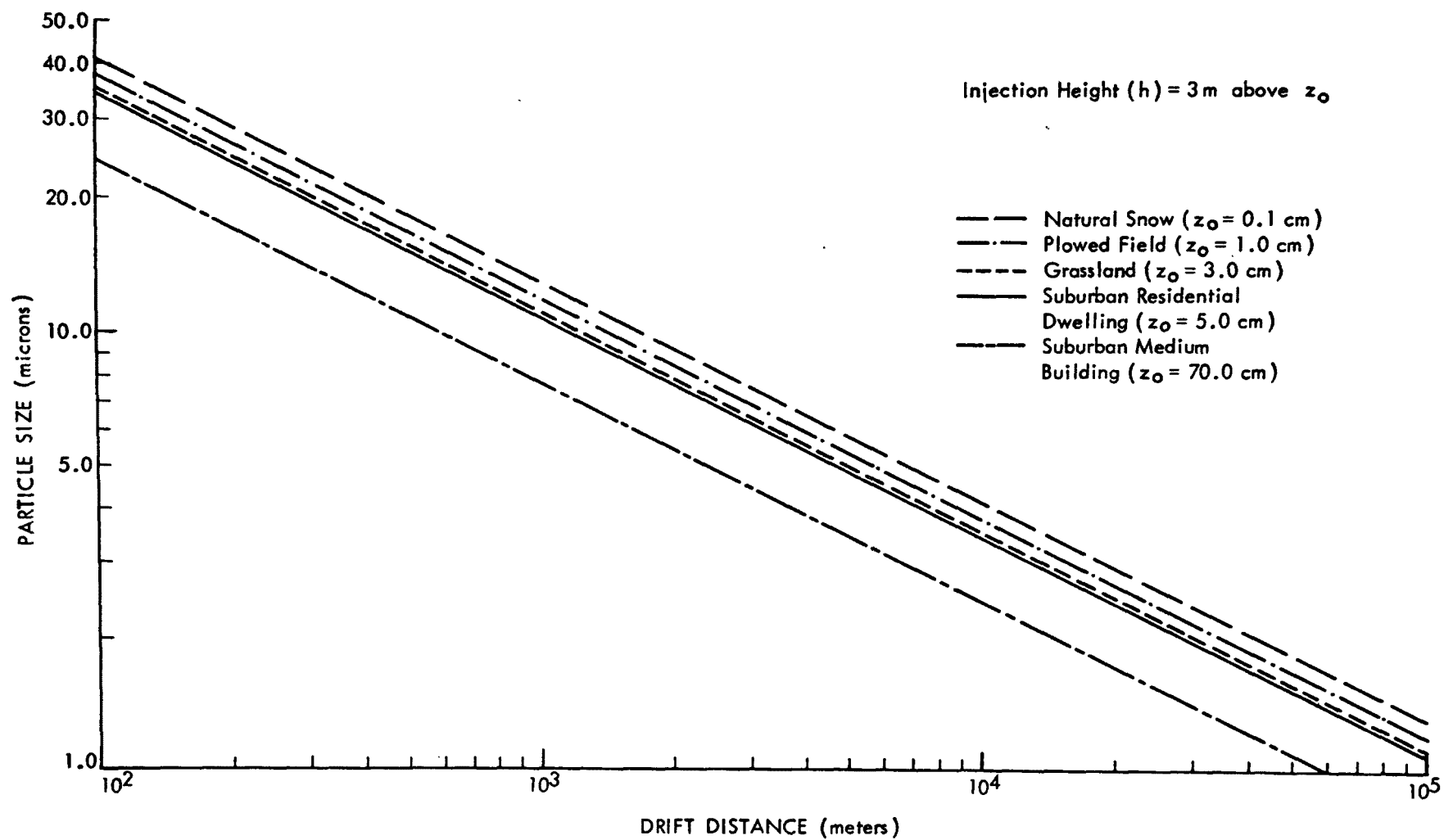


Figure 3 - Relationship Between Particle Size and Drift Distance

The analysis presented under Case 1 assumed that all particles generated from a particular fugitive dust source were deposited at the same point downwind (x_p). Clearly, however, particles subjected to a preponderance of downward turbulent velocity fluctuations will settle from the atmosphere at distances less than x_p and particles propelled above the trajectory defined above may drift far beyond x_p . In other words, because of the random nature of turbulent velocities, x_p approximates the distance at which half of the particles have deposited on the surface.

The specific question addressed here has to do with the form of the settling rate distribution. Recalling Eq. (7), this problem reduces to finding the distribution of ground-level concentration by solving the appropriate transport equations and accompanying boundary conditions.

The phenomena of quasi-steady advection and turbulent diffusion from a continuous line source under the condition of uniform wind speed is described by the following equation:

$$U \frac{dC}{dx} = pU \frac{d}{dz} \left(z \frac{dC}{dz} \right) + v_s \frac{dC}{dz} \quad (12)$$

where C = particulate concentration

U = uniform speed of crosswind

p = turbulence parameter

The uniform wind speed, U , is assumed to have the value given by the Case 1 velocity profile at $z = h$. The quantity pUz becomes the coefficient of eddy diffusivity.

Although Eq. (12) is not amenable to analytical solution for the case in point, it has been shown^{6/} that the distribution of ground-level concentration has the following form:

$$C_0(x) = A \frac{e^{-h/px}}{x^{1+\alpha}} \quad (13)$$

where A = constant

$$\alpha = \frac{v_s}{pU}$$

The function given in Eq. (13), and hence the settling rate, reaches a maximum at:

$$x_{\max} = \frac{h}{p(1+\alpha)} \quad (14)$$

and then decays to zero as $x \rightarrow \infty$. Values for x_{\max} are given in Table 2 based on values of p determined by comparing the two forms of the eddy diffusivity, yielding

$$p = ku_*/U \quad (15)$$

The constant A in Eq. (13) may be evaluated by equating the emission rate E to the integrated settling rate.

$$E = \int_0^{\infty} CoV_s dx = AV_s \int_0^{\infty} \frac{e^{-h/px}}{x^{1+\alpha}} dx \quad (16)$$

With the transformation $y = b/x$ where $b = h/p$, the above equation becomes

$$E = \frac{AV_s}{b^\alpha} \int_0^{\infty} e^{-y} y^{(\alpha-1)} dy = \frac{AV_s \Gamma(\alpha)}{b^\alpha} \quad (17)$$

where $\Gamma(\alpha)$ is the gamma function.

Similarly it can be shown that the mass fraction K of particles remaining suspended beyond some distance x is given by:

$$K = \frac{\Gamma\left(\alpha, \frac{b}{x}\right)}{\Gamma(\alpha)} \quad (18)$$

where the incomplete gamma function $\Gamma(\alpha, b/x)$ is defined as

$$\Gamma\left(\alpha, \frac{b}{x}\right) = \int_0^{b/x} e^{-y} y^{\alpha-1} dy \quad (19)$$

Table 2. DISTANCES TO POINT OF MAXIMUM SETTLING, x_{max} , CALCULATED FROM EQ. (14)

Injection height, h (m)	Roughness height, z _o (m)	Turbulence parameter, p	Friction u _* (cm/sec)	Values of α and x_{max} (m) by particle size									
				30 μ m		20 μ m		10 μ m		5 μ m		1 μ m	
				α	x_{max}	α	x_{max}	α	x_{max}	α	x_{max}	α	x_{max}
1	0.01	0.0347	30.0	0.564	18.4	0.251	23.0	0.0625	27.1	0.0157	28.4	0.00062	28.8
	0.05	0.0534	36.4	0.465	12.8	0.207	15.5	0.0515	17.8	0.0129	18.5	0.00052	18.7
	0.10	0.0695	40.0	0.423	10.1	0.188	12.1	0.0469	13.7	0.0118	14.2	0.00047	14.4
	0.50	0.2308	52.2	0.324	3.27	0.144	3.79	0.0359	4.18	0.0090	4.29	0.00036	4.33
3	0.01	0.0281	30.0	0.564	68.3	0.251	85.3	0.0625	100.5	0.0157	105.1	0.00062	106.7
	0.05	0.0391	36.4	0.465	52.4	0.207	63.6	0.0515	73.0	0.0129	75.7	0.00052	76.7
	0.10	0.0470	40.0	0.423	44.9	0.188	53.7	0.0469	61.0	0.0118	63.1	0.00047	63.8
	0.50	0.0893	52.2	0.324	25.4	0.144	29.4	0.0359	32.4	0.0090	33.3	0.00036	33.6
	1.00	0.1456	60.0	0.282	16.1	0.125	18.3	0.0312	20.0	0.0078	20.4	0.00031	20.6
10	0.01	0.0232	30.0	0.564	276	0.251	345	0.0625	406	0.0157	424	0.00062	431
	0.05	0.0302	36.4	0.465	226	0.207	274	0.0515	315	0.0129	327	0.00052	331
	0.10	0.0347	40.0	0.423	203	0.188	243	0.0469	275	0.0118	285	0.00047	288
	0.50	0.0534	52.2	0.324	141	0.144	164	0.0359	181	0.0090	186	0.00036	187
	1.00	0.0695	60.0	0.282	112	0.125	128	0.0312	140	0.0078	143	0.00031	144

The above analysis assumes that particles of all sizes are uniformly responsive to turbulent diffusion. More realistically, the time constant of particle response to vertical velocity fluctuations increases with increasing aerodynamic particle size.

In studies of the vertical flux of particulates over an agricultural field undergoing wind erosion, Gillette et al.^{2/} have characterized this phenomena in terms of the ratio V_s/u_* . If settling velocity is small compared to the root mean square velocity fluctuation, i.e., $V_s/u_* < 0.1$, the particulate is dispersed as a gas. On the other hand for $V_s/u_* \sim 1$, settling effects begin to predominate. Clearly, in the latter case, the settling distribution is more strongly focused around the distance X_p .

Case 3: Polydisperse particles, distributed injection height, turbulent atmosphere.

This case is treated by separately analyzing the dispersion of particles within narrow size ranges and injection height ranges and by superimposing the results. The analytical techniques to be used are those described above.

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FACTORS INFLUENCING EMISSIONS FROM FUGITIVE
DUST SOURCES

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FACTORS INFLUENCING EMISSIONS FROM FUGITIVE DUST SOURCES

by George A. Jutze and Kenneth Axetell

Emissions from fugitive dust sources are known to be highly variable over time and geographic area and dependent on a number of different parameters. While developing emission factors for fugitive dust sources during the past three years, PEDCo has attempted to identify the specific parameters which influence emission rates most and to derive general expressions describing the relationships between these parameters and resulting emissions (correction functions to the emission factors).

At least six different parameters have been found to substantially affect fugitive dust emission rates:

- ° soil type
- ° windspeed
- ° surface moisture
- ° precipitation
- ° vegetative cover
- ° traffic activity across the surface.

This paper discusses the theoretical basis for the relationship between each of these parameters and emission rate and presents a quantitative method for estimating the effect of each.

SOIL TYPE

Soil type affects almost all fugitive dust sources, since it is usually the native soil from the surface of the source that becomes airborne as the suspended particulate emission. Two different characteristics of soil are most important in determining emission rates: (1) soil structure, or its resistance to breakdown into its component particles; and (2) soil texture, or the size distribution of the individual component particles. Such factors as cloddiness (soil cohesion), surface crusting, bulk density, and mechanical stability give a soil its "structure." Soil texture provides an indication of the potential amount of suspended particulate that can result if wind erosion or repeated mechanical disturbance of the soil causes the disintegration of the soil's natural structure.

The particle size distribution, or texture, of a soil is an important factor in defining a soil's structure. For example, soils containing a high proportion of silt form the most stable clods and most compact crust.¹ Since the two soil characteristics which affect emission rate are somewhat dependent on one another, soil scientists have generally used soil texture as the single basis for comparing the erodibilities and potential dust emission rates of different soils.

The standard measure of dust losses from soil surfaces is the erodibility index, I , in units of ton/acre/yr. This index estimates the relative erodibility of different soil surfaces, not the actual quantities erodible under field conditions. The erodibility indexes for different soil textures are shown in Table 1.

In previous fugitive dust emission factor development, either the erodibility index or the percent silt was used as the correction function to account for different soil

Table 1. ERODIBILITY AND AVERAGE PERCENT SUSPENDED
PARTICULATE-SIZED PARTICLES BY SOIL TYPE

Soil texture	Erodibility index, ton/acre/yr ^a	Aggregates < 0.05 mm, percent by weight ^b
Sand	220	0.9
Loamy sand	134	1.0
Sandy loam	86	2.1
Clay	86	0.8
Silty clay	86	0.8
Loam	56	6.6
Sandy clay loam	56	4.1 ^c
Sandy clay	56	1.0 ^c
Silt loam	47	4.1
Clay loam	47	2.5 ^c
Silty clay loam	38	4.1
Silt	38	0.8 ^c

^a Source: D. G. Craig, J. W. Turelle. Guide for Wind Erosion Control on Cropland in the Great Plains States. U.S. Department of Agriculture, Soil Conservation Service. July 1964. Table 1.

^b Source: W. S. Chepil. Soil Conditions that Influence Wind Erosion. U.S. Department of Agriculture, Agricultural Research Service and Kansas Agricultural Experiment Station, Washington, D.C. Technical Bulletin Number 1185. June 1958. Table 1.

^c Estimated from most similar soil types.

types.^{2,3,4} However, both of these parameters have shortcomings. The erodibility index estimates the relative soil loss for different soil types, but does not adequately account for the greatly varying percentages of that total soil loss which are in the suspended particulate size range and thus available to become airborne. Example percentages are also shown in Table 1. Most of the eroded soil is suspended for a short distance and then resettles, or is pushed along the ground. Movement of larger soil particles near ground level is referred to as saltation or surface creep.

The use of silt content as the correction factor is an attempt to account for the amount of soil in the suspended particulate size range, but it does not consider the structural resistance of silty soils to breakdown into the individual particles.

Comparative particle size ranges for different soil types and soil movements are shown in Figure 1. The particle size limits shown in the figure are approximate--they depend on particle density, shape, and surface forces as well as size. Loam, the fourth soil texture used in the classification system, is a mixture of clay, silt, and sand.

It is recommended that the relative dusting potential of different soil types be estimated from the product of their erodibility index and average percent of suspended particulate-sized particles (less than 0.05 mm):

<u>Soil texture</u>	<u>Correction weighting factor</u>
Sand	1.98
Loamy sand	1.34
Sandy loam	1.81
Clay	0.69
Silty clay	0.69

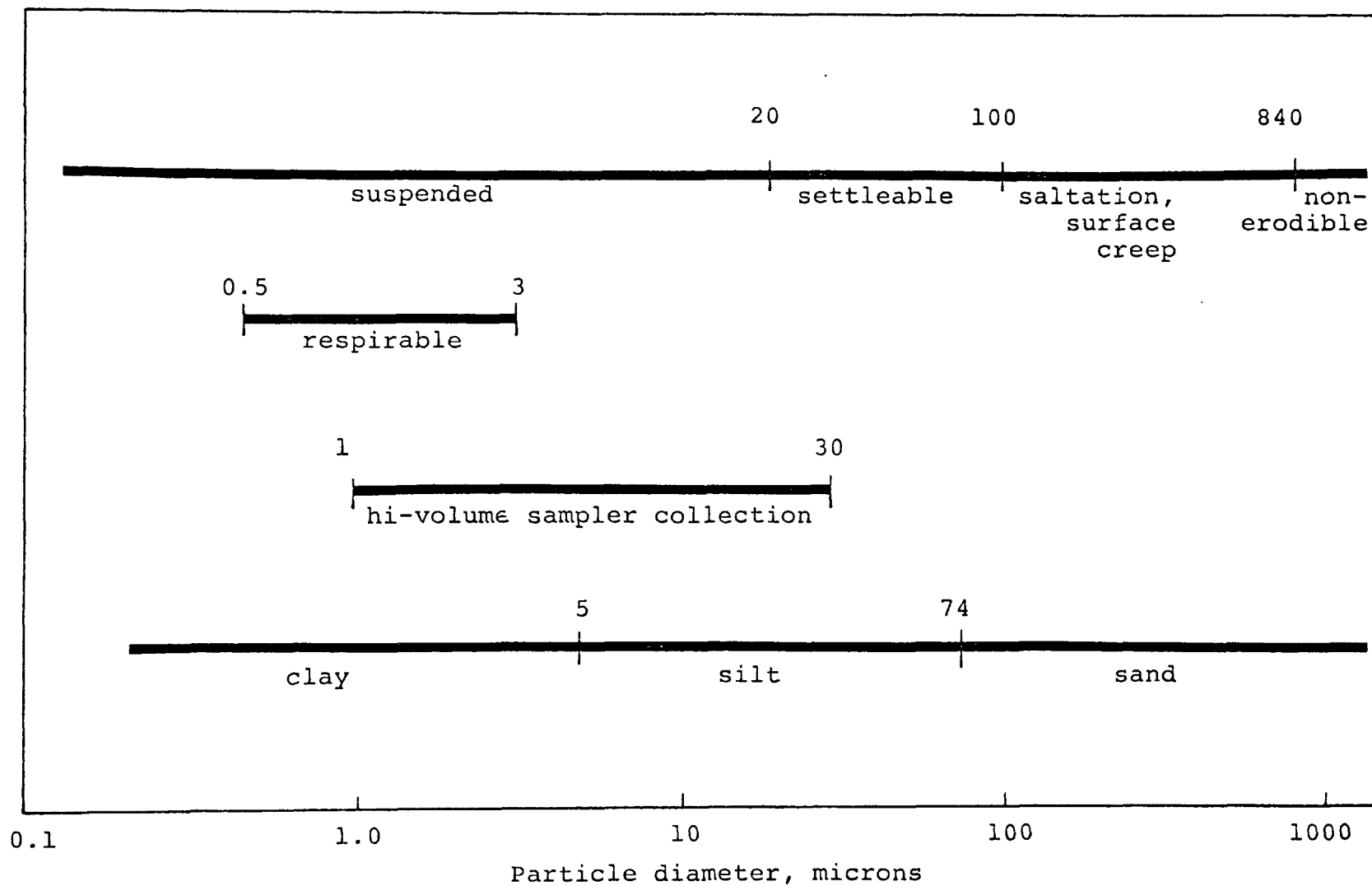


Figure 1. Particle size ranges for soil types and soil movements.

<u>Soil texture</u>	<u>Correction weighting factor</u>
Loam	3.70
Sandy clay loam	2.30
Sandy clay	0.56
Silt loam	1.93
Clay loam	0.94
Silty clay loam	1.56
Silt	0.30

The normal crusting characteristics of the soils are considered in their erodibility indexes. However, under certain conditions natural crusting can be enhanced by surface treatment with chemicals, compaction, or controlled watering. A soil surface that is well crusted loses dust at a rate about one-sixth that of the same soil in a non-crusted state.¹ If the surface is not completely crusted, or if the crust is weathered or damaged by traffic, the emission rate will be intermediate between an unprotected and a well-crusted surface.

A study of particle size distributions in the atmosphere indicated that these distributions are quite similar in all regions and appear to be independent of the prevailing soil types in the region (except in the case of dust storms).⁵ This conclusion is certainly not in conflict with the assumption made in the above discussion that emission rate from a soil surface is a function of the percent of soil particles in the suspended particulate size range. Essentially, the same sizes of particles appear to contribute to atmospheric particulate concentrations regardless of the size distribution in the parent soil.

Soils undergo seasonal changes in their erodibility as a result of biological activities, alternate wetting and drying, and freezing and thawing. These actions decrease

cloddiness and mechanical stability of the soils in winter in all areas where the soil is moistened at least occasionally and increase cloddiness and mechanical stability during the summer.¹ The amount of seasonal change in erodibility is also a function of soil type.

WINDSPEED

Windspeed affects emission rates primarily for the wind erosion sources, e.g., cleared land and agriculture.

Several investigators have found that when windspeed is greater than that required to barely move the soil, the rate of soil movement is directly proportional to the friction velocity cubed.⁶ The friction velocity at the surface is proportional to windspeed measured at a height above the ground, so the rate of soil movement is also proportional to the ambient windspeed cubed after it attains some minimum or threshold speed necessary to initiate movement.

One investigator reported threshold speeds to range from 13 to 30 miles per hour (mph) at 1-foot height, depending on the history of the field.⁷ Other investigators have found threshold velocities as low as 9 and 11 mph at about the same height.^{8,9}

The threshold velocity is influenced by the size of soil particles on the surface, and is lowest for aggregates of 0.10 to 0.15 mm in diameter. As shown in Figure 2, the threshold velocity increases with either an increase or decrease in the size of particles outside this range. The high resistance of fine dust particles to erosion by wind appears to be due partially to cohesion but mainly to the fact that the particles are too small to protrude above a laminar and viscous layer of air close to the surface of the ground. Fine dust is lifted from the ground mainly by impacts of larger grains, which are more erodible because

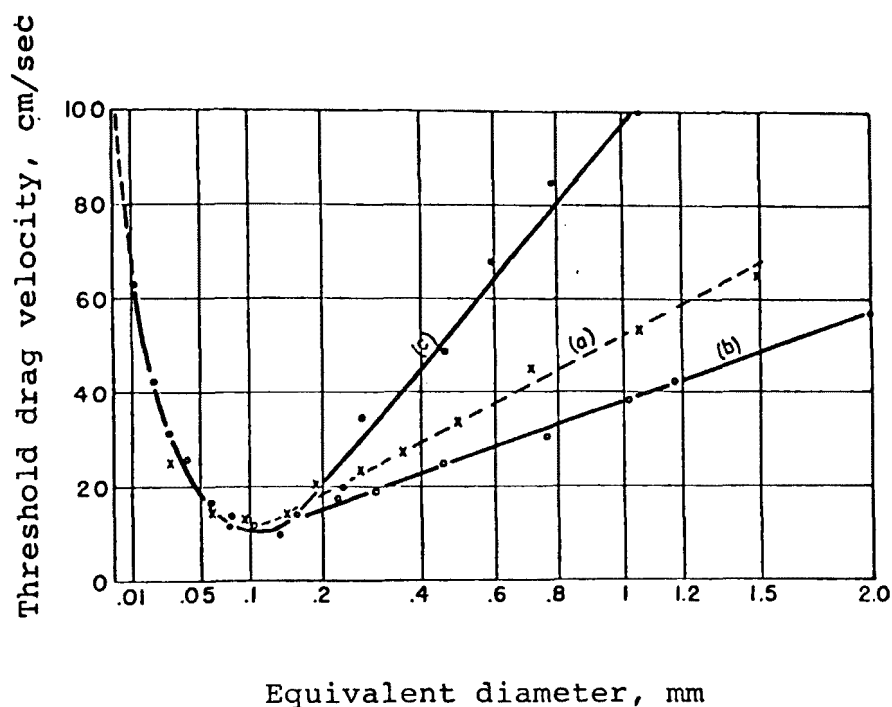


Figure 2. Relation between particle size and threshold drag velocity.

- (a) Sieved fractions with ratios of maximum to minimum diameters of 1.7.
- (b) Sieved fractions ranging in particle size from fine dust to diameter indicated by data point.
- (c) Soil containing 15 percent non-erodible clods.

Source: W. S. Chepil. Soil Conditions that Influence Wind Erosion. U.S. Department of Agriculture, Agricultural Research Service and Kansas Agricultural Experiment Station, Washington, D.C. Technical Bulletin Number 1185. June 1958.

they protrude farther into the fast moving, turbulent currents of higher air.

Turbulence is as important as average forward velocity in lifting and transporting the soil. Thus, maximum momentary wind velocity and gustiness also are factors in determining the impact of windspeed on fugitive dust emission rates. All surface wind velocities over about 2 mph create some turbulence.¹⁰

One research report⁶ recommended that the rate of soil movement be estimated as being proportional to the windspeed cubed times its duration above the threshold speed. In practice, it is difficult to obtain measurements of the duration above a given windspeed, such as 13 mph, because climatological summaries only record one- or three-hour averages. These averages are normally less than 13 mph, but may contain significant periods of time when the windspeed is greater than 13 mph. To avoid this problem, U.S. Department of Agriculture (USDA) researchers have assumed that all locations have a similar windspeed frequency distribution and, therefore, that wind erosion potential in an area is proportional to the cube of the average windspeed for any specified period.

This assumption in turn creates a problem with lack of standardization in height and exposure of recording anemometers; i.e., an anemometer at 30 ft may record an average windspeed one to two mph higher than the same instrument at 20 ft height. Since the erosion rate is related to the cube of windspeed, this small difference causes a change of 30 to 100 percent in the resulting correction function.

The climatic factor, C, in USDA's wind erosion equation¹¹ is an available correction function that includes the average windspeed cubed term and also a term (Thornthwaite's precipitation-evaporation index) to account for surface moisture. Values for C have been compiled for the entire

country with the exception of the four southwestern states of California, Nevada, Arizona, and New Mexico. These C values are shown in Figure 3.

Recognizing the limitations to accuracy of any correction function that utilizes average windspeed data, the climatic factor is recommended as the correction function to reflect emission differences due to windspeed. Many available emission factors already employ this term.

SURFACE MOISTURE

The soil moisture content of a fugitive dust source directly affects the emission rate from the source. For any surface moisture content above air-dried, a distinct decrease in erodibility is observed.¹² Soil movement is reduced only slightly by the first moisture, but decreases more rapidly with additional moisture. At a moisture content approximately corresponding to the permanent wilting point for vegetation, soil loss is completely arrested. For different soil types, the percent water required to eliminate soil loss is:¹²

<u>Soil type</u>	<u>Moisture content, %</u>
Dune sand	1.28
Sandy loam	3.89
Silt loam	11.21
Silty clay	20.71

Average surface moisture contents of soils are not routinely measured. Instead, Thornthwaite's precipitation-evaporation (P-E) index, calculated from the sum of 12 monthly ratios of measured precipitation to measured evapotranspiration, is used as an indicator.

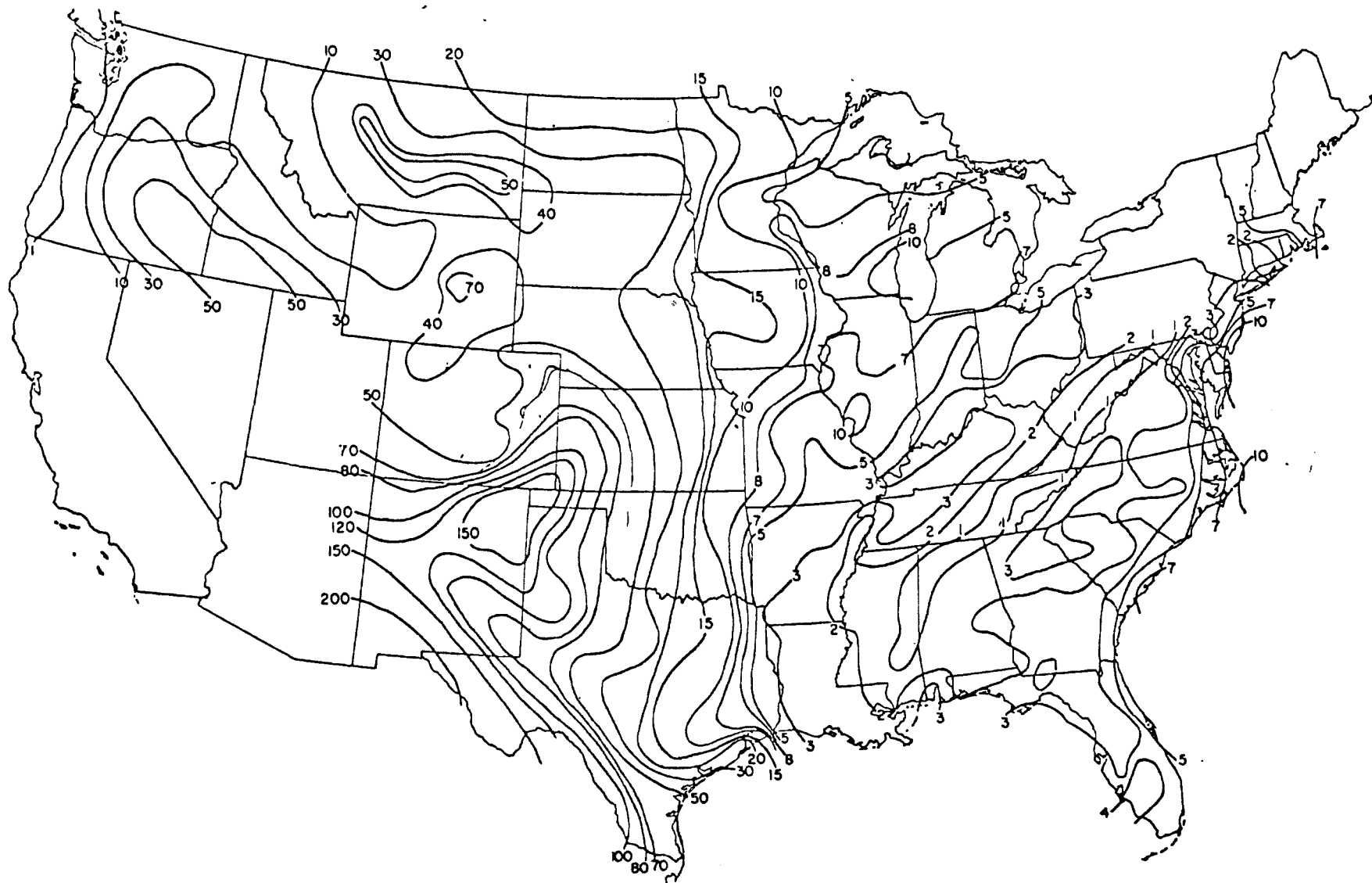


Figure 3. Climatic factors for use in the wind erosion equation.

Source: Armbrust, D. V. and N. P. Woodruff, 1968.

Empirical data show that soil loss varies inversely as the square of the P-E index. The P-E index value squared is included in the C factor previously described in the WINDSPEED section. Again, the C factor is recommended for use because of its acceptance as a broadly applicable correction factor.

Because of large variations in monthly rainfall, the monthly P-E values do not give meaningful monthly climatic factors.¹³ Also, the P-E index is not precise enough to evaluate soil moisture conditions for periods as short as one month. The U.S. Department of Agriculture has not developed any seasonal variation function for surface moisture.

The correction factor for moisture content overlaps somewhat with that for precipitation. The distinctions made between the two are:

- ° Only average soil moisture for the entire year is considered in the surface moisture function, and this value is used to adjust the locally applicable emission factor to reflect emission rates with normal retained moisture.
- ° The correction factor for precipitation only applies to days with rainfall or snow cover and accounts for the total absence of dust emissions from very wet soil surfaces.

While different soil types stop dusting with greatly varying amounts of surface moisture (see data above), there is no information to indicate that surface moisture affects the particle size distribution of fugitive dust emitted from a particular source. In other words, surface moisture does

not sort an erodible soil by differentially retaining certain fractions.

PRECIPITATION

As long as a soil surface is noticeably moist, it will not dust, even with traffic or surface activity.^{1,12}

EPA's recommended emission factors for emissions from unpaved roads⁴ assume that no emissions occur on days when there is measurable precipitation or snow cover. Analyses performed by PEDCo^{2,3,14} and others¹⁵ on particulate concentrations near fugitive dust sources on days with rain and no rain showed that hi-vol readings on rainy days are generally about half of those on non-rain days. Since these hi-vol measurements include some contribution from point sources, conventional area sources, and influx background in addition to fugitive dust, it appears that the combination of large reductions in fugitive dust emissions and rainout (during actual periods of rain) account for this 50 percent reduction in ambient concentrations. It can be shown that in some cases fugitive dust still occurs on days with rain, and may be even higher than normal due to strong winds that sometime precede thunderstorms. However, on an annual or seasonal basis, the current assumption embodied in EPA emission factors of no emissions on days with rain seems reasonable.

In most soils, crust formation due to rainfall has no permanent effect in reducing fugitive dust emission rates.¹ In fact, small showers tend to smooth the soil surface, loosen some surface particles, and accelerate rather than alleviate soil movement by wind. Rains may also bring additional fines to the surface in areas that have been cleared or disturbed and cause a temporary increase in dusting after the surface dries. A related natural phenomenon, the seasonal deposition onto a lake bottom of fine

particles from water erosion and subsequent wind erosion of the dried lake bed, provides another example of the negative secondary effects of rainfall on fugitive dust emission rates.

Several attempts have been made to demonstrate that the dust emission rate in an area increases with the number of days since the last rain, as the surface becomes progressively drier (see SURFACE MOISTURE). This relationship has never been shown to be significant, possibly due to actions such as those described above. Therefore, the assumption is made that emission rate does not increase with time since rain.

The proposed method for incorporating the correction for precipitation is to count the number of days during the period of interest with rain and snow cover (using a National Weather Service climatological summary for a local station), convert this count to a percentage of the total days, and reduce the emission factor by this percentage. This method is directly adaptable to estimating seasonal variations.

VEGETATIVE COVER

Ground cover primarily affects fugitive dust generated by wind erosion. Vegetation reduces wind erosion losses in two different ways. First, it absorbs some of the drag at the surface and decreases the wind velocity locally. Secondly, the roots of the plants act as a soil binder. The retained moisture necessary to support shallow-rooted vegetation also reduces the rate of soil loss.

The initial ground cover on a field usually provides more protection than additional increments of vegetation.¹ The relationship between the amount of vegetation and wind erosion rate is best quantified in the USDA wind erosion equation, which has been developed with over 30 years of experimental data.

The amount of vegetation is expressed in lb/acre of air-dried residue in the wind erosion equation. For ground cover more than about 250 lb/acre, additional vegetation reduces soil loss approximately linearly. Depending upon the potential for wind erosion in a particular geographic area, ground cover of 1000 to 2000 lb/acre are required to essentially eliminate soil loss.

The amount of vegetation obviously changes seasonally. Due to the wide range in seasonal changes for different plants, it is difficult to quantify this factor for use as a seasonal correction function. Deciduous trees and bushes are reported¹⁶ to lose 20 to 40 percent of their effectiveness when they are defoliated in the winter. This range may provide a usable estimate of average seasonal variations due to vegetation--a 20 percent increase from annual emission rates during winter and a 20 percent reduction during summer for sources with ground cover.

TRAFFIC ACTIVITY

Traffic movement over unpaved surfaces causes dust to be thrown into the air at contact points between the vehicle and the surface or pulled into the air by the vehicles' wakes. Also, traffic may break protective surface crusts and leave the soil more susceptible to subsequent wind erosion. Thus, surface activity has a direct, immediate effect on emission rates and an indirect, longer term impact.

The fugitive dust emitted by traffic movement is related to the amount of traffic or activity and its average speed. For many source categories, such as road shoulders and construction, it is difficult to find an activity parameter for which data are generally available. For sources with automotive traffic (unpaved roads, unpaved parking

lots, paved streets, and street sanding for snow control), the emissions are assumed to be directly proportional to vehicle miles of travel (VMT) even though there would be a small amount of emissions from these sources with no traffic. For agricultural tilling, the number of tilling operations per year on the fields is used as the measure of activity. For aggregate storage operations, traffic levels in the storage area are simply classified as active or inactive.

Several studies^{2,3,17,18} have examined the effect of average vehicle speed on emission rates from unpaved roads and have variously proposed that the emission rate is linearly dependent on,² a function of the square of,¹⁷ or exponentially related to³ vehicle speed. Since these analyses were each based on data for only three or four different speeds, it could be anticipated that many different curves would approximately fit the experimental data points. The important conclusion is that all these studies show emission rates increase with higher vehicle speeds throughout the normal operating range of the vehicles. Within the range of accuracy of the basic emission factor and the experiment designs it may be assumed that the actual relationship and the resulting correction function should be linear. The current EPA-recommended emission factor for unpaved roads incorporates a linear correction for average speed.

The only study¹⁹ identified which investigated the effect of speed on emissions from paved streets concluded that the relationship is also linear. However, the availability of material on the road surface for resuspension may be a limiting factor on the emission rate from this source. More data are needed to establish this correction factor.

A speed correction function for agricultural tilling has also been reported.^{2,20} However, most farm implements

are designed to operate over a narrow speed range and, as a practical consideration, there are usually no means of obtaining data on actual operating speeds when performing an emission inventory. Similar situations exist for vehicular traffic at construction sites and aggregate storage areas. Therefore, it is recommended that speed correction functions not be developed for any source categories other than paved and unpaved roads.

For sources such as roads and construction sites that have continual traffic, it has been determined that most of the fugitive dust results directly from traffic movement. For plowed fields and cleared areas, the total emissions are due to a combination of surface traffic and wind erosion. Total agricultural emissions may be estimated by calculating the tilling and wind erosion components separately. A proposed emission estimation procedure for cleared areas utilizes the wind erosion equation, but it may be appropriate to add a specified percentage to the estimated emissions if it is determined that there is surface traffic over the cleared area. Correction values of five percent for occasional traffic (1 veh/day) and 20 percent for regular traffic (more than 5 veh/day) were calculated from the previous estimates of the relative impacts of traffic and wind erosion on unpaved roads.

Traffic activity over native surfaces tends to destroy the surface crust or layer of pebble-sized particles ("desert pavement") that normally shield the surface from further wind erosion. The increase in emissions due to this traffic is a function of the surface area with tire tracks and the extent of damage to the natural protection. As mentioned previously in the SOIL TYPE section, an uncrusted surface loses soil at a rate about six times as great as a completely crusted surface. Therefore, a correction function

for areas with off-road traffic could be calculated as follows:

$$\text{correction function} = 1 + 5 \left(\frac{\text{area with damaged surface}}{\text{total area}} \right) \left(\frac{\text{fraction of crusting lost}}{\text{in disturbed areas}} \right)$$

Seasonal variations in a traffic correction function are dependent primarily on seasonal variations in activity (VMT, plowing operations, etc). The decision of whether to apply seasonal corrections should be based on the availability and quality of such data.

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STUDY OF THE EFFECT OF ASBESTOS WASTE PILES ON AMBIENT AIR

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Mary Stinson**
Paul Ase*

ABSTRACT

The fabrication of asbestos products is a major industry involving about one million tons per annum of asbestos. Asbestos in the atmosphere is associated with increasing incidences of cancer in the general populace. One source of the atmospheric asbestos is fugitive emissions from the waste piles located throughout the United States. Individual sources of emissions from the steps involved in the formation of waste piles are considered. Methods available to control these fugitive emissions are discussed and estimates presented on the relative costs of mitigation.

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STUDY OF THE EFFECT OF ASBESTOS WASTE PILES ON AMBIENT AIR

Introduction

Waste dumps from asbestos product manufacturing operations are frequently located in high density population areas. Fugitive asbestos emissions are created as the material is transferred to the dump and also as the surface of the pile is eroded by weather action. At the present time, fugitive dust control techniques are seldom used to mitigate the asbestos emissions.

The lack of adequate emission control at asbestos waste dumps is regrettable since there is growing evidence that low levels of asbestos exposure may be harmful to health. Evidence from the literature is presented which suggests that industrial activity involving asbestos is leading increasing numbers of mesothelioma cases. Mesothelioma, or cancer of pleura, is a rare disease known to be caused by asbestos.

This paper discusses methods by means of which the fugitive asbestos emissions may be mitigated. The technology which has been developed and applied to other industries is considered for application to the case of asbestos emissions.

Asbestos Waste Dumps

The asbestos industry is not a small industry; world consumption approaches five million tons per annum, while in the United States the amount is nearly one million tons per annum (see Table 1 for 1973 figures). Chrysotile asbestos accounts for approximately 96% of all the asbestos used in the United States.

Asbestos cement products account for 70% of the total United States usage. Products include asbestos cement pipe, asbestos cement siding, asbestos cement shingles, asbestos cement wallboard, and insulation products. It is estimated that there is between 5-10% of the product material dumped

Table 1
 ASBESTOS PRODUCTION, 1973
 (Minerals Yearbook)

	<u>Short Tons</u>
World Production	4,598,000
U.S. Production	150,000
U.S. Consumption	876,000
Chrysotile	839,200
Crocidolite	18,000
Amosite	4,300
Anthophyllite	1,200

as scrap every year. The scrap material is about 10% of fine material collected from baghouses and 90% of coarse scrap from cuttings and drillings and from products which have failed quality assurance tests (Table 2). Thus, the total annual scrap from asbestos cement operations alone may be calculated. If 1,000,000 tons of asbestos are used per annum, and of this 70% is for asbestos products, and there is a 7.5% scrap rate, then the total waste asbestos dumped is:

$$1,000,000 \times \frac{70}{100} \times \frac{7.5}{100} = 52,500 \text{ tons}$$

Assuming that the average asbestos cement product contains 25% asbestos, then the total asbestos cement waste is:

$$52,500 \times \frac{100}{25} = 210,000 \text{ tons}$$

Thus, the nation faces an annual problem of disposing of 210,000 tons of waste material containing hazardous asbestos.

The Hazardous Nature of Asbestos

Recognition of the hazardous nature of asbestos is relatively new. Although asbestos is associated with various cancers of the stomach, colon, etc., there is no doubt of its effect in producing two illnesses, asbestosis and mesothelioma.

Asbestosis is a non-malignant fibrosis of the lung and is only found among asbestos workers with a relatively heavy exposure level. Asbestos was first observed by Murray (1) in 1907. In 1930, Mereweather (2) gave a detailed description of the disease, and this led to the 1931 United Kingdom regulations on asbestos usage. Similar regulations were issued in the United States in 1938 following the studies by Dressen, et al. (3).

Mesothelioma, or cancer of the lung, was not fully recognized as a disease until 1960 when Wagner, et al. (4) published their work on the South African mine areas. They

Table 2
ASBESTOS CEMENT WASTE

Dust from baghouse collectors and sweepers	10%
Aggregates from breakage, cuttings, and drillings	90%
Waste pH	~12.5
Waste composition:	
Cured Portland cement	40 to 55%
Quartz silica sand	24 to 33%
Asbestos	15 to 35%

cited 33 cases of mesothelioma and, of these, 17 were not occupationally exposed. After 1960, the standard medical textbooks, e.g., Willis (5), were changed to include mesothelioma; until this time mesothelioma had been described as a metastasis from a primary site.

A number of studies have shown that asbestos is ubiquitous to the ambient air [Selikoff and Nicholson, 1970 (6); Holt and Young, 1973 (7)]. Other researchers have shown asbestos is commonly found in the lungs of urban dwellers [Um, 1971 (8)].

Case history studies in various countries have given increasing evidence of mesothelioma occurring in people non-occupationally exposed to asbestos. Newhouse and Thompson (9) in 1965 reported in England 76 cases of mesothelioma; only 40% had occupational exposure. Lieben and Pistawka (10) in 1967 working from Pennsylvania hospitals discovered 42 mesothelioma cases; only 24% of these were occupationally exposed. A further 24% could not be established, while 52% were not occupationally exposed. Borrow (11) in 1967 studied mesothelioma cases in the town of Manville, New Jersey (population 15,000), the site of a large asbestos products plant. Over a three-year period, 17 mesothelioma cases were found with two being non-occupationally exposed. Bohlig (12) in 1969 reported his study of the population surrounding a German asbestos plant. He found 319 mesothelioma cases between 1958 and 1968. He was able to follow the case histories of 119 and found only 46% of these were occupationally exposed to asbestos.

Much further research work is needed to answer the many questions raised concerning asbestos exposure and health. However, enough evidence is available to make it mandatory that modern technology be applied to limit the emission of asbestos to ambient air. Such techniques will be discussed in the following pages.

Utilization and Disposal Options

There are a number of options available for the utilization of asbestos wastes, Table 3. The first option to be considered is the re-use of the material. The industry makes every attempt to re-use asbestos cement waste by recycling it into the production line. However, there is a limit on the amount that can be recycled set by the exacting standards on the quality of the product. This is particularly true for asbestos cement pipe where every section of pipe is tested for its ability to withstand pressure and also its ability to withstand flexing.

Attempts have been made to use the waste material in building blocks. It has been shown that asbestos cement product waste can be fabricated into bricks which meet the standards of the ASTM C-90 and the ASTM C73-67 specifications. To date, asbestos waste has not been used commercially to produce these bricks because of the fear that a health hazard might be associated with such a product.

Another suggestion for a use of the waste material is to mix it with acid waste from other mining operations; the hope being that the neutralized waste would be more suitable for reclamation and plant growth.

Waste alteration by thermal or chemical decomposition has also been suggested. Chrysotile asbestos when heated above 600°F decomposes due to loss of water of crystallization to give forsterite and talc, both of which are thought to be harmless amorphous particles. Chemical leaching has been found to remove the magnesia from the crystal structure of chrysotile asbestos leaving behind the silica skeleton. While both thermal and chemical decomposition is possible, it is economically impractical.

The only viable alternate to waste re-use or alteration is dumping. The methods which may be applied to mitigate the emissions from dumping operations will now be considered.

Table 3

UTILIZATION AND DISPOSAL OPTIONS

Waste Re-use	-- recycle, cement blocks, steam cured bricks, waste neutraliza- tion with acid wastes
Waste Alteration	-- thermal decomposition, chemical leaching
Waste Dumping	-- waste aggregates, waste fines

The Dumping of Asbestos Wastes

The dumping of asbestos waste creates fugitive emissions at a number of points. The flow sheet for the dumping of waste materials is shown in Figure 1. The sources of asbestos emissions are given in Table 4 along with an estimate of the relative significance of each emission source. It should be noted that in these figures are not emissions rates but rather the relative magnitude of the total emissions set out such that the importance of each source may be assessed. Thus, even though the crushing and leveling is observed to have a high emission rate, the operation is of such a short time that its total contribution to the fugitive emissions is relatively minor. Obviously then, the major effort in reducing emissions should concentrate on the dump rather than the dumping, crushing, and leveling operations.

Emission Control Options

In Plant Options

The best method of controlling the dust is at the source, before they ever become fugitive emissions. Fine waste can be slurried, agglomerated, bagged, or at least wetted down. Larger waste material or aggregates may be crushed, washed free of fines, and kept wet. The use of various water-additive agents will extend the effectiveness of wetting techniques.

Dump Emission Control

Control of the emissions from waste dumps includes physical, chemical, and vegetative.

Physical include the use of physical barriers including straw, bark, or gravel applied directly to the surface of the dump. Other physical methods involve the judicious placement of wind breaks or banks of trees, placed in the direction of the prevailing winds, which protect the dump from wind erosion.

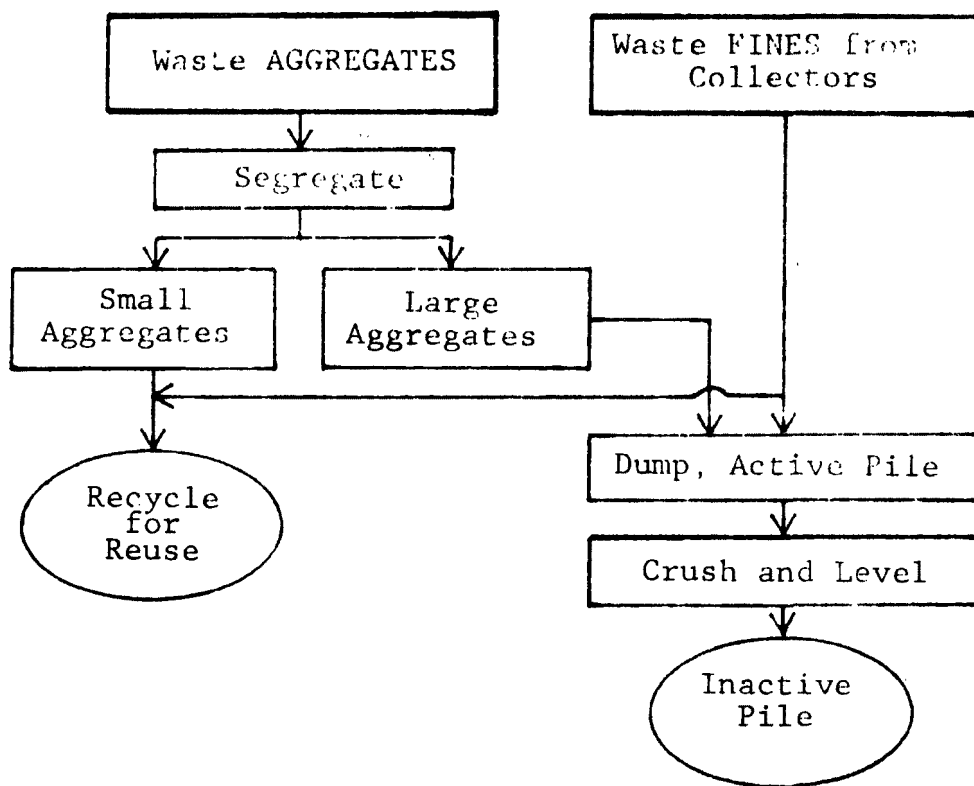


Figure 1
ASBESTOS CEMENT WASTE DISPOSAL

Table 4
WASTE DUMPING EMISSION SOURCES

Transfer Emissions	-- dumping, crushing, and leveling
Dump Emissions	-- active pile weathering, inactive pile weathering
Emission rate estimates:	
Fines dumping	22%
Aggregate crushing and leveling	6%
Active pile weathering	60%
Inactive pile weathering	12%

Chemical binders are finding increasing utility in reducing erosion from soil banks. The selection of the binder is important and the ideal binder would have the following properties:

- Low application cost
- Water soluble
- High bondability to the particles under consideration
- Long life and stability
- Resistant to heat and cold
- Non-toxic
- Biodegradable
- No water pollution problems from drainage water
- Easy to clean from application devices
- Effective in low dilutions

Obviously no one binder material is able to score highest in each of the above categories. For the purposes of the present study, about 30 binders were considered (some of these are listed in Table 5). Of these, five were considered in greater detail. Field experiments were conducted using Coherex which, although it was not the most efficient binder in terms of stabilizing soil, it did have the best overall properties including water solubility, biodegradability, and resistance to leaching once applied.

Vegetative covers, if properly applied and maintained, offer the surest, most permanent method of eliminating emissions caused by surface erosion. A cover of 6 to 12 inches of soil vegetated with fertilizer, seed, and mulch, and regularly watered is generally found to be effective. It is important to overcome the problems inherent in the waste material such as the lack of plant nutrients and microbial populations, and also the pH of the waste. For this reason, plants cannot generally be grown directly on the waste, and

Table 5
MATERIALS

<u>Product</u>	<u>Chemical Identification</u>	<u>Supplier</u>	<u>Cost, Dollars/lb</u>
Amine D Acetate, 50S	Stabilized abietyl amine	Hercules, Inc. Hattiesburg, Miss.	0.625
Polyrad 1110A	High molecular-weight amine ethylene oxide adducts	Hercules, Inc. Hattiesburg, Miss.	0.780
Vinsol Emulsion	Water emulsion of aliphatic resin	Hercules, Inc., Kalamazoo, Mich.	0.126
Defloc 50	Cationic polymer	Hercules, Inc., Milwaukee, Wisc.	0.170
Abitol	Hydroabietyl alcohol	Hercules, Inc., Burlington, N.J.	0.8375
Paracol emulsion	Wax-rosin emulsion	Hercules, Inc.	
Piccolyte Dipentene	Polyterpene adhesive resin	Hercules, Inc.	
Reten 421	Anionic acrylic polymer	Hercules, Inc., Hopewell, Va.	1.40
Neuphor 100	Anionic emulsion	Hercules, Inc., Milwaukee, Wisc.	0.2335
Kymene 557	Cationic polyamide-epichlor-hydrin resin	Hercules, Inc.	
Landlock XA2440	Adhesive binder	3 M Co., St. Paul, Minn.	
Latex M145 or M166	Latex binder	Dowell Div., Dow Chemical Co., Tulsa, Okla.	2.00/gal
Elvanol	Polyvinyl alcohol	E.I. Du Pont de Nemours & Co.	
Vinylac	Polyvinyl acetate, tackified dispersion	Borden Chemical Co.	0.51
ARQUAD 2HT	Quaternary ammonium compound	Armak Chemicals Co.	

Table 5 (continued)

<u>Product</u>	<u>Chemical Identification</u>	<u>Supplier</u>	<u>Cost, Dollars/lb</u>
ARQUAD 2S	Quaternary ammonium compound	Armak Chemicals Co.	1.09
Ethomeen T/12	Tertiary aliphatic amine	Armak Chemicals Co.	0.725
Curde Amine	Amine compound, unpurified	Armak Chemicals Co.	0.340
Krilium CRD-186	Vinyl acetate/maleic acid	Monsanto Chemical Co., St. Louis, Mo.	
Sodium alginate	Sodium alginate	Rolakem Co., Teaneck, N.J.	2.00
Polyacrylic acid	Polyacrylic acid amine	Rohm & Haas Co., Philadelphia, Pa.	
Superfloc 16	Flocculant	American Cyanamid Co., St. Louis, Mo.	
Coherex	Resinous binder	Witco Chemical Co., Hammond, Inc.	0.36/gal
Rezosol 5411B	Cationic resin emulsion	E. F. Houghton Co., Philadelphia, Pa.	0.245/gal
Dextran	Dextran	Howard Hall Co., Cos Cob, Conn.	4.00

a soil layer must be applied. It is generally found to be advisable to use soil types which fit in with the local area and also to use plant seeds of indigenous plants.

Costs of Various Control Options

To estimate the cost of controlling emissions from transfer operations at the waste and from the waste dump itself, a hypothetical plant was considered. The plant disposed of 13.2 metric tons of reject product and other waste aggregates per day; in addition, its plant disposed of 0.9 metric tons of baghouse waste fines per day.

Fugitive emissions were assumed to result from four basic operations:

- Daily dumping of fines onto "active" pile
- Crushing of reject pipe by bulldozer, once a month, on active pile
- "Weathering" of active piles
- "Weathering" of inactive piles

The emission rates associated with each source were based on reported fugitive dust emission rates and from IITRI field experiences.

Control techniques for reducing emissions from the waste disposal activities vary considerably in the annual costs and the emission reduction achieved. Thirteen basic control options were evaluated. They were considered separately and in combinations to ascertain the lowest cost methods for achieving emission reductions. Table 6 summarizes the results of this cost analysis.

The cost of controlling emissions from fines dumping was estimated for a water spray, water plus surfactant, pelletizing of fines, water slurry, water slurry with chemical binder, and bagging of fines. The bagging of the fines was the most efficient control scheme for the fines dumping operation (100% efficient) found in this analysis while a water

Table 6
SUMMARY OF CONTROL OPTIONS

Control Method	Total Annual Cost, \$	Percent Reduction in Emissions From:				
		Fines Dumpings	Aggregate Crushing	Active Pile	Inactive Pile	Total Emissions
1. Water Spray at Fines Dumping	2,800	10	--	--	--	2
2. Water and Surfactant at Fines Dumping	3,400	20	--	--	--	4
3. Agglomeration of Fines with Water	10,000	90	--	5	--	23
4. Agglomeration of Fines with Binder	13,000	90	--	25	--	35
5. Water Slurrying of Fines	4,100	85	--	--	--	19
6a. Chemical Binder with Water + 0.25% Binder	5,800	85	--	45	--	46
6b. Chemical Binder with Water + 0.20% Binder	5,400	85	--	27	--	35
6c. Chemical Binder with Water + 0.10% Binder	5,000	85	--	14	--	27
7. Bagging of Baghouse Fines	10,500	100	--	45	22	52
8. Chemical-Vegetative Control of Inactive Pile	3,380	--	--	--	90	11
9. Water Spray on Active Pile	3,570	--	--	50	--	30
10a. Chemical Stabilize Active Pile Once/ Week	8,970	--	--	90	--	54

Table 6 (continued)

<u>Control Method</u>	<u>Total Annual Cost, \$</u>	<u>Percent Reduction in Emissions From:</u>				
		<u>Fines Dumpings</u>	<u>Aggregate Crushing</u>	<u>Active Pile</u>	<u>Inactive Pile</u>	<u>Total Emissions</u>
10b. Chemical Stabilize Active Pile Once/ Month	3,970	--	--	80	--	48
11. Landfilling Active Pile Once/Month	8,700	--	--	73	20	46

spray at the dump site was the least efficient (10% efficient). The control costs for the fines dumping operation varied from \$2,800 per year (water spray) to \$13,000 per year (pelletizing).

The fugitive emissions from an active pile could be reduced with methods similar to those used to obtain fines control. Water spray, chemical stabilization, foaming agent, and landfill techniques were all considered as possible mitigation methods. The cost of these control methods varied from \$3,750 to \$8,970 per year. The emission reduction from the active pile ranged from 50% (water spray) to 90% (chemical stabilization of pile once per week).

A permanent cover on the inactive pile was required to prevent the source emissions from increasing each year as the size of the pile increased. A combination chemical stabilization-vegetative cover was assumed to be the method most likely to yield a permanent cover. The cost for developing the permanent cover on the inactive asbestos pile was calculated to be \$3,400 per year.

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It should be noted that the views and comments expressed in this paper are those of the authors and does not necessarily reflect those of the EPA.

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AN ASSESSMENT OF FUGITIVE EMISSIONS IN
THE PRIMARY ALUMINUM INDUSTRY

BY
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PRIMARY ALUMINUM INDUSTRY

The primary industry supplies all new, or virgin, aluminum, since aluminum is not produced as a by-product of other industrial activity.

It extracts chemically bound aluminum from its commercial ore, bauxite, and converts it to metal in two stages.

In the first stage, bauxite is treated by the Bayer Process to obtain aluminum oxide in a high state of purity.

In the second step, metallurgical grade alumina is converted electrolytically to the metal.

The aluminum industry, in common with other major materials producing industries, must process necessarily large amounts of raw materials by economic methods. In 1974 approximately 5 million tons of aluminum was produced in the United States. Since one pound of aluminum requires an input of 5.6 pounds of raw materials, starting with the refining of bauxite, the overall magnitude of the materials handling is obvious.

At each successive stage in the production of aluminum, the value of the product increases substantially. And with the increase in value, the incentive to conserve materials. But the incremental costs to control further losses beyond a certain level of control tend to rise rapidly and finally reach a point of extreme cost ineffectiveness.

FLOW OF RAW MATERIALS AND PROCESSES

Domestic production of bauxite supplies less than 8% of current U.S. demand for metallurgical grade alumina.

At the present time, domestic bauxite is actively mined only in Arkansas. Arkansas bauxite is produced mostly from underground mines. It is a fairly hard material and requires special processing because of higher than normal amounts of alumino-silicate materials.

Most other bauxite is a soft, clay-like deposit--not readily distinguished from the other soil in areas where it is found. It may be hauled by truck or rail from the mine site to a nearby alumina plant or to shiploading site for export.

Foreign sources of bauxite now account for somewhat more than 92% of U.S. alumina consumption--imported partly as bauxite, partly as refined alumina.

At the Bayer plant bauxite is unloaded from ships by bucket or clam-type crane, or in certain instances, by conveyor mechanisms, and transferred to storage buildings.

As needed, bauxite is transferred from storage to a crushing and milling operation. This step may be accompanied by blending of two or more bauxites from different sources to achieve desired properties.

The foregoing operations may be accompanied by the generation of dust, depending on the moisture content of the bauxite and weather conditions and on provisions for capture and control of dusts that may be operated at a particular plant. It should

be borne in mind in this regard that, with the one exception of an alumina plant operated in the Virgin Islands, the construction of all domestic alumina plants were begun before the late 1950's.

The remaining steps in the Bayer Process, until final calcining, are totally enclosed.

Bauxite is mixed with hot caustic and heated under pressure to dissolve hydrated aluminum oxide to produce a solution of sodium aluminate. This step is known as "digestion." Impurities --mostly iron oxide and free silica or quartz--remain essentially undissolved.

Insoluble impurities are removed by filtration.

The clear sodium aluminate solution is cooled, accompanied by precipitation of alumina-trihydrate.

The trihydrate filter-cake is calcined at 1200°C removing practically all chemically bound water and producing a metallurgical grade alumina of about 99.4% purity.

Other raw materials used in the Bayer Process are lime and caustic. The caustic is recycled to the process and is handled as a liquid throughout.

Today the production of primary aluminum relies exclusively on the Hall Process.

In the Hall Process, aluminum is produced by the passage of direct current through a cell containing alumina dissolved in molten electrolyte. The electrolyte is primarily cryolite. The molten cryolite is contained in a carbon-lined refractory box, called a pot. As electrolysis proceeds, molten aluminum

collects in the bottom of the cell. Electrical current passes through the electrolyte between a large block of carbon, suspended over and partially immersed in the electrolyte--this is the anode--and the cathode which is the pool of molten aluminum resting on the carbon lining. During the course of electrolysis, the anode carbon is gradually consumed by reaction with oxygen generated by the cell.

Thus, there are two major materials input to the manufacture of primary aluminum: alumina and anode carbon, prepared from petroleum coke.

Alumina and petroleum coke typically arrive at most aluminum smelters by rail, barge or ship.

Alumina may be unloaded from hopper cars pneumatically and transferred to storage bins referred to as "ore" silos.

The petroleum coke is moved to the carbon plant where it is stored until needed to manufacture pre-cast anodes or anode paste for Soderberg-type plants.

Distribution of alumina to the individual cells requires handling that tends to create some dust. Virtually all of this dust is settleable.

Likewise, also the preparation of anode carbon from coke involves operations which tend to create dust.

Cellroom and anode plant emissions are covered by current or proposed EPA point source and fugitive emission regulations and will not be further discussed.

NATURE AND SIGNIFICANCE OF FUGITIVE LOSSES

1. At the source of bauxite

Operations: Earth Moving
Loading
Transportation

Type of Materials: The bauxite contains principally hydrated oxides of aluminum with lesser, variable amounts of iron oxides, free silica, hydrated alumino-silicates, titanium dioxide, and other minor impurities.

Significance: Generation of dust depends very much on moisture content of ore--this, of course, depends on weather conditions. Generally speaking, bauxite dusts contain particles larger than 20 microns in diameter and are considered settleable--other than a potential nuisance in the immediate vicinity of bauxite mining operations, these dusts are considered to have a minor to insignificant impact beyond the locus of operations --the dusts are virtually indistinguishable in composition from endemic soils.

2. At the Bayer Plant

Operations: Unloading Bauxite
Transfer and storage of bauxite
Crushing and milling
Loading of calcined alumina

Materials: The composition of bauxite was just described. A bauxite may be very similar chemically and mineralogically, but usually not identical to native soils at the Bayer Plant. They may contrast somewhat in color.

They will support vegetation.

Metallurgical alumina is high purity aluminum oxide, snow-white in color.

Significance: Very little airborne material actually leaves the plant property; bauxite itself is a soil which will support vegetation. Bauxite can be thought of as the last stage of evolution in the

formation of laterite wherein most of the former constituents have been leached away over geologic time, leaving aluminum oxide.

Alumina is exceptionally inert, chemically and biologically, and is considered to have very little if any adverse effect on the environment.

3. At the Primary Smelter

Operations:	Unloading and transfer of alumina Distribution of alumina to the cells Unloading and handling of petroleum coke
Materials:	High purity alumina, petroleum coke is a pure form of commercial carbon; mechanical handling of P. coke produces coarse dust.
Significance:	From the calcination step in the production of alumina through the smelting of primary aluminum, about 99% of the alumina is converted to metal. This is a remarkable efficiency considering the many opportunities for losses: transportation and transfer, housekeeping, and oxidation losses due to formation of aluminum oxide skim and dross.

SUMMARY

Although the scope of presentation bars any detailed assessment of fugitive emissions at particular plants, it is possible to examine several factors that have a bearing on the matter:

- | | |
|----------------|--|
| - Siting | In most instances, alumina and primary aluminum facilities are situated well away from population centers |
| - Prior Action | The aluminum industry has consistently instituted measures to reduce fugitive losses. |
| - Character | Fugitive dusts, represented by large particle size, tend not to leave the plant site. |
| - Chemistry | Calcined alumina is inert; the chemical composition and mineralogy of bauxite so strongly resembles that of native soils at many plants that it is very difficult to distinguish from soil derived dusts from other sources such as local agriculture. |

- The aluminum industry has spent considerable money and effort to contain fluoride emissions--these are the subject of EPA New Source Performance Standards, published recently. EPA Guidelines to the States for Existing Aluminum Plants are pending.
- Finally, because of potential litigation on the new source standards for aluminum, I have avoided reference to cell-room emissions or of emissions from any other source that might contain fluoride.

MEASUREMENT OF IRON FOUNDRY FUGITIVE EMISSIONS

by

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Presented at the

SYMPOSIUM ON FUGITIVE EMISSIONS:
Measurement and Control

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MEASUREMENT OF IRON FOUNDRY FUGITIVE EMISSIONS

INTRODUCTION

An iron foundry has traditionally been thought of as an unpleasant place to work; foundry workers may be exposed to dust, free silica, heat, chemical emissions, and noise during molding and casting.

Nonetheless, the foundry industry is one of the largest and most basic industries in the United States; it supplies components used in the manufacture of a great variety of products by other industries. The industry is the sixth largest of all U. S. manufacturing industries, according to the 1970 U. S. Census. In 1974 the annual U. S. casting production was about 22,000,000 tons with a direct product value of approximately \$13 billion.

This paper reports some of the experimental techniques and results employed to quantify point source emissions in the foundry. The work included characterization of general particulate, free silica, and chemical emissions from typical green sand foundry molds.

The detailed results of the chemical emissions portion of the program have been reported elsewhere.^{1,2*} Various organic materials are added to the sand mixture so that their thermal decomposition produces a reducing atmosphere in the mold, which is necessary for good casting quality. The gases or products of decomposition are then released into the foundry atmosphere. Adequate ventilation is necessary to prevent build-up of noxious gases such as carbon monoxide, hydrogen cyanide, ammonia, methane, and other more complex species.

This ventilation is often in the form of forced air drafts that are exhausted directly from the plant without treatment. Design and

* Superscript figures refer to items in "References."

operation of these ventilation systems determine the actual concentrations of gases present in the working environment and also those released outside the plant. A fundamental knowledge of the point source emissions in the foundry allows estimations of the total amount of objectionable materials produced.

In addition to gaseous emissions, particulate emissions are also generated in the green sand foundry process. It is estimated that between 9 and 15 tons of sand must be processed for every ton of castings made.

An estimate of the particulate emissions generated during casting production in a typical foundry using sand molds is presented in Table I.³ The major plant areas that contribute are (a) molding, pouring and shakeout, (b) cleaning and finishing, and (c) sand conditioning areas.⁴ The total estimate of non-melting operations indicates that 115 lb. of emissions is generated for each ton of metal poured. Normal collection equipment reduces this to 60 lb/ton in the plant atmosphere, of which 51 lb. settles out. About six pounds of this total is released outside the plant.

In addition to the dust from the sand handling, the emissions from the shakeout and pouring operation include organic compounds of potentially harmful character. These most often are expected to condense on the particulate matter and thus are available for respiration. The types of compounds that are included in this portion of the emissions have also been reported elsewhere.⁵

Table I
Particulate Emissions Factors
from Non-Melting Operations

		(1)	(2)	(3)	(4)	(5)	(6)
		Emissions Generated	Normal Collection	Emissions to Foundry Environment	Settling Factor	Emissions Released to Atmosphere	Emissions to Atmosphere Nationwide*
Department	Operation	Lb./Ton Melt	Percent	Lb./Ton Melt	Percent	Lb./Ton Melt	Lb./Ton Melt
Scrap Yard	Raw Material Handling	.20	0	.20	80%	.04	.04
	Charge Make-up	.15	0	.15	80	.03	.03
	Charge Pre-heating	.20	10%	.18	40	.11	.01
Molding, Pouring and Shakeout	Molding	.50	0	.50	90	.05	.04
	Magnesium Treatment	5.00	10	4.50	75	1.12	.11
	Pouring	5.10	10	4.59	60	1.84	1.45
	Cooling	10.30	10	9.27	90	.93	.75
	Shakeout	32.20	60	12.80	90	1.28	1.02
Cleaning and Finishing	Shot Blast	15.50	99	.18	80	.03	.02
	Grinding	1.60	95	.08	80	.02	.01
	Annealing	.10	0	.10	30	.07	.03
	Painting	.02	95	—	40	—	—
Sand Conditioning	Dry Sand Handling	10.30	10	9.27	90	.93	.75
	Prepared Sand Handling	.50	0	.50	90	.05	.04
	Screening	10.00	20	8.00	90	.80	.64
	Mulling	20.60	60	8.24	90	.82	.65
	Drying and Sand Reclamation	1.50	60	.60	80	.12	.01
Core Department	Oil Sand Core Baking	.10	5	.05	0	.05	.03
	Shell, Hot Box and Cold Set Sand Cores	1.02	0	1.02	0	1.02	.20
Pattern Shop	Wood Patternmaking	.01	80	—	50	—	—
	Metal Patternmaking	.02	80	—	50	—	—
Total		114.92		60.21		9.31	5.83

Note: *Incidence factor has been applied to these emissions rates to reflect actual industry application of each operation.

SAMPLING PROCEDURES

The sampling procedure used to determine the point source emissions from typical foundry molds employed an exhaust hood and canopy that covered the source. This arrangement has been referred to as a quasi-stack method and it is shown schematically in Figure 1. An orifice plate and DC motor-driven exhaust fan in the stack were used to establish a known flow rate through the hood. The flow in the stack was made turbulent to insure a homogeneous sample.

The draft air volume was controlled with a rheostat on the fan motor and was set to maintain a constant pressure drop of 1/2 inch of water, as monitored with a manometer. This pressure drop corresponded to a volume flow of 35 cfm in the stack, or an air velocity of 50 ft/min over the mold surface. This velocity was made high enough to prevent significant losses of effluent by diffusion under the hood.

Gaseous emissions were periodically sampled by drawing the effluent into 250-ml evacuated bulbs. This technique is shown in Figure 2. These grab samples were taken at intervals starting immediately after the casting had been poured, while the castings cooled in the sand molds, and as the castings were separated from the sand in a simulated shakeout. These samples were analyzed by gas chromatography. This procedure allowed the preparation of a time-concentration profile.

A constant stream of gas from the stack was passed through dimpled bubblers containing specific absorbing reagents for the determination of average concentrations of gases such as ammonia and hydrogen cyanide that were present in low concentrations. These bubblers are shown in place on the stack in Figure 3. Figure 4 shows the metal

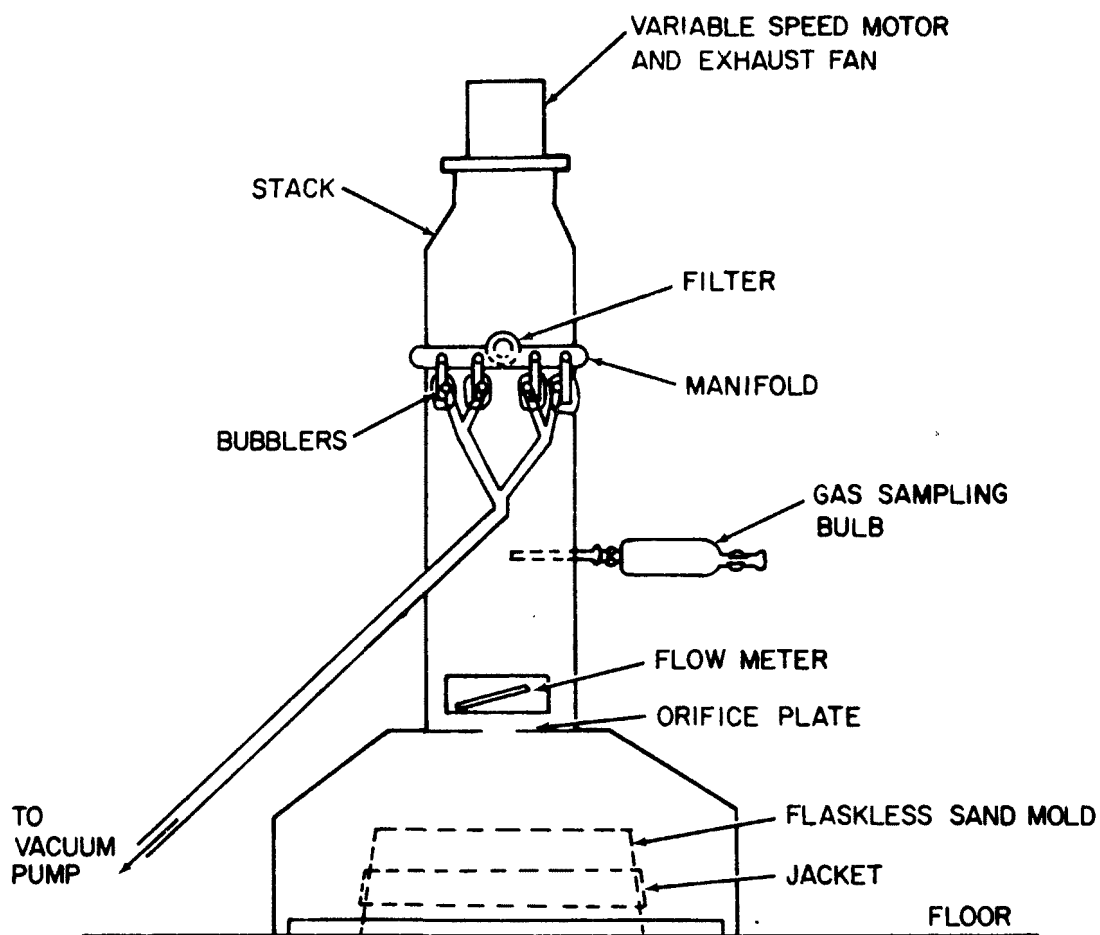


Figure 1. Schematic of portable gas sampling hood.

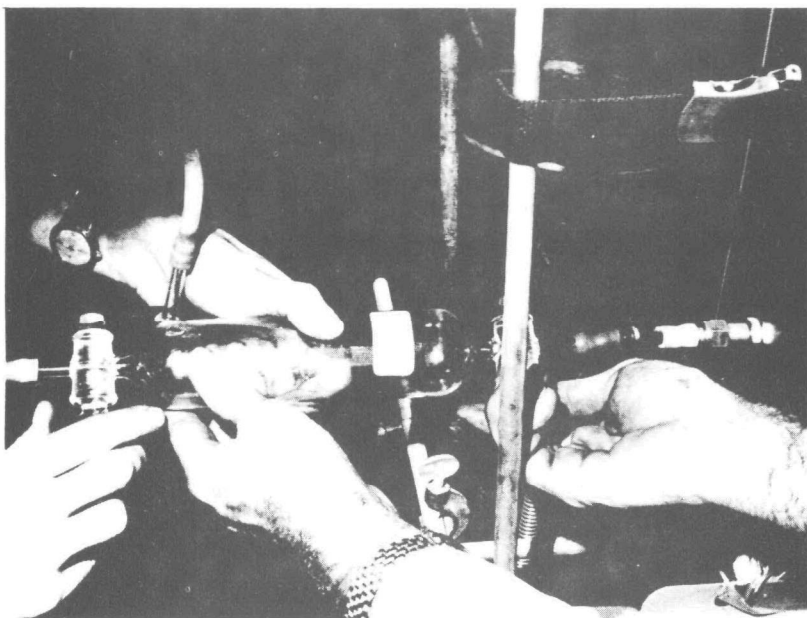


Figure 2. Taking a grab sample for gas analysis above an open mold.

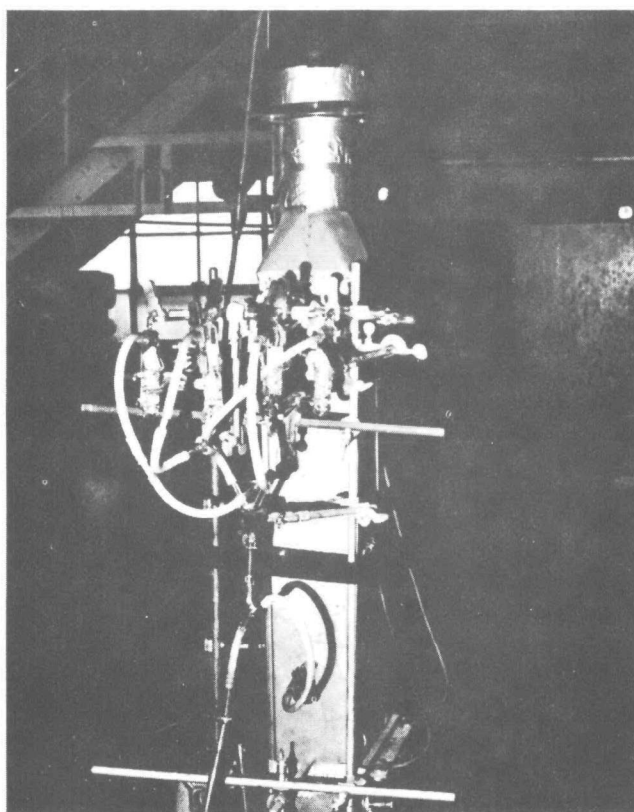


Figure 3. Top of sampling hood, showing bubblers for trace gas detection.



Figure 4. Pouring an open mold.

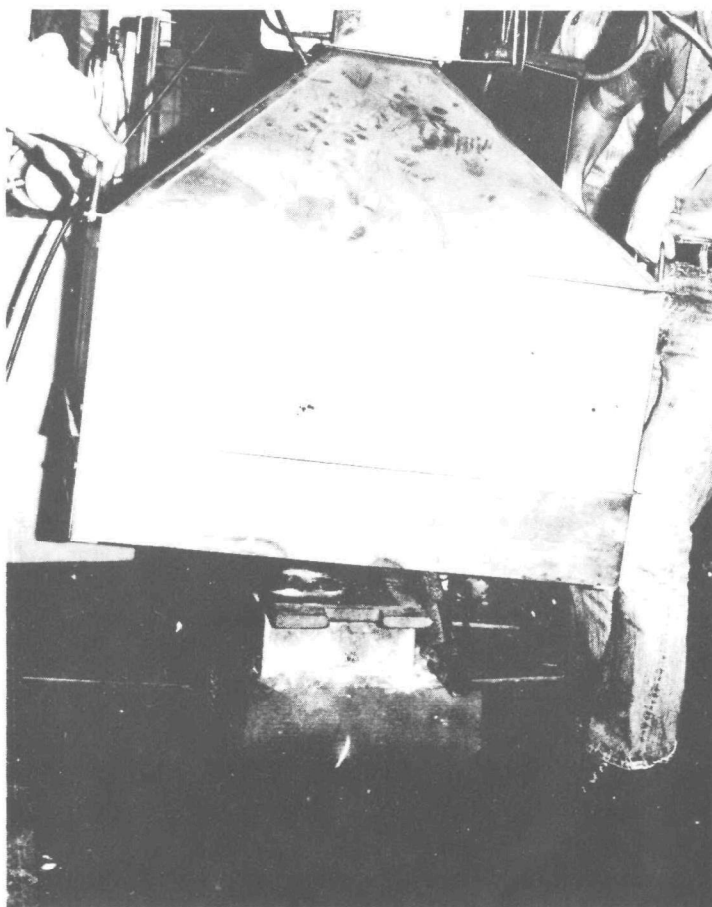


Figure 5. Placing the collection hood over an open mold. Note mold gases burning at the parting line.

being poured into a mold. Figure 5 shows the hood being positioned over the mold after the pour. Positioning the hood required approximately 15 seconds from the start of the pour.

The hood was also used to sample the air above the muller and shakeout bin in a pilot plant. In these tests, the hood was lifted into place over the operation of interest with a forklift, and then a polyethylene canopy was suspended to cover the entire operation.

Cascade impactors were positioned in the stack to obtain particle mass-size distributions over the range of particle diameters of 0.5 to 14 μm . Brink and Andersen designs of cascade impactors were used, depending on the dust loading of the atmosphere. The Brink impactor, which has a low flow rate, was used at high loadings. At low loadings, the Andersen impactor, which has a higher flow rate, was used to keep the sampling time reasonable. In either impactor, the particulate material entrained in the air is separated according to particle size by suitable orifice designs in the impactor, and deposited upon collector plates.

The Brink impactor uses aluminum foil plates that may later be dissolved in acid as the first step in the free silica determination by the Talvitie colorimetric method.⁶ By summing up the weights of the particles on all of the collector plates, and dividing the result by the volume of air drawn through the impactor, an average dust loading was determined. The Andersen impactor will also allow a dust loading to be calculated as well as size distribution, but the particulate matter may not easily be chemically analyzed.

Particles in the 0.3 to 1.0 μm size range were counted with an optical particle size analyzer. In operation, a sample of effluent from the hood was continuously removed, diluted with clean air, and

passed through an orifice and then through a collimated light beam. The amount of scattering caused by individual particles was measured with a detector tube. Since small particles scatter more light than large particles, the signal from the detector could be electronically analyzed to both size and count the number of particles in the gas stream.⁷ This allowed data in the form of concentration-versus-time to be obtained.

RESULTS

Gaseous Emissions from Green Sand Foundry Molds

Gaseous emissions from green sand molds were determined as described above by collecting emissions as a function of time after pouring and during shakeout. A constant rate of air flow of 35 cfm through the exhaust hood was maintained for these experiments. This air flow dilutes the effluent and permits partial oxidation of the combustible gases present in the effluent. This simulates normal foundry practice where air flow from mechanical ventilation is used and the burning of mold gases is observed.

Concentration-time plots of the carbon monoxide from green sand molds containing about 5% seacoal are shown in Figure 6. The castings were 4" cubes with appropriate ingates and pouring basins, which gave a total weight of approximately 30 lb. The castings were poured at a sand:metal ratio of approximately 3:1. The first peak in the concentration-time curve represents the carbon monoxide concentration in the effluent after pouring. The second peak represents the CO concentration when the mold was broken open.

The carbon monoxide concentration increased from a low value just after pouring to approximately 1900 ppm about five minutes

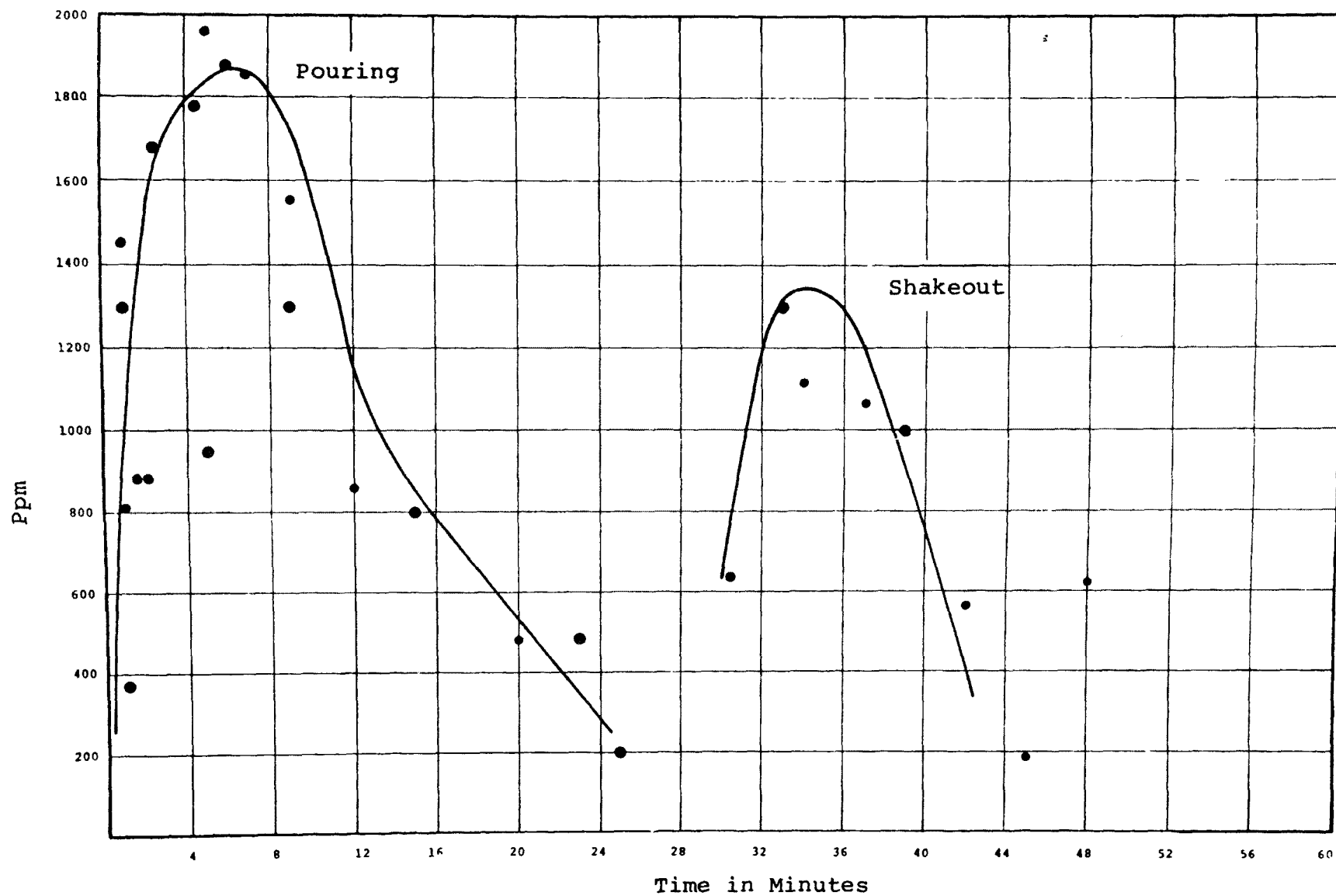


Figure 6. Variation of carbon monoxide concentration with time for green sand mold under the hood.

after pouring, and then began to gradually decrease. The casting was held in the mold for approximately 25 minutes and then was manually removed from the mold under the hooded sampling system. During the simulated shakeout operation, the CO content of the effluent increased as the mold was broken open and the hot sand exposed to the atmosphere. The CO concentration reached a level of about 1350 ppm during this operation and then began to decrease as the sand cooled.

Similar data is shown in Figure 7 for the concentrations of total hydrocarbons evolved after pouring and during shakeout of green sand molds. The hydrocarbon concentration increased after the mold was poured, to a value of about 1200 ppm at six minutes, and then began to decrease. The casting was broken out of the mold about 30 minutes after pouring and maximum hydrocarbon concentrations of about 1500 ppm were observed.

The average concentrations of the major gaseous constituents of the effluents from the green sand are shown in Table II. Two different castings were made: the 4-inch cube discussed above and a set of bars on an ingate that weighed about 15 lb. (with corresponding sand-to-metal ratio of 7:1). The values listed in Table II represent the average maximum concentrations observed during the pouring and breaking out of the castings. The size of the casting does not appear to be of major importance when the mold is well ventilated, and the values for the concentrations are not significantly different.

Hydrogen was not detected in these samples; it can be expected to burn in the mold. This burning, shown in Figure 5, may also account for some of the variations in the values obtained for carbon monoxide and carbon dioxide.

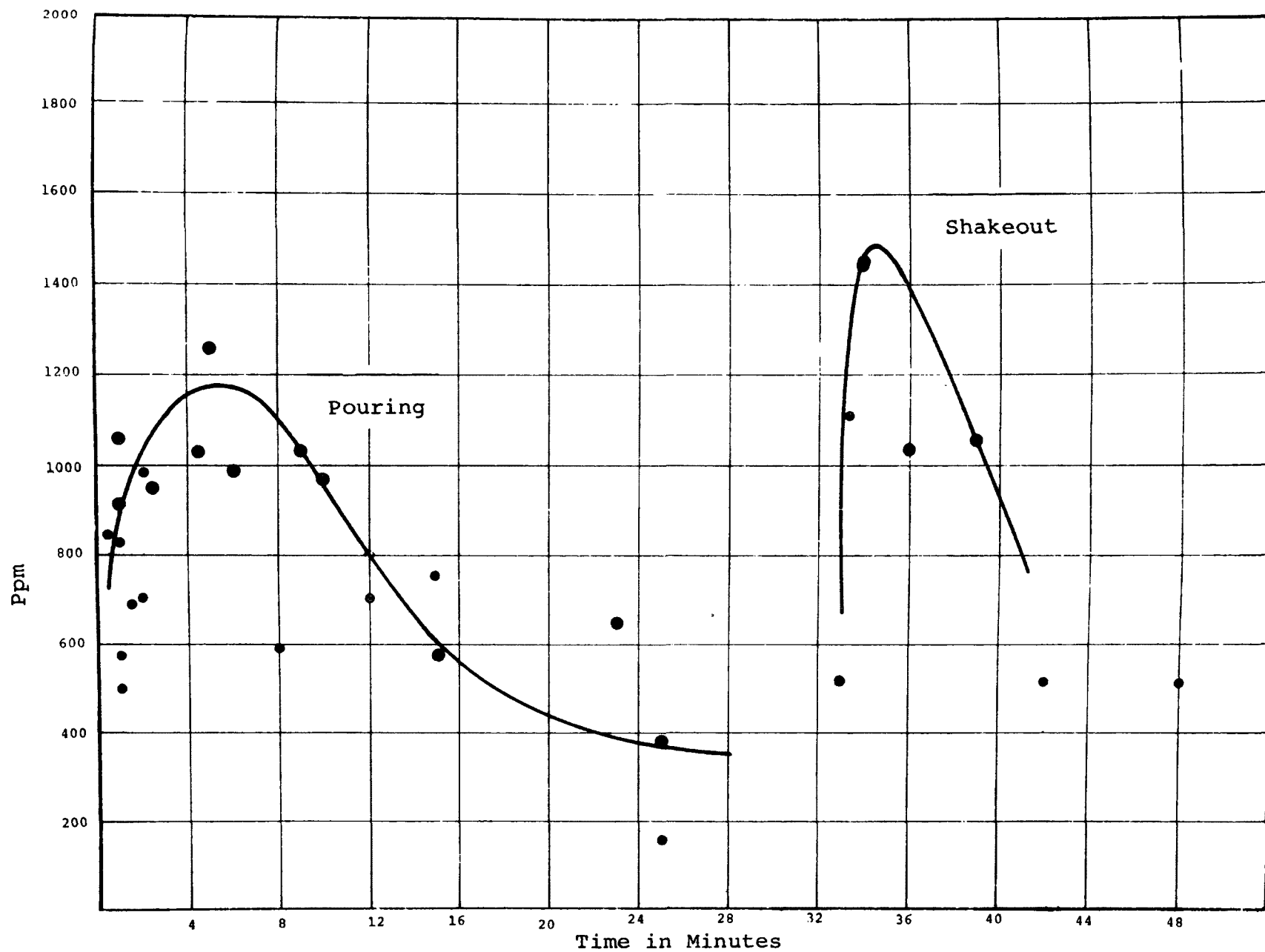


Figure 7. Variation of total hydrocarbons concentrated with time for green sand under mold.

Table II

Green Sand Emissions for Uncored Castings

Element		Bar Mold Sand:Metal Ratio = 7:1		Cube Mold Sand:Metal Ratio = 3:1	
		Pour	Shakeout	Pour	Shakeout
Carbon monoxide	(ppm)	1350	230	1510	650
Carbon dioxide	(ppm)	4920	2360	-	-
Total hydrocarbons	(ppm)	1780	640	1400	470
Methane	(ppm)	630	80	520	250
Cyanide	(ppm)	0.6	0.4	1.3	3.3
Ammonia	(ppm)	1.4	1.1	1.1	3.4

Particulate Emissions from Green Sand Molds

Considerable amounts of particulate material are evolved from the green sand molds after pouring, and during the breaking open of the molds to remove the castings. The particulate matter contains carbonaceous material from the burning organic material in the sand, silica fines and clay from the molding aggregate, other fines present in the mold, and metallic fumes.

The particle-size distributions of particulate matter collected in the cascade impactors during pouring and breakout of the green sand molds are presented in Table III. These values are for the 4" cube casting weighing about 30 pounds. During pouring, 95% of the particles evolved were less than 5 microns in diameter. During shakeout, 50% of the particles were less than 5 microns in diameter. Particles in this size range are considered more hazardous to human health than larger airborne particles because they penetrate into the lungs and are deposited there.

The dust loading of the effluent averaged 0.0625 grains/scf after pouring and during solidification, and 0.0968 grains/scf during the breakout of the 30 lb. casting. These calculations were based on the total weight of material collected during the sampling time of the impactors, and so represent average concentrations of dust in the air.

However, a time profile of dust concentration, shown in Figure 8, shows that loading peaks occur soon after pouring and again just after the breaking open of the mold to release the casting. This type of data was generated using an optical particle counter sampling over a ten-second interval and scanning several size ranges. The data shown in Figure 8 is for particles in the size range of 0.35 to 1.00 microns.

Table III

Particle Size Distributions of Green Sand
Emissions for 4" Cube Pattern

<u>Size (microns)</u>	<u>Pouring</u>		<u>Shakeout</u>	
	<u>Mass(grams)</u>	<u>% of Total</u>	<u>Mass(grams)</u>	<u>% of Total</u>
Less than 0.54	3.98	7.0	5.14	24.0
0.54 - 0.83	8.35	14.8	2.28	10.6
0.84 - 1.34	23.01	40.7	1.36	6.3
1.35 - 2.67	16.69	29.5	0.36	1.7
2.68 - 4.14	1.86	3.3	0.56	2.6
4.15 - 6.08	.97	1.7	0.24	1.1
6.09 - 8.95	.53	0.9	10.88	50.7
8.96 -14.36	.40	0.7	0.34	1.6
More than 14.36	.68	1.2	0.28	1.3

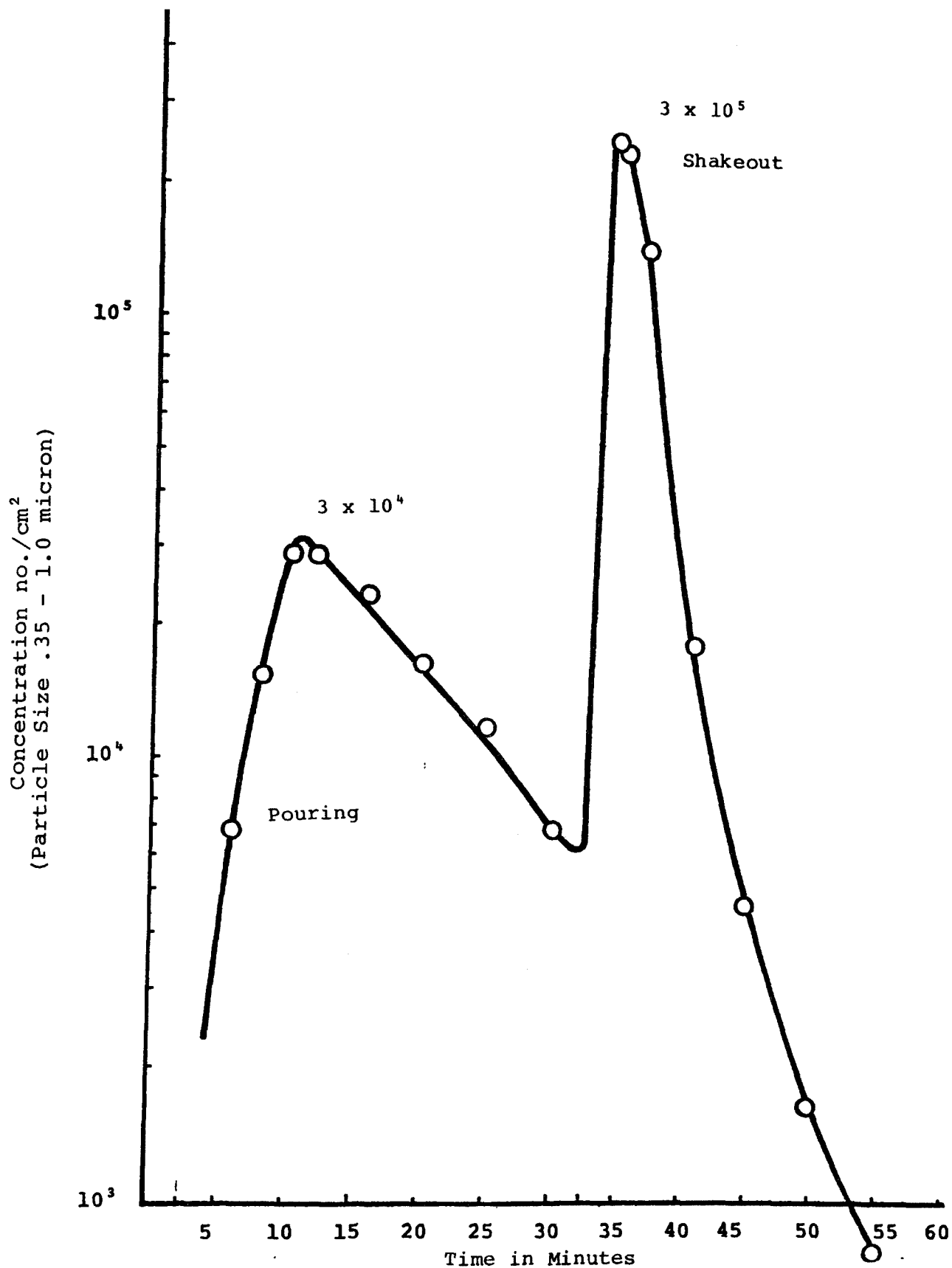


Figure 8. Time profile of dust concentration from cube casting in green sand.

A similar peaking is observed for larger particles, but the concentrations are much lower.

Particle counts increase very rapidly after pouring to a count of about 30,000 per cubic centimeter and then exponentially decrease with time. After allowing the casting to cool for 30 minutes in the mold, the mold was broken open in a simulated shakeout operation and the number of particles in the effluent increased dramatically to about 300,000 per cubic centimeter, an order of magnitude greater than the particulate emissions observed during pouring. This agrees with the impactor data that showed shakeout to have a higher dust loading than pouring.

The dust samples of particles less than 5 microns in diameter collected in the impactor were analyzed for free silica by the Talvite colorimetric method recommended by NIOSH.⁶ The free silica content of the dust ranged from 0.67% to 7.3%, with the mean value slightly over 1%. Other constituents of the dust were not identified.

Pilot Plant Studies

The sand casting laboratories were used for evaluating emissions from a small scale foundry operation. The molding sand used is high in natural clay content and is referred to as "Yellow Velvet." This sand has no seacoal in it, but 1% cereal binder is added. The water content is nominally 5%. Emissions were measured during mulling, at an automatic molding machine, and during pouring and shakeout.

In these trials, a plate casting with a sand-to-metal ratio of 4:1 and an open riser was cast. No mold ignition was observed, either during pouring or while on the runout line cooling, although steam was visibly given off. Table IV lists the results for the pilot plant

Table IV

Pilot Plant Emission Data

Muller:

Dust loading	.00396 gr/scf
CO	<50 ppm
CO ₂	850 ppm
Total Hydrocarbons	200 ppm
CH ₄	7 ppm

Hunter Molding Machine:

Dust loading	.00373 gr/scf
CO	<50 ppm
CO ₂	775 ppm
Total Hydrocarbons	180 ppm
CH ₄	2 ppm

Pouring:

Dust loading	.00291 gr/scf
CO	1500 ppm
CO ₂	2500 ppm
Total Hydrocarbons	250 ppm
CH ₄	15 ppm

Shakeout:

Dust loading	.01654 gr/scf
CO ₂	<50 ppm
CO	670 ppm
Total Hydrocarbons	215 ppm
CH ₄	5 ppm

study. The pouring emissions, collected under the hood, show a grain loading in the effluent of .00291 gr/scf. Carbon monoxide reached a maximum value of 1500 ppm, and the carbon dioxide level was a maximum of 2500 ppm. The total hydrocarbon content, calculated relative to methane, was 250 ppm.

The shakeout was accomplished by dumping the molds into a basket that moved up and down vigorously, so that the sand fell away from the castings and out the bottom. The atmosphere in this operation was extremely humid. The dust loading measured in the impactor was .01654 gr/scf. However, the gaseous effluent using this sand was much lower, with a carbon monoxide level less than 50 ppm, and carbon dioxide registering at only 670 ppm. Total hydrocarbons were 215 ppm.

Table IV also contains data on several other sand mold preparation operations found in the foundry. In mulling, batches of 150 lb. were prepared by adding the sand, then the clay, and finally the water, and mulling for three minutes. Total mulling cycle time was approximately six minutes, and sampling was continued through four complete cycles to insure that impactor catches were large enough for accurate weighing. Average sand temperature was 80°F.

The grain loading was observed to be .00396 gr/scf. The carbon monoxide level was below 50 ppm, and the carbon dioxide was near 850 ppm. Total hydrocarbons were present at 200 ppm. The operation in the pilot plant was considerably cleaner than with the green sand containing seacoal that was sampled in the laboratory muller. Table V lists the effluent analysis from the laboratory tests for the various sand preparation steps. The moisture content of the sand is the largest variable. A hot, dry sand as it is returned from the

Table V

Laboratory Sand Preparation Effluent

Muller

Dust loading (dry)	.192 gr/SCF
(wet)	.007 gr/SCF
Average CO	18 ppm
Average CO ₂	1530 ppm
Average Total Hydro-carbons	115 ppm

Jolt-squeeze molding

Average CO	<20 ppm
Average CO ₂	800 ppm
Average Total Hydro-carbons	<10 ppm

Shakeout

Dust loading	.149 gr/SCF
Average CO	20 ppm
Average CO ₂	510 ppm
Average Total Hydro-carbons	80 ppm

shakeout area produces a much higher grain loading than the wet ready-to-mold sand.

Discussion

This research has shown that a number of undesirable substances can be emitted into the foundry atmosphere from the green sand molding operation as it is commonly employed. Data from this research should be useful in developing suitable designs of ventilation systems, and it certainly shows the necessity of proper ventilation in the pouring area and in the run-out and shakeout areas of the foundry.

A typical foundry may pour over a hundred molds an hour. Each mold can produce an average of about 750 ppm of CO, or 150 ft.³, over a period of an hour, as estimated by integration of the area under the curve in Figure 6. The other gaseous constituents will be a fraction of this amount, but the combined amounts are large enough to be important.

In a foundry where the ventilation system is good, these species may well be diluted to the point where their discharge is harmless. The peak concentrations will tend to be levelled out as a new mold is poured every 30 to 45 seconds. In those foundries that have not considered the problem, the amount and nature of the environmental pollution may be significant.

Likewise the particulate matter introduced into the foundry atmosphere by the sand casting process is approximately 5.5 grams per mold for one of the castings selected in this investigation. The particle size distribution in Table III indicates that much of this material, especially that from the shakeout, will settle out inside the plant, or in the vicinity if it is entrained in the ventilation

system and exhausted from the plant. However, there are appreciable amounts of fine particles in these emissions that may present environmental problems. The low content of free silica in this material indicates that it may not present a serious health problem. These particles are small enough to remain airborne for several days and they are in the respirable size range. If some of the organic compounds emitted in the molding operation (such as the polynuclear aromatic hydrocarbons) are condensed on them, as is quite possible, they may present a hazard to human health. Much work needs to be done in the area of qualification and quantification of these compounds in the foundry emissions before the scope of the problem is properly understood.

There are several different ways to produce molds, and perhaps one of these will decrease the severity of the problem. There are sea-coal substitutes being marketed that, it is claimed, will decrease the amount of carbon monoxide emitted. However, these are polystyrenes and it is possible that some of their thermal decomposition products could be equally undesirable. Unfortunately, the reducing atmosphere at the mold-metal interface which is needed to make a good casting almost necessarily results in the production of noxious organic emissions.^{8,9}

In addition to the traditional clay bonded molding aggregates, there have been a number of chemical binders introduced in the last fifteen years. It is possible that these no-bake binders may improve the emissions. There are several of these binders that have little or no organic constituents, and perhaps one of these can be developed to where it can produce acceptable castings with relatively few harmful emissions.

Recent work with permanent mold casting processes for iron castings has shown that an increasing number of castings may be made by this process without loss of desired physical properties and with significantly less environmental contamination.^{2,10} The weight of particulate introduced into the atmosphere on a per casting basis was reduced from 5.5 grams to 0.15 grams for one particular configuration. Likewise, there were significant reductions in both peak and average gas concentrations. The average carbon monoxide concentration was less than 35 ppm. Unfortunately, this process is not suitable for all castings, but it could be used more extensively than at the present.

In conclusion, the foundry industry as a whole may have another problem in that their fugitive emissions appear to present an undesirable environmental contamination. Control will be difficult and expensive, due to the large amounts of air involved. The cost of control will have to be borne on top of the costs of control of stack emissions and requirements for meeting Department of Labor (OSHA) compliance. Many in the industry feel that they may not be able to afford another costly compliance program. There is evidence that a number of foundries have already been forced to close due to the cost of EPA and OSHA compliance requirements. In addition, the money spent by the foundries for compliance has been diverted from needed capital expenditures that would normally upgrade the technology in the foundry. A great deal of work needs to be done to identify and quantify the fugitive emissions from a foundry and to determine the importance of their contribution to the whole environment as weighed against

the importance of the industry to our society. If control should still be necessary, it then becomes paramount that the methods be fully developed before being implemented to insure the best capital utilization.

Acknowledgements

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Tuesday Afternoon, May 18, 1976

SESSION IV: CONTROL TECHNOLOGY

CONTROL OF FUGITIVE EMISSIONS IN PETROLEUM REFINING

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(Representing the American Petroleum Institute)

Mr. Weiland holds the title of Coordinator in the Environmental Protection Department of Texaco Inc. His primary responsibility is coordinating Texaco's activities in the areas of stationary source emissions and environmental noise. Mr. Weiland has been actively involved in the American Petroleum Institute's environmental affairs activities for the past 10 years, having served as Chairman and member of a number of task forces investigating various industry problems associated with stationary source emissions. He is currently Chairman of the Stationary Source Emissions Committee of the API Division of Environmental Affairs.

* * * * *

For the purpose of this discussion fugitive emissions are defined as any emissions which are not released through a stack or duct. In petroleum refining, under this definition, volatile hydrocarbons are the primary fugitive emissions of concern. It is proposed to review some of the emission sources, discuss briefly the emission factors that are commonly used to attempt to get some fix on these emissions, and then discuss some of the control methods that may be used.

There are a great many possible sources of fugitive hydrocarbon emissions in a refinery. A modern refinery is a very complex installation consisting of numerous different processes, all with associated pumps, compressors, pipe line flanges, valves, including relief valves, tanks, and so on. There may be thousands of flanges and valves in a large complex refinery. Pumps and compressors can number in the hundreds.

Other possible sources of fugitive emissions, much fewer in number, include vacuum jets, cooling towers, blowdown systems, hydrocarbon loading operations, sampling of the hydrocarbon streams, process drains, and oil-water separators.

The emission factors in EPA publication AP-42* basically go back to studies made in the Los Angeles area in the last of the fifties. These studies were conducted by the U.S. Public Health Service in cooperation with the Western Oil & Gas Association and the Los Angeles Air Pollution Control District. This was a very excellent study in that an effort was made to thoroughly inventory the equipment and facilities which actually existed in these plants and the services in which the equipment was operating. Then "representative" individual items were selected for emission testing. Various procedures were used, including enclosing the equipment in plastic "tents" where feasible to capture and measure the emissions.

There have been a number of comments made during the course of this conference which have emphasized that an emission factor for a given piece of equipment or process is not necessarily applicable across the board to all superficially similar equipment in a whole industry. Thus, it is important to keep in mind that, in an absolute sense, the factors that were derived for petroleum refineries were specific to the

*"Compilation of Air Pollutant Emission Factors," U.S. Environmental Protection Agency, March, 1975.

pieces of equipment which were measured and the extrapolation of these factors to any refinery, in the gross sense, or to any other petroleum operation is not technically completely accurate. The results, however, have been useful to give a general idea of where major problem areas may be and to help evaluate the possible order of magnitude of refinery emissions.

At the present time the emission factors developed in the late 1950's are no longer believed generally applicable in many cases. The technology's improved a very great deal; the operating and maintenance procedures have improved over the last 15 years and some of these factors are undoubtedly high by an order of magnitude or more when applied to modern technology and operations. Consideration has been and is being given by industry and various regulatory agencies to the updating of these emission factors. This may indeed become necessary in order to obtain a current assessment of the present order of magnitude of fugitive hydrocarbon emissions from modern refineries. However, it is not an easy or inexpensive thing to do.

Now, let's consider possible control methods. Good housekeeping and good maintenance is the key to eliminating or minimizing hydrocarbon losses from many of the sources mentioned. It is necessary that operating personnel realize that hydrocarbons must be contained within the appropriate lines and vessels and that any leaks or malfunctions should be corrected promptly. Specific comments on the various sources follow:

- Flanges and valves - Leaks can be eliminated or held to an absolute minimum with adequate inspection procedures and prompt maintenance to correct any leaks noted.

- Relief valves - Venting of relief valves to the refinery flare system to the maximum extent feasible will reduce hydrocarbon emissions from this source to a minimum.

- Pumps and compressors - There has been a great deal of both technical and housekeeping improvement in this area. The industry is going more and more to the use of mechanical seals on new equipment as opposed to packing glands. Properly designed and maintained mechanical seals reduce emissions to insignificance in pumps and compressors. In cases where there are older pumps with packing glands or where packing glands are needed because of the type of service a technique that can be used is to collect any drips and route them into an underground slop oil tank for later pumpout to a slop oil system.

- Tankage - The control of volatile hydrocarbons emissions from tankage is accomplished either by the use of floating roofs or vapor recovery systems. Both of these are very effective in controlling these emissions, and over the years the industry has increasingly utilized such systems.

- Vacuum jets - In the past many vacuum towers on crude units used a barometric condenser system to condense the steam from the vacuum jets. This requires the injection of large volumes of water into the steam jet. The water and condensed steam was discharged into a sump at the base of the jets. There is some carryover of hydrocarbon

fractions into the barometric sump resulting in some hydrocarbon evaporation into the atmosphere. This can be corrected by replacing barometric condensers using direct water injection with surface condensers.

- Cooling Towers - The control of cooling towers is obviously a question of maintenance. There will be no hydrocarbons in cooling towers unless there is a leak in condensers or coolers somewhere in the system and unless the pressure differential is such that the hydrocarbon pressure in the equipment is in excess of the water pressure.

- Blowdown systems - In preparing a unit for test and inspection, any vessels, exchangers, lines, etc. that are going to be opened must, as a matter of safety, be thoroughly purged of hydrocarbons. To minimize hydrocarbon emissions the unit can be depressured to a flare and the liquid hydrocarbons that remain pumped to a slop tank. The remaining hydrocarbons -- and there'll still be little residues here and there -- can be drained to an underground slop tank.

- Process drains, oil/water sewers, and oil/water separators - The best solution to these, of course, is to keep the amount of hydrocarbons entering them to a minimum. Over and beyond that, use of covered-drain systems and covered oil/water separators will prevent or minimize hydrocarbon emissions to the atmosphere. In addition, segregation of steam condensate and any other high temperature water to prevent its discharge into oil/water sewers or process drains will reduce volatilization of any hydrocarbons in the system.

- Hydrocarbon loading operations - Hydrocarbon emissions during loading of tank cars and tank trucks can be controlled by use of a vapor-recovery system which will recover the hydrocarbons during the loading operation.

- Fugitive dust emissions - The major possible source of such emissions in a refinery is from coke piles associated with coking units. There are at least three ways of handling any problem of dust emissions from a coke pile. One involves the use of a large building in which to store the coke under cover. If outside storage is used, the pile can be wet down with water. This can create a problem in that any water runoff must be settled to remove coke dust. Another way that has been used where the pile is going to be inactive for a time is to spray the pile with some type of a polymer solution which will seal the surface.

In summary, a well controlled and maintained refinery should have minimal problems with the types of emissions discussed. One of the ways to measure the progress that a given refinery has made over the years is to compare the odor emanating from a well-controlled refinery today with how the refinery smelled say 35 years ago. Part of the improvement, of course, is associated with control of stack emissions. But a great deal of it is due to the better housekeeping and to the use of improved equipment and procedures. This improvement is certainly indicative that the refinery is indeed doing a good job of controlling fugitive emissions.

THE COST EFFECTIVENESS OF
COKE OVEN CONTROL TECHNOLOGY

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THE COST EFFECTIVENESS OF COKE OVEN CONTROL TECHNOLOGY -
STUDY OF AMBIENT AIR IMPACT

1.0 Introduction

The state-of-the art of coke oven emissions has progressed dramatically in the last decade. Substantial reductions in charging, pushing, and quenching emissions are possible; however, there are large costs associated with this new control technology. Four questions need answering:

1. What effect do controls have on ambient air quality?
(Primary standards and/or allowed significant deterioration increment)
2. What is feasible for retrofit on present coke ovens?
3. What is feasible for installation with new coke ovens?
4. What controls are most cost effective?

TRC air quality studies for industrial clients have shown that low level or fugitive emissions, such as those usually found in coke oven operations, are often the critical factor in determining local ambient air quality rather than the stack emissions.

The steel industry is faced with the following dilemma:

- o Ambient air quality standards are not being met in many urban areas where integrated steel mills are located.
- o Substantial sums of money have already been expended by steel companies to control stack emissions.
- o The highly visible fugitive emissions from operations such as coke oven batteries have been identified as the next targets for control equipment installation.
- o The cost of fugitive emission controls is high and not as well proven as those for stack emissions.
- o If, after installation of these controls, ambient air quality does not substantially improve, more pressure will be on for tighter controls on all steel mill air emission sources.

It is the purpose of this paper to show how to assess the cost effectiveness of coke oven emission controls. The approach uses measured coke oven emission rates or best estimates of them as inputs to a computer model which predicts their impact on ambient air quality. Knowledge of the costs of each control concept and the degree of control of coke oven emissions for each concept permits the determination of the incremental cost of incremental improvements in ambient air quality. When these are plotted, cost effectiveness curves are developed which answer two serious questions. These questions are:

- o Will coke oven emission controls solve the ambient air quality problem?

- o If so, what is the minimum cost solution to the problem?

If emission controls are installed, they must solve the right problems.

2.0 Coke Emission Control Technology

Although the purpose of this paper was not to establish the state-of-the-art in coke oven control technology, an evaluation of the feasibility (commercial and technical) of various proposed control methods was required. The most proven technologies were identified for control of:

- o Coke oven charging emissions
- o Coke pushing emissions
- o Coke quenching emissions

OTHER SOURCES (door leaks, underfiring, coal and coke handling) were not evaluated here.

2.1 Coke Oven Charging

The charging of coal into coke ovens results in a fugitive emission release consisting of coal dust, tars and gases from the charging hole. Control technologies considered commercially feasible for prevention of substantial charging emissions include:

- o Staged charging with oven evacuation
- o Larry cars equipped with gas collecting systems and wet scrubbers
- o Pipeline charging

The former two have been considered as retrofits for present coke ovens as well as feasible for new coke oven battery construction. In some cases, pipeline charging, which is a technology considered suitable for new construction, has been installed in rebuilt batteries to meet the need for strict control of particulate emissions.

2.2 Coking

Leakage of emissions (gases, fumes) from the coke oven doors and other openings in the ovens are minor but hard to control sources of emissions. Although improved door sealing is a potential control, it is hard to estimate the degree of control achieved by this technique. The only controls considered here were sheds with scrubbers whose primary purpose was to control pushing emissions.

2.3 Coke Pushing

The pushing of the incandescent coke from the oven into the quench

car results in emission of hot coke particles and tars as well as gases from the coke as it leaves the oven and dumps into the quench car. Although there have been commercial control equipment installations, the technology is undergoing change and new concepts are now in design stage. Commercially feasible controls include:

- o Coke side sheds ducted to wet scrubbers or electrostatic precipitators
- o Coke guide and hooded quench car

Both have been considered for retrofit to present ovens and for new construction. In new construction, the hooded quench car can contain a mobile quench station which eliminates quench towers.

2.4 Coke Quenching

Although changes in this technology may be more related to water reuse and pollution, they do result in lower air pollution emissions. Commercially feasible control technologies include:

- o Dry quenching
- o Coke guide and hooded quench car (with mobile quench station)

Both technologies have been primarily considered for new installations rather than retrofit.

2.5 Effectiveness of Controls

Because of the problems associated with fugitive emission measurement and the small number of present control equipment installations, the effective percent emission control of many of the technologies described can only be estimated. Conservative estimates were used for calculations in this paper, and deviations from these in actual practice can be shown not to be critical for analysis of the data. Comprehensive and accurate measurement programs will be required to give us better emission factors for coke oven operations. Table II-I sums up the emission estimates used in the prediction of the effect of coke oven emissions on ambient air quality.

3.0 Air Quality Prediction

3.1 Methodology for Prediction of Impact

The basic procedures for the accurate prediction of the impact on ambient air of any emissions, whether fugitive or stack, include:

- o Identification of all significant emission points
- o Estimation of emission release heights, temperatures and exit velocities
- o Estimation of emission rates and particle size/density for particulates (latter for deposition models)

- o Evaluation of availability and validity of local ambient air quality and meteorology data
- o Application of a reliable, tested short term (1 - 24 hour) or long term (annual average) diffusion model to worst case air quality time period or periods
- o Comparison of measured and predicted air quality to establish the accuracy of the calculations
- o Use of auxiliary techniques (tracer studies, morphological/chemical analysis of particulates, etc.) to enhance accuracy of predictions if required.

TRC has applied these techniques to the study for the steel industry of coke emission impacts for several projected and existing facilities. The predicted and observed air quality data (in the case of existing facilities) has shown the validity and the basic accuracy of the methodology mentioned above.

3.2 Case Studies

As a case study, TRC chose a hypothetical coke oven facility which could represent a proposed or present facility. The facility specifications are given in Table III-1. This represents a medium size "grassroots" mill or an expansion of a present integrated steel mill.

The emission factors of Table II-1 were used to represent those of the actual facilities with and without various add-on controls. A TRC developed short-term air quality diffusion model was used in conjunction with "worst case" 24-hour meteorology from typical steel mill locations in East Coast and Pittsburgh areas to show the impact of coke oven emissions on ambient air quality at two different site types (coastal and river valley). Particle size was assumed to be 1.0μ and particle density was assumed to be 1.0 gm/cc . Emissions from coal handling or oven underfiring were not modeled.

3.3 Results of Modelling

Figure 3-1 is an isopleth map of the "worst case" 24-hour particulate concentrations for the East Coast location with no coke oven emission controls. As a comparison, Table III-2 shows the "fenceline" air quality results and results at $1/4$, $1/2$ and 1 mile from the "fenceline" for both controlled and uncontrolled coke oven emissions.

The results show that uncontrolled coke oven emissions have a significant impact on local ambient air quality. In order to ensure that off-plant ambient air samples met the 24-hour primary particulate standards (considering here also the other potential fugitive sources in-plant and background particulate levels), an uncontrolled coke oven would have to be located about $1 \frac{1}{2}$ miles from any steel plant boundary. In fact, it would be imperative that significant modelling predictions be run before the location for such a facility is chosen. The impact decreases drastically with distance from the plant "fenceline" and is also directionally dependent.

A similar analysis can be given to the predicted particulate concentrations for the Pittsburgh area location. Figure 3-2 is an isopleth map for particulate concentrations for this case, and Table II-3 summarizes particulate concentrations as a function of distance from plant "fence-line". The comparison of controlled vs uncontrolled coke oven emissions is similar to that for the East Coast location.

One point worth noting is the impact of quench tower emissions on ambient air quality. Because they act as a high velocity elevated plume from a stack (except for "rain out" close to the tower) rather than as a fugitive emission, their impact would be at a greater distance from the plant than low level fugitive emission from changing or pushing. In fact, their local impact might be increased by their control by a mobil quench facility whose off-gasses are wet scrubbed even though overall impact is decreased off plant property. The low buoyancy, low velocity plume if released from a stub stack will be transported in a similar manner to the true fugitive emissions.

The two case studies shown are just examples of potential impact of coke oven emissions on ambient air quality. Each specific site may lead to quantitatively different results. However, the two cases cited show, for locations of many present and proposed steel mill facilities, the environmental problems of coke oven site selection.

4.0 Cost Effectiveness of Controls

In order to determine the cost effectiveness of the potential coke oven emission control technologies, it was necessary to compare the costs of each of these. Table IV-1 shows updated cost data developed from current literature sources. It shows that there are a wide range of costs associated with the emission controls considered for present and proposed coke ovens. Their cost effectiveness is determined by plotting the predicted ambient air quality at a specific location against the cost of the control technology required to obtain that air quality.

Figures 4-1 and 4-2 show such plots for two ambient locations beyond plant "fenceline" (East Coast and Pittsburgh locations are plotted on the same curves). As better and better air quality is required, the costs rise significantly and not in any direct proportion to the air quality improvement. These cases have in addition neglected the fugitive emissions from such integrated steel mill sources as blast furnaces, BOF shops, electric furnace shops, sinter plants and ore/limestone/coal/coke storage. The cost curves are similar for control of these. It is therefore important to ask the question: Is there a significant overall benefit to the population in reducing coke oven (and other fugitive source) emissions below a certain level where controls are no longer cost-effective in order to meet air quality standards? An additional question is: Is there another control strategy which would achieve the same result?

5.0 Conclusions

The results of this case study have the following implications for present coke oven operations:

- o Stringent controls may be required on coke ovens to help

the attainment of ambient air quality standards in the local area.

- o It is important to identify the sources which significantly impact the ambient air so that the controls can be applied to the proper sources.
- o The controls may be costly and require rebuilding of the ovens.
- o It is possible, even after the implementation of controls, especially where property lines are close to the coke ovens, to have ambient air quality standard violations partially caused by coke oven emissions.
- o It is important for steel mill management to examine the cost-effectiveness (air quality benefit vs incremental control costs) of several control system alternatives for coke oven emissions before implementing any of them.
- o Attention should also be paid to control of other in-plant and non-plant fugitive sources.

For proposed coke oven operations, the aforementioned hold and, as well, there are the following implications:

- o The environmental as well as construction/process engineering aspects of coke oven battery placement must be taken into account in design and placement of a new facility.
- o Even with stringent coke oven controls, problems may occur in meeting the air quality deterioration criteria for new facilities even in areas where such industrial expansion would be allowed.

The technology of coke oven emission control is advancing, however it is important to assess the impact on ambient air quality of the expenditure of such large sums of capital. TRC has identified, and is now pursuing, the following areas of future needs to improve our ability to predict the impact of fugitive emissions (such as coke ovens) on ambient air quality:

- o Methods for measurement of mass rate, particle size/density of fugitive emissions.
- o Methods for the measurement of time dependent and ill-defined fugitive emission sources.
- o Methods for the calibration of models using tracers to develop site-specific detailed models for industrial sites.
- o Methods for better defining non-industrial emission

sources or ill-defined fugitive sources to allow better predictions of ambient air quality.

These areas of development will allow us to give a more thorough analysis of what affects local ambient air quality and how to improve it in the most cost-effective manner.

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2. P. W. Kalika, P. T. Bartlett, R. E. Kenson and J. E. Yocom, "Measurement of Fugitive Emissions", 68th Annual APCA Meeting, Boston, Mass., June 1975.
3. L. F. Kutcher and B. Linsky, "Economics of Coke Oven Charging Controls", Air Pollution Control Association, 24:765 (1974).
4. Air Pollution Emission Factors, EPA Publication AP-42, April 1973.

Table II - 1

**Emission Factors* Used for the Prediction of the
Impact of Coke Oven Emissions on Ambient Air Quality**

Operation	Controls technology	Estimated per cent control	Particulate emissions (lbs./ton coal)
Charging	None	0	1.5
Charging	Staged charging	90	0.15
Charging	Scrubber on Larry car	90	0.15
Charging	Pipeline charging	98	0.03
Coking	None	0	0.1
Coking	Coke side shed/scrubber	42	0.06
Coking	Push/coke side shed/scrubber	83	0.02
Pushing	None	0	0.6
Pushing	Coke side shed/scrubber	83	0.1
Pushing	Coke guide/hood	95	0.03
Quench	None	0	0.9
Quench	Coke guide/hood	98	0.02
Quench	Dry quench	98	0.02

*Uncontrolled taken from
EPA Publication AP-42

Table III - 1
Hypothetical Coke Oven Facility For Case Study

Steel Production	2.5 mm tons/year (6850 tons/day)
Coke Production	4100 tons/day
Coke Batteries	2
Ovens/Battery	100
Oven Capacity	25 tons
Coking Cycle	16 hours
Oven Reliability	90%

Oven Site Size: 1000' x 3000' (including coal storage)
Steel Plant Size: 6000' x 8000' (centered on coke ovens)

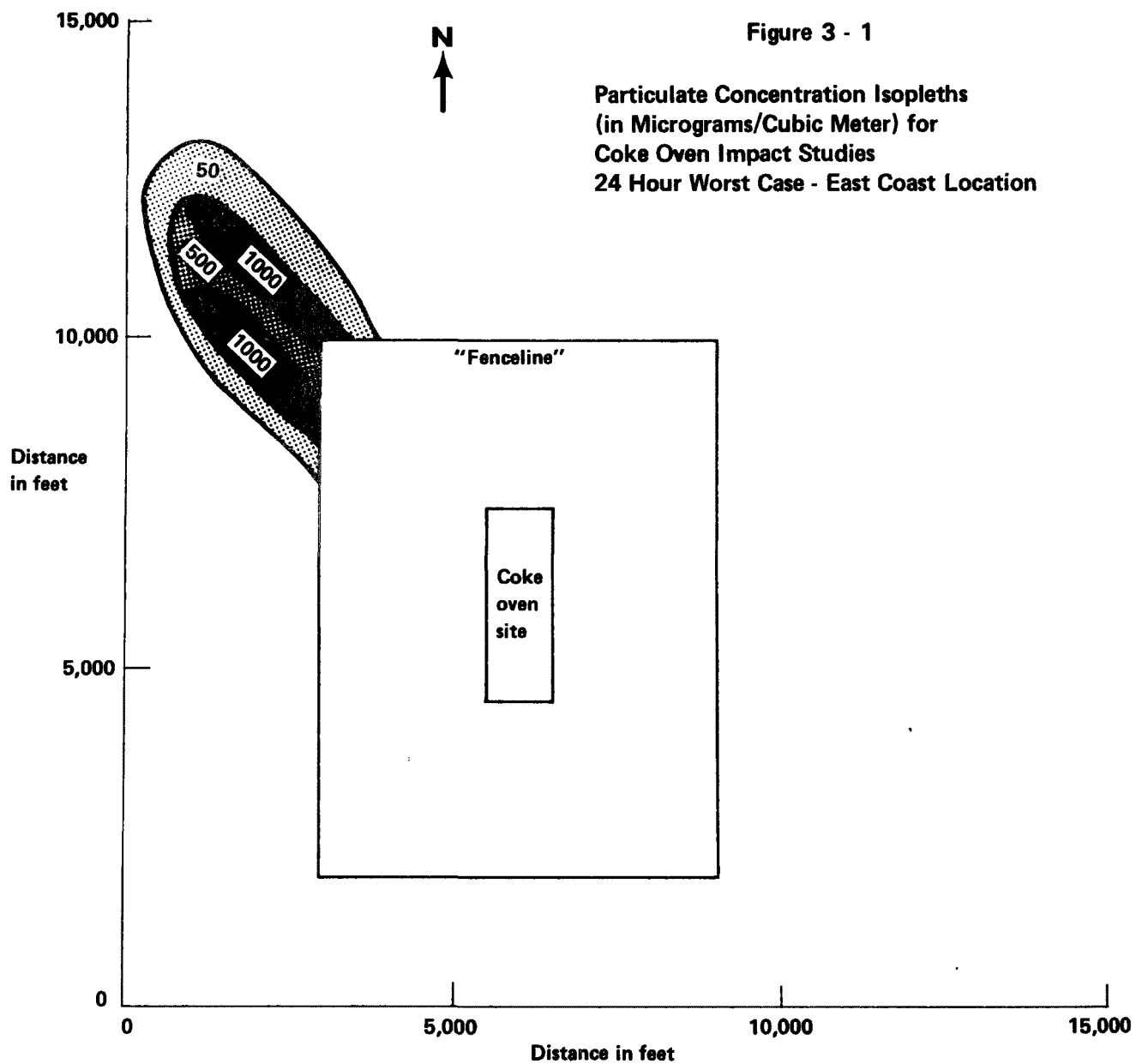


Table III - 2

**Predicted Ambient Air Quality (Micrograms/Cubic Meter)
for Particulates - 24 Hour Worst Case for East Coast Coke Oven Location**

	Fenceline sampler	1/4 mile from Fenceline	1/2 mile from Fenceline	1 mile from Fenceline
Uncontrolled	1290	1275	295	30
Staged charging	500	490	115	12
Staged charging plus coke side shed with scrubber	182	180	40	4
Staged charging plus coke guide/hood	165	160	37	4
Pipeline charging plus coke side shed with scrubber	110	109	25	3
Pipeline charging plus coke guide/hood	94	93	22	2
Pipeline charging plus push/coke side sheds with scrubber plus coke guide/hood plus dry quench	53	52	13	1

Table III - 3

**Predicted Ambient Air Quality (Micrograms/Cubic Meter)
for Particulates - 24 Hour Worst Case for Pittsburgh Coke Oven Location**

	Fenceline sampler	1/4 mile from Fenceline	1/2 mile from Fenceline	1 mile from Fenceline
Uncontrolled	750	480	315	67
Staged charging	290	195	125	28
Staged charging plus shed with scrubber/precipitator	95	60	40	9
Pipeline charging plus coke side shed with scrubber	65	40	27	6
Staged charging plus coke guide/hood	55	35	23	5
Pipeline charging plus push/coke side sheds with scrubber plus coke guide hood plus dry quench	30	20	13	3

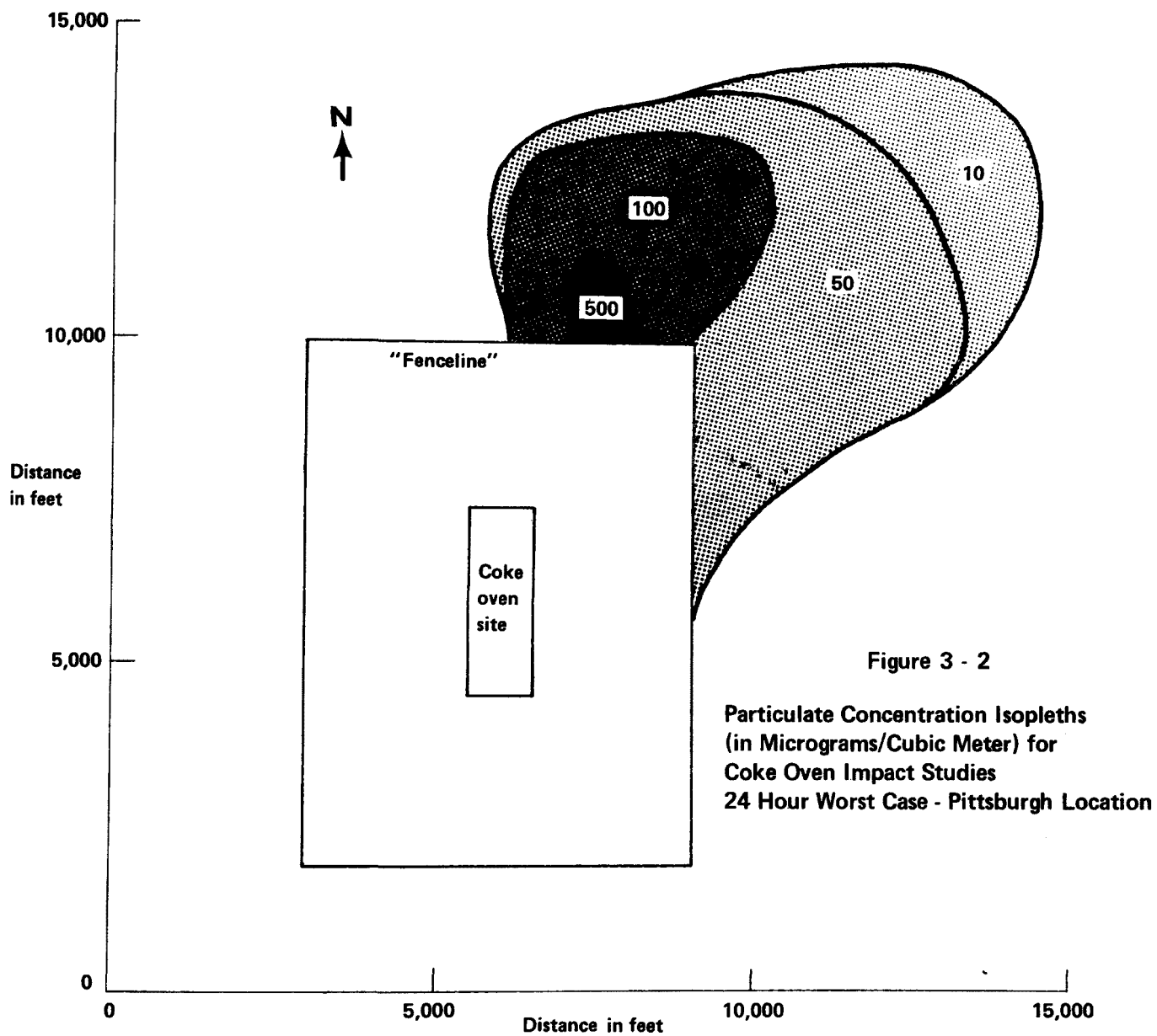


Figure 3 - 2

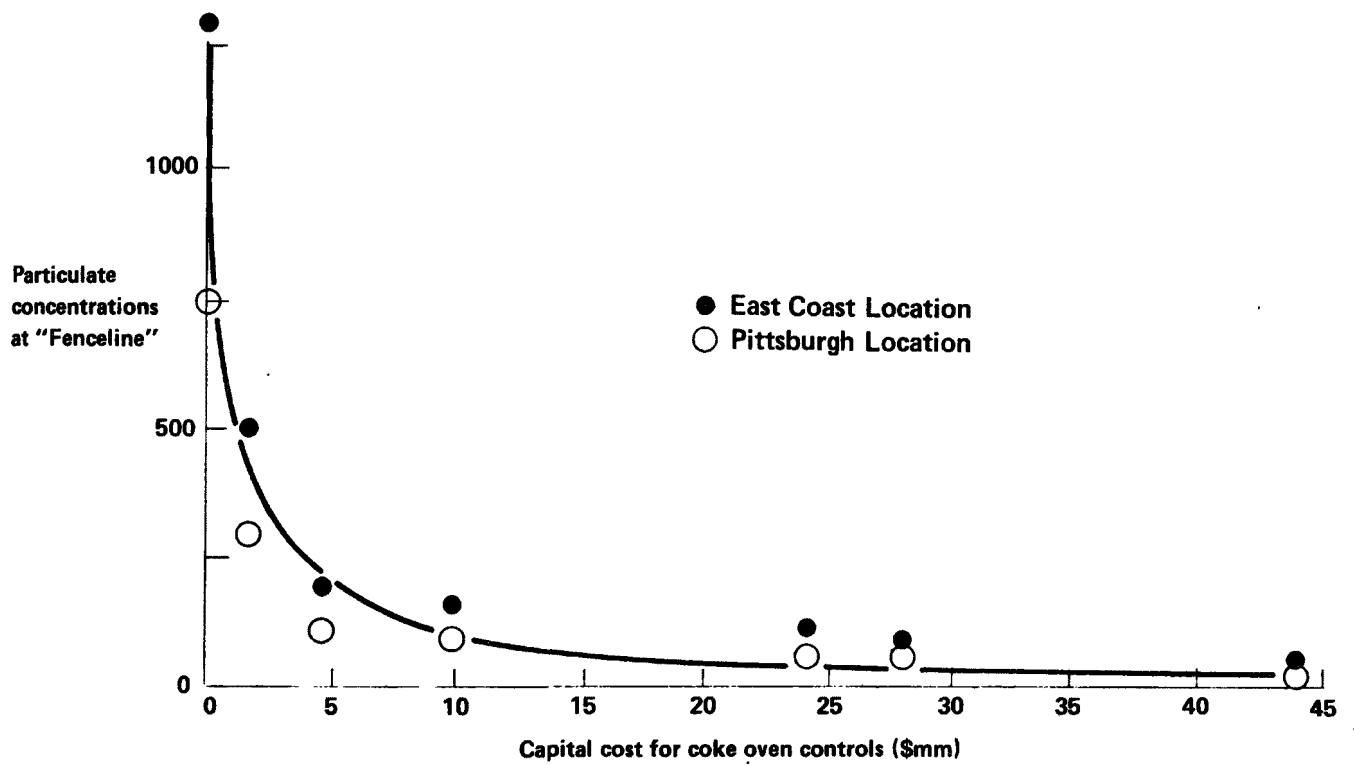
Particulate Concentration Isopleths
(in Micrograms/Cubic Meter) for
Coke Oven Impact Studies
24 Hour Worst Case - Pittsburgh Location

Table IV - 1

**Estimated Costs of Emission Control Systems for
Coke Oven Cases Studied***

Controls installed	Capital costs \$		Operating, maintenance and repair costs \$/year	
	2 batteries	4 batteries	2 batteries	4 batteries
1. AISI staged charging	1,500,000	3,000,000	200,000	400,000
2. Coke side shed				
a) with scrubber	3,000,000	6,000,000	450,000	900,000
b) with precipitator	6,000,000	12,000,000	200,000	400,000
3. Charging air with scrubber	1,500,000	3,000,000	225,000	450,000
4. Enclosed coke pushing and quench car with scrubber	8,000,000	16,000,000	800,000	1,600,000
5. Pipeline charging (Retrofit)	20,000,000	40,000,000	2,000,000	4,000,000
6. Dry quenching	10,000,000	20,000,000	650,000	1,300,000

*Using updated literature data and equipment vendor quotations



Cost Effectiveness of Coke Oven
Emission Controls

Figure 4 - 1

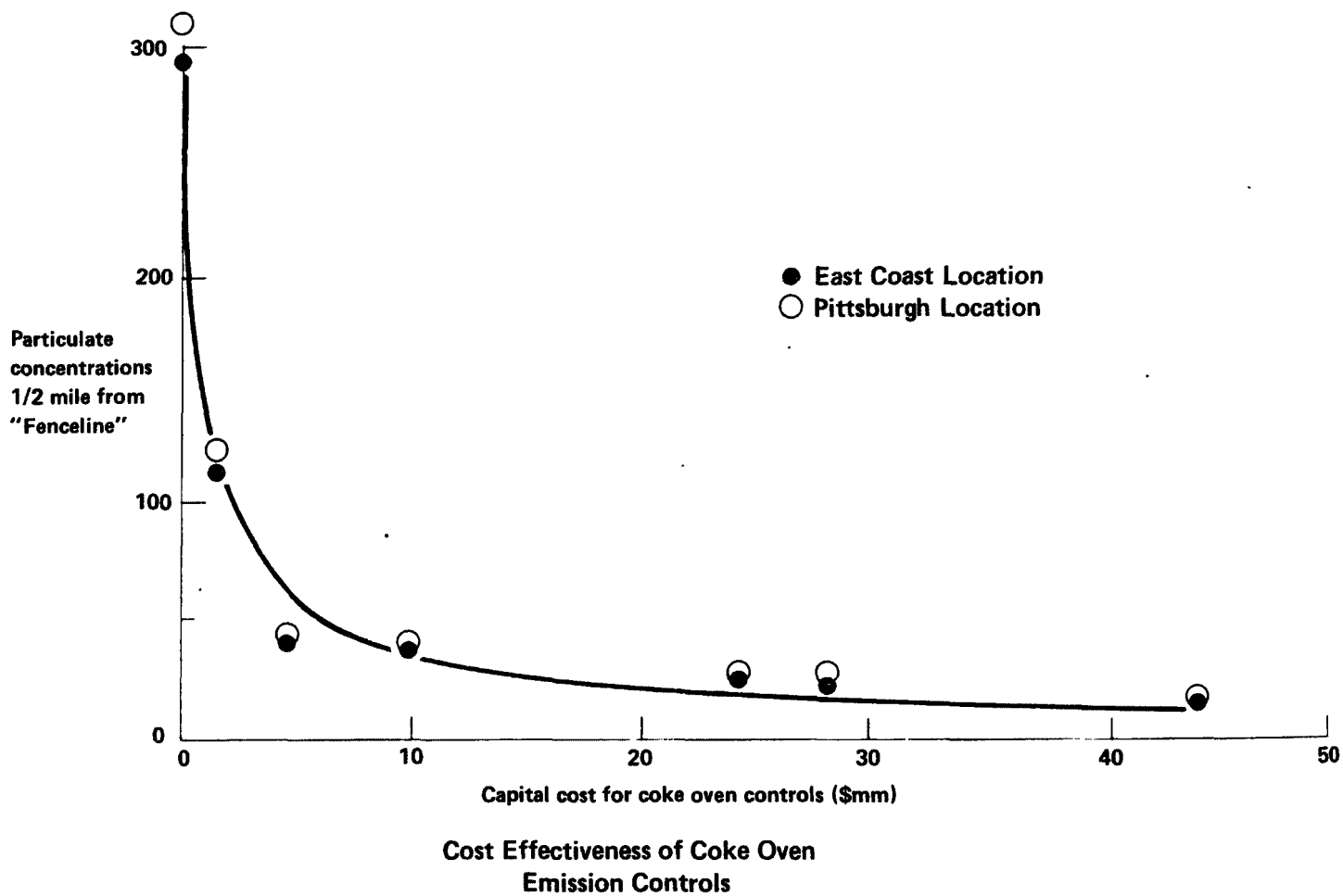


Figure 4 - 2

Session V:

FUTURE NEEDS FOR MEASUREMENTS AND CONTROL TECHNOLOGY

Robert M. Statnick, Ph.D.
Session Chairman

FUTURE NEEDS FOR MEASUREMENT AND CONTROL OF FUGITIVE DUST

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* * *

This is not a technical paper in any sense of the word. It is a review of how the crushed stone industry looks at a problem, which is basically a dust problem...fugitive dust problems. Dr. Thomas Blackwood has laid some very excellent groundwork for this review with his studies of the impact of open sources on over-all air quality.

To begin this presentation, here is a bit of background on the crushed stone industry. The crushed stone industry is the largest non-fuel mining industry in the country. Approximately one billion tons of rock are processed per year. These billion tons are produced from approximately 4500 to 4700 individual quarry sites around the country. These quarries vary from extremely small, independently-owned operations to extremely large, corporate-held locations. For instance, the 1900 smallest quarries account for less than 2 percent of the total production; the 180 largest quarries account for over 50 percent of the production. There is a tremendous size variation and there is also a tremendous difference in the ability of individual companies to deal with the many technical problems facing the industry today.

BACKGROUND

Concern with the ambient dust situation in quarries started in the early 1960's. In 1963 it was decided that the time had come to try to determine what was meant by the numbers everybody was talking about with respect to a rock quarry.

Considerable time was spent in the field and some very inter-

esting things were discovered. The only types of applicable measuring procedures available at that time were, of course, the ambient air measuring techniques. But, it was found that there is a very definite area of influence around a rock quarry. It doesn't really make much difference whether one is upwind or downwind of the site. The fact that there is a big hole in the ground tends to set up a micrometeorology in the area and an upwind/downwind relationship does not truly develop. A scattering phenomenon in all directions is more common. If one were to take a compass and draw a circle approximately 3500 feet in radius from some central point in the operation, that circle would encompass, for all practical purposes, the impact area of that rock quarry. In other words, at about this distance both the suspended air levels and the settled dust levels essentially return to the normal background in the area. High volume samples within this area literally have dust particles falling off the filter paper. But, as Dr. Blackwood previously stated, one is not really measuring suspended dust, but rather both suspended dust and a substantial amount of dust that is on its way out of the atmosphere...very heavy, large pieces of rock.

Since the above original work, this same pattern has developed at a number of locations. There seems to be almost a halving effect for every thousand feet one moves away from some central point within the operation. Within this area, however, there is without question, some impact---not a toxic situation, but rather a nuisance situation.

For years our industry has talked about the captive or capturable dust, which is generated as a result of the processing

operation...crushing, screening and conveying. This is opposed to the fugitive dust, which is generated from things like the unpaved roads, the open quarry floor, the area that is stripped bare of vegetation, etc. The plant-generated dust is fairly easily controllable and is not truly a problem. Collecting and suppression equipment can be installed. However, there is some question as to whether or not this truly solves the problem. There was a study conducted a number of years ago by the Manatee County Health Department in the State of Florida around a dolomitic lime plant -- quarry and plant operation. The significant finding of that study was that regardless of whether the control equipment was operating or not, the impact on the downwind ambient air concentrations was negligible. In other words, no real difference could be detected between the ambient levels in that plant area when that plant's control equipment was operating.

An assumption is that 75 to 80% of the crushed stone industry's problem is probably due to what was defined above as fugitive dusts. This has never truly been documented because normal test methods have not permitted isolation of the various fugitive sources. It has been inferred from looking at an operation that open road areas and the open quarry floor were probably the primary problems.

Some of the data that was presented at this conference by Dr. Blackwood tends to indicate that this assumption is correct. As an example, he demonstrated that for a typical limestone quarrying operation, there was a total emission in the neighborhood of .007 pounds per ton, and that roughly two-thirds of that was attributable to the fugitive sources, primarily the haul roads and quarry floor areas.

CURRENT STATUS

The crushed stone industry has two control options. The first option is the dry collection, the second is wet suppression. Each option has certain inherent problems.

The major problem with the dry collection option, of course, is the creation of a substantial solid waste disposal problem. There are quarries that have collection equipment in operation and are collecting from 50 or more tons of dry dust a day. They have a gigantic solid-waste disposal problem. Much of this dry dust is not saleable; therefore, it must be disposed of, dumped, or otherwise handled. Another problems created by the installation of dry collection equipment is the creation of a stack or stacks which in turn subject the operation to stack emission codes. Since there is evidence that there is very little measurable effect on ambient air levels whether the control equipment is operating or not, one must ask whether a problem has really been solved. Just recently the stone industry has been involved with the Environmental Protection Agency as they have attempted to develop new source performance standards. Two plants that were on the drawing board were studied. They were roughly 300-350 ton-an-hour operations. Note what would happen, not in terms of cost, but rather in terms of power consumption, if adequate dry collection equipment to meet a 0.02 to 0.03 grain loading standard were installed. On these two plants, the power required to operate the control equipment approached 20 to 26% of the power to operate the plant. That is a substantial amount of additional power. Forget about the cost of the control equipment; forget about the cost of that power. Just consider the

total number of kilowatt hours in that 20 to 26% excess power required to operate this collection equipment. Under those circumstances, has the problem been solved or has a new problem been created? In other words, somewhere, someplace, somebody has to burn some coal or other fuel to generate that power being used to operate the control equipment to collect the dust which doesn't have an impact beyond a half mile. This is a very real question and one which warrants serious consideration right now!

The stone industry currently has a program underway to try to determine those areas in which the power currently used in the production process can be conserved. There are projections with respect to stone production which tend to indicate that by the year 2000, the industry's ability to supply the demand will be severely limited if the same amount of power per ton for production is used. By the year 2000 the industry's ability to meet demands will be jeopardized if power conservation is not achieved. The dry collection option, in effect, nullifies power conservation techniques. Roughly increased power must be saved before reaching the break-even point, but even at the break-even point, the production demands will not be met by the year 2000.

The second control option is the wet suppression system. This is a highly-effective system. Go out to a typical plant operation and look at it with those suppression systems operating. Have the operator turn them off and look at it again, 15 minutes later. One can definitely see what those suppression systems are doing. The biggest single problem is that it cannot be measured quantitatively. Are suppression systems 50% effective--90% effective or are they

95% effective? As yet, there is no answer. They are economical to use from the standpoint of capital investment, operating cost and negligible power consumption.

Some 60 operations with only suppression equipment operating, were observed, with ambient air samples taken outside the plant boundaries. (Remember, there was the full effect of all the fugitive sources, and the only control in the plant itself was the wet suppression equipment.) The same dispersion model used to measure the impact of stack emissions from crushed stone operations was taken and worked backwards. The ambient air levels converted to an equivalent stack emission of approximately 0.02 grains per standard cubic foot. This was a measure of the full impact of all fugitive sources, as well as a plant controlled by simply wet suppression equipment.

One other area of importance is types of regulations...not specific regulations, but the types of regulations which have been applied over the past 10 or 12 years. There are the stack emission codes which can be applied if the industry either elects or is forced to create stacks. Those are very readily understandable. One can measure whether or not one complies with a stack emission code. The fugitive sources have been handled by rather subjective regulations. A typical one might say that an operator or an individual shall take reasonable precautions to control fugitive emissions and such precautions might include a whole list of things. The problem with this is that nobody--not the control agency enforcing the regulation nor the operator upon whom the regulation is being enforced--can determine whether or not compliance is achieved. These types of regulations have presented considerable problems.

Dr. Blackwood mentioned the respirable fraction, which he defined as that below 10 microns, and then mentioned the total suspended portion as being less than 50 microns making no reference at all to those dusts above 50 microns. The question was raised, from the audience, that those dusts are, in fact, nuisance dusts and ought to be controlled. That is true. However, whether or not they ought to be controlled under air pollution regulations is another question, particularly as they relate to the stone industry. It is almost virtually impossible, for one reason or another, in the country today, to open a new crushed stone or sand and gravel operation, the reasons being zoning restrictions, land use planning restrictions, or general public apathy. All plants operate by virtue of operating permits, which are reviewable. These larger dusts that settle in the immediate area of a plant are viewed primarily as a public relations problem. It just simply makes good sense to do something about them if one expects to continue to operate. The industry is making an effort to control fugitive emissions by oiling, watering, or even paving haul roads, and quarry floor areas, by shielding stockpile areas and by planting substantial vegetation. Incidentally, a tree buffer is an extremely effective mechanism for controlling large fugitive dust particles. During one of the first studies it was observed in the fall of the year, the settled dust concentrations tended to increase, even though the plant operations were decreasing. In the spring season, they tended to decrease, even though the activity at the plant was increasing. This phenomena was observed for a period of about two years and correlated very well with the foliation and defoliation of the trees.

SUMMARY

The above are some of the problems and concerns the crushed stone industry is facing. The biggest concern has to do with the tendency to more or less force an industry, by virtue of regulation, to move towards collection equipment. Our industry is not convinced that, all things considered, that this is the best approach for the stone quarry. The entire plant area and the quarry operation can be treated and controlled as a fugitive source. Its area of impact is extremely limited. The number of individuals involved or affected is another area that more emphasis ought to be placed upon. Dr. Blackwood touched on it in his remarks. For the most part, a great majority of quarry operations tend to be located in rural low-density population areas. The number of individuals affected and the effect upon those individuals should dictate more strongly the nature and type of controls to be demanded by various control agencies. Test methods to truly measure the impact of open sources are sorely needed. Most of the work up to now has been done by inference. Available tests are run and based upon the results of those tests the impact and/or true levels have been inferred. A method of assessment must be developed so that a move away from subjective toward more definitive control regulations can be achieved. Industry, the public, and control officials should know the true impact of a given fugitive dust source.

DETERMINING EMISSIONS MEASUREMENTS NEEDS FOR AN EMERGING
INDUSTRY-ADVANCED FOSSIL FUELS UTILIZATION*

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DETERMINING EMISSIONS MEASUREMENTS NEEDS FOR AN EMERGING INDUSTRY-ADVANCED FOSSIL FUELS UTILIZATION

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Introductory Remarks

Converting coal or the organic matter in oil shale to petroleum-like liquids or to fuel gases is predicted (1) to be a major component of the nations energy base. Liquid products may also constitute a significant source of raw materials for the petrochemical industry. Successful processes are likely to constitute a new major industry in this country.

The many processes (2-5) now under study have the common objective of extracting hydrocarbons with an atomic hydrogen to carbon ratio approximating that of natural crude oils or natural gas from the solids. Pyrolysis and hydrogenation at elevated pressure and temperature are therefore required to chemically convert the solids to usable liquids or gases. The COED Process, designed to produce a high quality char, is illustrated in Figure 1 as an example. The complexity of the starting material and of the chemical reactions occurring during conversion suggest the possibility of both point and fugitive emissions. Evidence of potential occupational health problems is available in the literature (6-10) for coal hydrogenation.

The existence or extent of environmental and health problems cannot be determined at present because commercial scale plants do not exist. Few processes have yet reached the demonstration plant scale. Methods are required, however, to quickly and reliably assess the extent of the problem as large

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² Biology Division

scale operations are initiated. Data generated in the course of methods development can also be useful in designing control technologies. The finding (11) that conditions can be modified in the Synthane coal gasification process to reduce emissions without sacrificing conversion efficiency provides additional impetus for early emissions characterization.

This paper presents one approach to establishing reliable methods and generating data of value in prioritizing environmental and health studies. Measurements needs are identified through an experimental assessment of existing methods.

Chemical Screening of Complex Mixtures

Solid, aqueous, gaseous, and liquid emissions (4) are theoretically possible from each process. Liquid products themselves must be considered until it is demonstrated that they can be transported and handled as are natural crudes. Each of these materials is expected to consist of a highly complex mixture of chemicals varying in chemical type and concentration prior to applying emission control methods.

At least three methods are commonly used to determine the chemical nature of complex mixtures. Individual constituents, e.g., benzo(a)pyrene, indicative of chemical classes, e.g., polynuclear aromatic hydrocarbons, can be determined. The material can be subjected to class fractionation to obtain a weight percent distribution of constituent types. Multicomponent chromatographic profiling or direct spectral analysis of the untreated material can be carried out to estimate chemical nature and complexity.

Each of these approaches has been used in the work reported here. Multicomponent chromatographic profiling, particularly when combined with class fractionation or the isolation of specific subfractions has been found most useful. Individual constituents must also be determined because biological effects are stereospecific.

Methodology

Samples which have been surveyed in varying detail to date (12-16) and their sources are summarized as follows:

- (a) Coal liquefaction product from the Synthoil Process and the aqueous condensate from the Synthane Gasification Process courtesy of the Pittsburgh Energy Research Center.
- (b) Samples from the points in the COED Pyrolysis Process numbered in Figure 1 courtesy of FMC.
- (c) Shale oil from an above ground simulated insitu retorting courtesy of the Laramie Energy Research Center. Product water was obtained in-house by batch centrifugation of the oil-water emulsion.
- (d) A Louisiana-Mississippi Sweet crude oil courtesy of Dr. J. A. Carter of the Analytical Chemistry Division, Oak Ridge National Laboratory.

Gas samples and product headspace volatiles are profiled using on-column cryothermal trapping and subsequent temperature programmed capillary column gas liquid chromatography (12). Polynuclear aromatic hydrocarbons are isolated as a group by sequential liquid-liquid extraction followed by florisil and alumina column chromatography (15). Carbon-14 isotope dilution is used to locate eluting constituents and compute recoveries. The isolate is profiled gas chromatographically using a 22 ft. long by 1/8 inch O.D. glass column of 3% Dexsil 400 on 80/100 mesh HP Chromosorb G programmed from 110°C to 320°C at a rate of 1°C/min. Benzo(a)pyrene is determined using the same extraction procedure but is isolated from benzo(e)pyrene and other isomers by acetylated cellulose paper or column chromatography and determined spectrophotofluorimetrically. Alkyl chrysenes are determined using the method of Hecht (17). Aqueous samples

are gas chromatographed without prior treatment using Tenax as the column packing (13). The class fractionation method (16) used most extensively is that developed (18) to elucidate the carcinogenic properties of condensed tobacco smokes. Most of the constituents listed have been identified only by isolation and co-chromatography. Identifications must therefore be considered preliminary.

Preliminary Results

Figure 2 illustrates the results of direct gas chromatographic profiling of a stack gas sample and of the material volatilized from Synthoil at 50°C. The stack sample is illustrative of a point emission and the volatiles are illustrative of a possible fugitive emission. The profiles visualize organic constituents containing from one to approximately seven carbon atoms. Very low molecular weight compounds, hydrocarbons containing from one to four carbon atoms, are not resolved under the conditions used. Both samples are seen to consist of a large number of isomeric organic compounds. Use of the flame photometric detector allows selectively visualizing sulfur containing constituents including hydrogen sulfide, carbonylsulfide, methyldisulfide, and thiophene in gas samples (12).

Aqueous samples from the COED Process, Synthane Process, and simulated in-situ oil shale retorting also contain a large number of constituents (Figure 3) at high concentrations (Table 1). Waters derived from coal processing are generally found to contain phenol, the cresols, and other isomeric phenols as the primary contaminants. Oil shale derived water contains a homologous series of saturated carboxylic acids. Flame-photometric detection indicates (12) the presence of at least ten sulfur containing constituents in a product separator liquor from the COED process. All of these constituents would be greatly reduced in effluent waters by standard control methods.

Direct chromatographic profiling easily and rapidly provides a great deal of information but is limited in two important ways, (a) chromatographic conditions optimized to visualize non-polar compounds often preclude visualizing polar compounds and vice versa, and (b) compounds present at low concentrations are obscured by the major constituents. Procedures which combine isolating the compound class of interest and chromatographic conditions optimized for profiling that class represent the next level of complexity.

Figure 4 illustrates the chromatographic profiles of the polynuclear aromatic hydrocarbon (PAH) isolates from condensed cigarette smoke and a coal liquefaction process. The comparison is more than academic because this fraction is thought to be the primary contributor to mouse skin carcinogenicity of condensed smoke and skin contact is a primary health concern (9) in handling coal liquids. For purposes of quantitative comparison, the coal derived profile was obtained at a detector sensitivity one eighth that used for the smoke condensate. The PAH isolate from the coal product contains a wider variety of constituents present at concentrations at least an order of magnitude greater than in smokes.

Multicomponent profiling of isolates is readily carried out for qualitative or semi-quantitative analytical purposes. Table 2 summarizes estimates of PAH's in products and related aqueous samples obtained using the profiles. Multicomponent quantitative determination is also possible if recoveries and identities are known. Recoveries of 80% or more and relative standard deviations of 10% for triplicate determinations are common with experience.

The complexity of the isolates and the importance of specific isomers limits the utility of even quantitative multicomponent analyses. In the examples shown, benzo(a)pyrene cannot be distinguished from benzo(e)pyrene. Methods optimized for the determination of selected constituents must supple-

ment profiling. We find (12) approximately 40 ppm benzo(a)pyrene in one synthetic crude oil by extractive and chromatographic isolation, isotope dilution, and fluorescence measurement. Interest in methyl chrysene concentrations requires a comparable isolation procedure plus the use of Diels-Adler adducts to remove interfering benzanthraces. Results of this study indicate the presence of chrysene (98 ppm), 2 methyl chrysene (102 ppm), 3-methyl chrysene (106 ppm), and 6-methyl chrysene (64 ppm). The highly carcinogenic (17) 5-methyl chrysene has not yet been adequately resolved from the 4-methyl isomer to establish its presence but the combined concentration of both constituents is less than 20 ppm.

Figure 5 illustrates a traditional approach to the class fractionation of complex organic mixtures. The percentages included in the figure correspond to the weight percentage of those fractions obtained when syncrude from the COED Process is subjected to the procedure. The primary results of this approach are weight percent distributions of constituent types and partially defined fractions for further study. The procedure has been applied to products and aqueous samples from both shale and coal processing. Table 3 illustrates results obtained for the COED and Synthoil products. Reproducibility is typical of qualitative procedures requiring much manual manipulation. Studies are presently underway of column chromatographic fractionation methods which promise to be more reproducible and less likely to produce chemical artifacts.

Sufficient sensitivity and resolution is likely to allow the detection of chemicals of every type. High molecular weight paraffins can be detected in shale derived waters, as is illustrated in Table 4, for example. Information is required to prioritize chemical types and sample types for measurements research. Chemical fractionation followed by biological screening is in use here (16) to prioritize chemical needs while developing the bioassay systems.

Mutagenicity Screening of Complex Mixtures

The potential predictive value of assays for genetic damage and the correlation with carcinogenic damage has been emphasized in a number of recent reports (19, 20, 21). The wide applicability of the bacterial test system developed by Ames has been illustrated for a large number of "pure" compounds but also may be used as a prescreen for ascertaining the genetic and potential carcinogenic hazard of complex environmental effluents or products e.g., tobacco smoke condensates (22), soot from city air (23), hair dyes (24) and in our preliminary work, synthetic crude oil (16). Furthermore, the overall economy and rapidity of the Ames test, along with the high resolution, also allows the detection of potential mutagenic/carcinogenic activity in human body fluids, e.g., monitoring urines from occupationally exposed workers (20).

The Ames test system is a highly sensitive yet simple bacterial assay for chemical mutagens. Compounds (or mixtures) are tested with a group of well-characterized mutants of Salmonella typhimurium requiring histidine for growth. Simply stated, the assay detects genetic damage induced by chemicals by the reversion of these specific strains to the wild-type or prototrophic state (the ability to grow without histidine supplement). Additionally, the particular type of genetic damage - base alterations, frame shifts (addition or deletions) - can be detected with the appropriate strain.

Furthermore, since we now realize that many carcinogens and mutagens require metabolic activation in order to reach their ultimate form, the test can be modified to include an in vitro activation of the chemical under test. Here, homogenates from rat or human liver or other tissues can be applied and the mutagenic activity of the activated form can be detected. The inclusion of this metabolic activation has led to the detection of a wide array of carcinogens as mutagens (21).

The Salmonella strains and procedures used have been described in detail by Ames (23). For the study of the feasibility of application of mutagenicity testing to environmental effluents and crude products from the synthetic fuels technology, we attempted to perform wide range screening with two of the highly sensitive strains known to respond to a wide variety of known mutagens/carcinogens. The working hypothesis was that sensitive detection of potential mutagens in fractionated complex mixtures could be used to isolate and identify the biohazard. In addition, the information could be helpful in establishing priorities for further testing, either with other genetic assays or carcinogenic assays. Finally, the procedures might show utility in monitoring plant processes, effluents, or personnel early in the formation of the engineering and environmental technology that will eventually evolve in the synthetic fuels industry. The approach and preliminary results cited here show that the coupled analytical-biological scheme is a feasible research mechanism and is applicable to the ascertainment of potential human health hazard of a wide variety of environmental exposures, either occupationally or to the population in general.

Methodology

The Salmonella strains used in the various assays are listed below. All strains were obtained through the courtesy of Dr. Bruce Ames, Berkeley, California.

Salmonella typhimurium Strains

TA 1535	hisG46, uvrB, rfa	(missense)
TA 100	hisG46, uvrB, rfa	(missense plus R factor)
TA 1537	hisC3076, uvrB, rfa	(frameshift)
TA 1538	hisD3052, uvrB, rfa	(frameshift)
TA 98	hisD3052, uvrB, rfa	(frameshift plus R factor)

In the screening of fractionated materials the two strains TA98 and TA100 were generally employed. Standard experimental procedures have been given by Ames, McCann, and Yamasaki (23). Briefly, the strain to be treated with the potential mutagen(s) is added to soft agar containing a low level of histidine and biotin along with varying amounts of the test substance. The suspension containing approximately 2×10^8 bacteria is overlaid on minimal agar plates. The bacteria undergo several divisions with the reduced level of histidine, thus forming a light lawn of background growth on the plate and allowing the mutagen to act. Revertants to the wild-type state appear as obvious large colonies on the plate. The assay can be quantitated with respect to dose (added amount) of mutagen and modified to include "on-the-plate" treatment with the liver homogenate required to metabolically activate many compounds.

Fractions and/or control compounds to be tested were suspended in dimethylsulfoxide (DMSO, supplied sterile, spectrophotometric grade from Schwarz-Mann) to concentrations in the range of 10-20 mg solids. The potential mutagen was in some cases assayed for general toxicity (bacterial survival) with strain TA1537. Generally, the fraction was tested with the plate assay over at least a 1000-fold concentration range with the two tester strains TA98 and TA100. Revertant colonies were counted after 48 hours incubation. Data were recorded and plotted versus added concentration and the approximate slope of the induction curve was determined. It is assumed that the slope of the linear dose-response range reflects the mutagenic activity. Positive or questionable results were clarified using a narrower range of concentrations. All studies were carried out with parallel series of plates plus and minus the liver enzyme preparation for metabolic activation. The background lawn of bacterial growth was routinely examined so that any effects attributed to massive cell death and subsequent growth of the few surviving bacteria (availability of more histidine)

could be differentiated from mutation induction. Routine controls demonstrating the sterility of samples, enzyme or S-9 preparations, and reagents were performed. Positive controls with known mutagens were carried out in order to recheck strain response and enzyme preparations.

Preliminary Results

To demonstrate the feasibility of the coupled analytical-biological approach, we derived primary fractions from a number of crude products and effluents from various fossil fuel technologies. For example, Table 5 lists the mutagenicity testing results from three crude oils: Syncrude from the COED process, shale oil, and a natural crude. The important neutral fraction was subfractionated and the individual values are listed. Since in most cases the original crude product was too toxic to test, the total represents the summation of the assays of all fractions. We have assumed that the most accurate measure of the total potential of the neutral fraction is the sum of the tested subfractions.

An overview of the preliminary results points to a number of consistencies: (a) all crudes showed some mutagenic potential, (b) the neutral and basic fractions showed activities regardless of the source of the sample, and (c) the relative total mutagenic potentials vary from 516 revertants/mg for Syncrude, 178 revertants/mg for shale oil, to 76 revertants/mg for the natural crude oil. Whether these results reflect a comparative biohazard is not the point in question here. The results simply show that biological testing - genetic reversion assays in this case - can be carried out with the newly developed tester systems but only when coupled with the appropriate analytical separation schemes.

In addition to the obvious bias that could accompany the choice of samples and their solubility, or the time and method of storage, a number of biological

discrepancies can also enter into the determinations. For example, concomitant bacterial toxicity can nullify any genetic damage assay that might be carried out; the choice of inducer for the liver enzymes involved can be wrong for selected compounds; the choice of strain could be inappropriate to, again, selected compounds; and additionally, the applicability of the Salmonelle test to other genetic assays and the validation of the apparent correlation between mutagenicity and carcinogenicity remains a point of significant fundamental research. Furthermore, the short term assays chronically show negative results with, e.g., heavy metals. Similarly, compounds involved in or requiring co-carcinogenic phenomena would presumably go undetected. However, in the context of a prescreen to aid the investigators in ordering their priorities, the short-term tests appear to be a valid testing approach to address the dilemma of the magnitude of the number of hazardous compounds and complex mixtures that man encounters in his environment.

Perhaps more appropriate to environmental effects, we have extended the fractionation procedures and mutagenicity assay to the organics recovered from a number of aqueous samples from various technologies. In parallel with the assays listed above, we have investigated product water from the shale oil process, the separator liquor from the COED process, and condensate from the Synthane process. Again, the mutagenic potential of the various fractions can be ascertained. The studies are not being extended to the isolation and identification of the active components. In correlation with the biological effort, the chemical analyses of the materials and fractions are being extended. A concentrated effort designed to genetically assay the known or predicted constituents of effluents from various synthetic fuel processes is also underway, utilizing a number of biological systems in a "tier approach" to mutagenicity testing.

Comments

The single most important measurement "need" encountered in our chemical studies to date is the need to prioritize measurements research activities. The number of combinations of processes, sample types, and chemicals available for study is staggering. Bioassay data when health effects are of interest and environmental data when environmental impact is of interest provide a rational basis for prioritizing research. If carcinogenesis is of interest, results of the Ames test are accepted as indicators of carcinogenesis, and the additivity of mutagenic activities of chemical fractions are accepted, bioassay results reported here suggest that the neutral and ether soluble basic constituents of coal derived samples should receive priority attention. The assumptions leading to this conclusion and the bioassay methodologies require considerable basic research for validation but any guidance at this point is invaluable.

A related problem (raised by Dr. James Dorsey, EPA/RTP following this presentation) is that of "decision points" - quantifying bioassays responses, at least in a relative sense, so that subsequent action is indicated. Such action can range from prioritizing samples for study to dictating control technology requirements. "Natural" materials of epidemiologically known health effect, cigarette smoke for inhalation exposure or soot for skin contact as examples, might be used to establish baseline biological responses.

The large number of questions associated with the relationship between bioassay systems, human health effects, and exposure to complex mixtures of materials, preclude positive judgments of environmental and health impacts of advanced fossil fuels processes. Judgments are made essentially impossible by the absence of commercial scale conversion plants and uncertainties about the relationship between aged untreated samples from pilot plant experiments

and materials issued from commercial scale operations which will incorporate environmental control methods. Studies carried out now must emphasize methods development in preparation for process evaluation.

Measurements research needs identified from work referenced and reported here include:

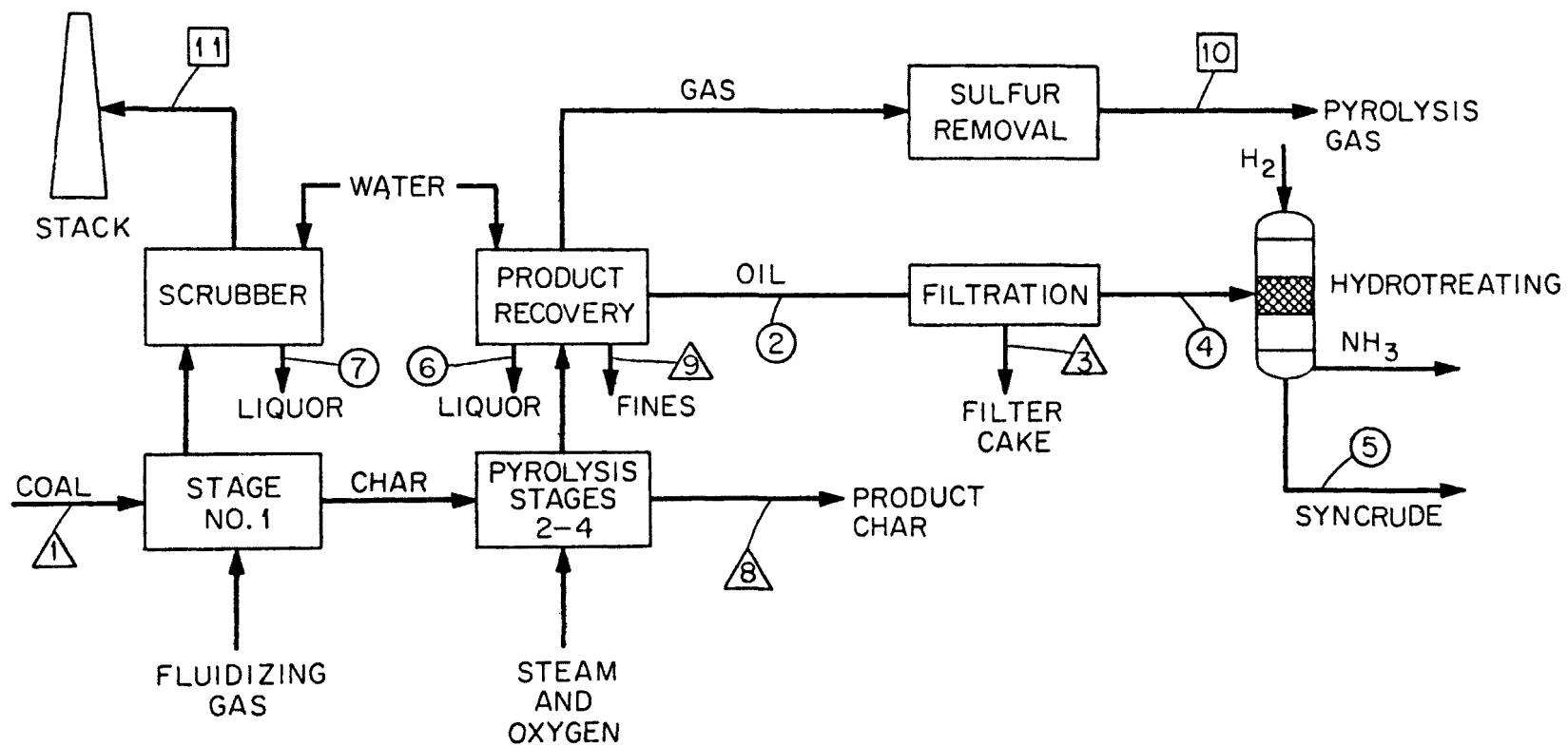
- (a) Development of chemical class fractionation procedures for bio-testing support which are free of artifact formation and routinely applicable at preparatory (100+ gm) levels.
- (b) Development of multicomponent quantitative analytical methods for carcinogenic, co-carcinogenic, and tumor promoting fractions.
- (c) Identification and quantitation of stereoisomers in the polynuclear aromatic hydrocarbon subfractions of liquid products and aqueous effluents.
- (d) Identification of individual constituents or physico-chemical parameters (e.g., fluorescence) which can be monitored as indicators of the polynuclear aromatic hydrocarbon, weak acid, and basic fractions of liquid products and effluents. Development of monitoring instrumentation follows.
- (e) Identification of constituents in aqueous leachates from spent shale or solid residues from coal processing.
- (f) Development of methods for the rapid, highly selective determination of stereoisomers in environmental samples.
- (g) Identification of individual constituents in airborne materials available for occupational inhalation exposure.

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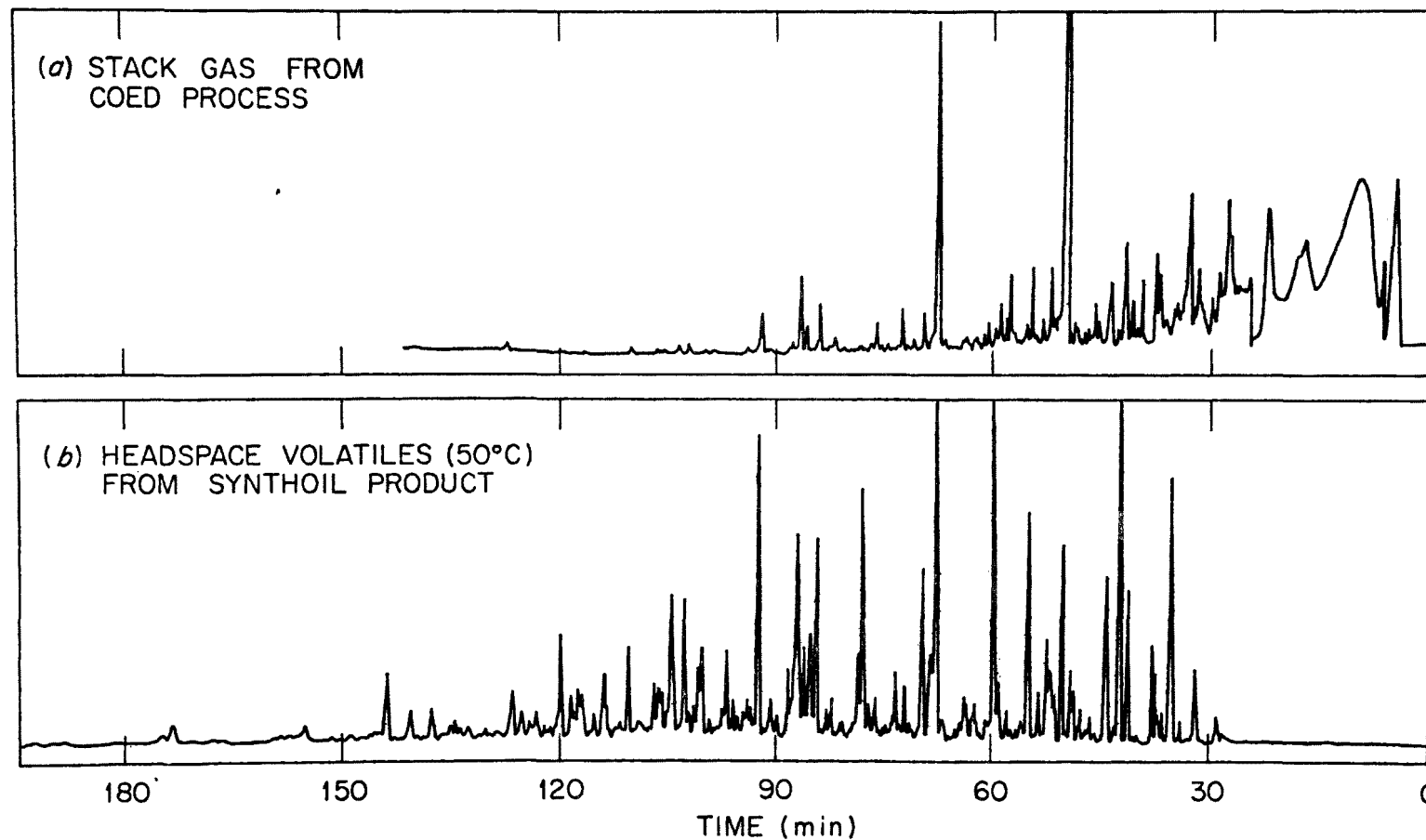
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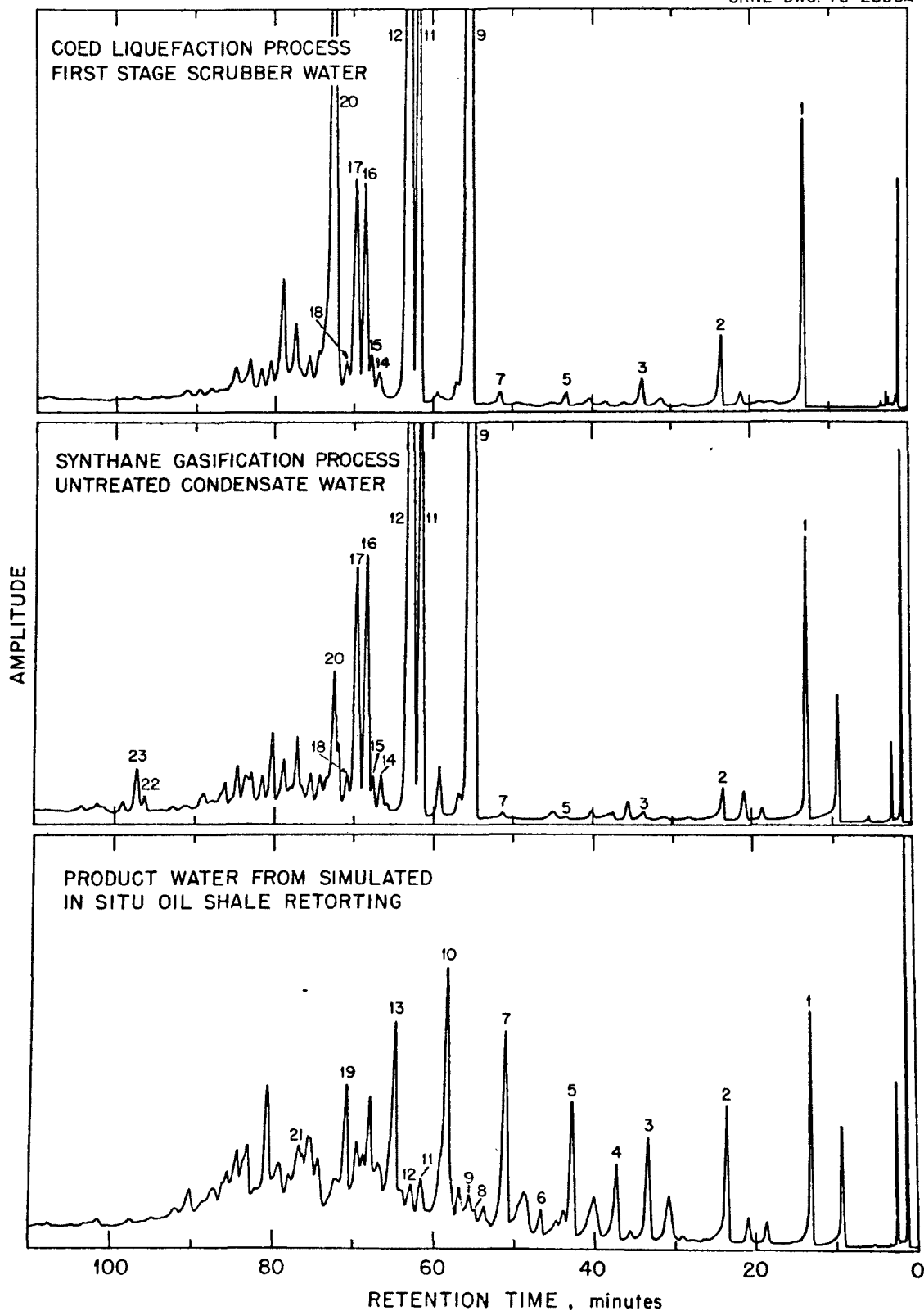
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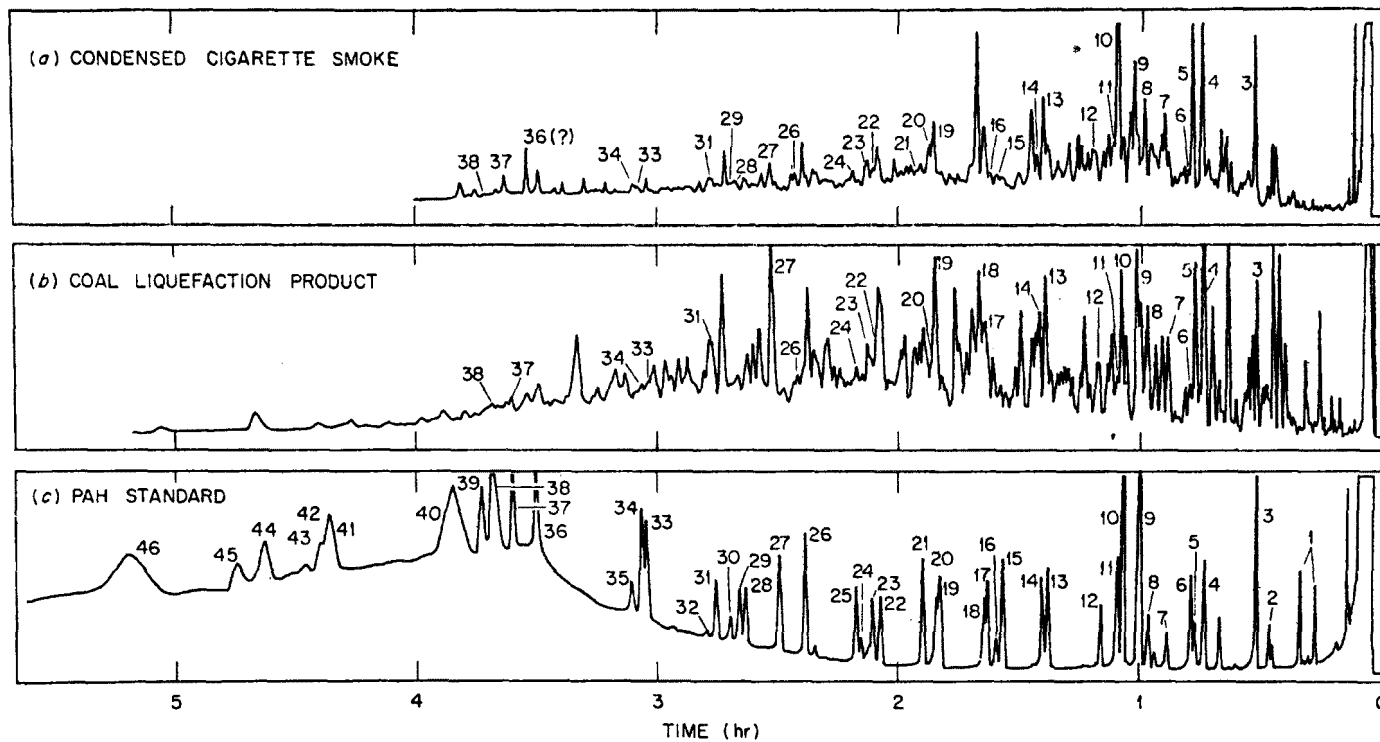
- Figure 1. 75-4852 Schematic Representation of the COED Coal Conversion Process with Sampling Points Indicated
- Figure 2. 75-12340 Capillary Column Gas Chromatographic Profiles of Organic Constituents in Untreated Stack Gas and Headspace
- Figure 3. 76-2806A Direct Injection Gas Chromatographic Profiles of Coal and Shale Derived Waters
- Figure 4. 75-7368R Gas Chromatographic Profiles of Polynuclear Aromatic Hydrocarbon Isolates
- Figure 5. 75-9451 Extractive Fractionation of COED Syncrude



COED Process.







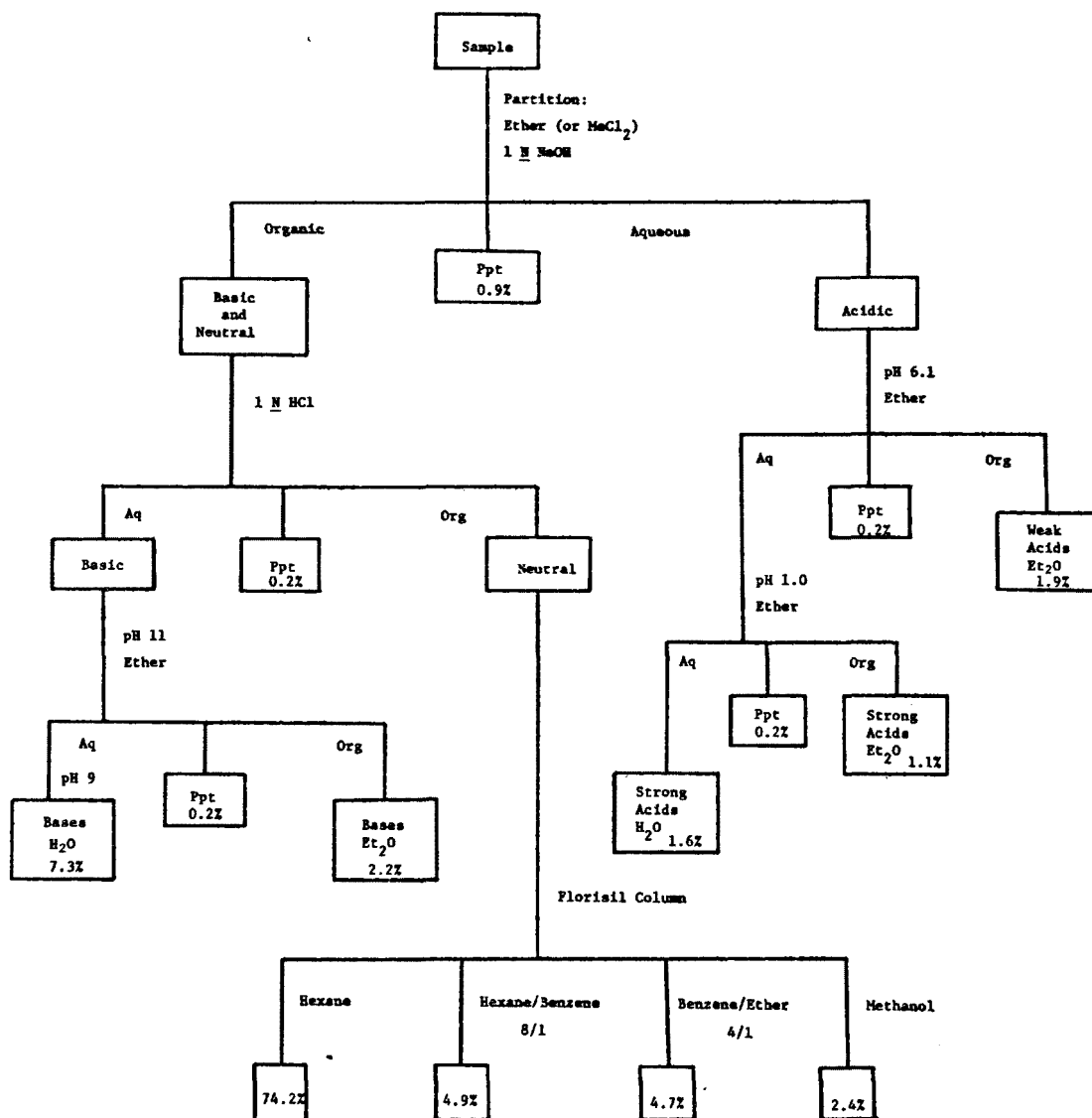


Table 1

Organic Compounds Determined in By-Produce Waters
from Fossil Fuel Conversion Processes

Peak ¹	Co-Chromatographic Identification	Concentration (µg/ml)		
		Oil Shale Retorting	Synthane Coal Gasification	COED Coal Liquefaction
1	Acetic Acid	600	620	600
2	Propanoic Acid	210	60	90
3	n-Butanoic Acid	130	20	40
4	Acetamide	230	--	--
5	n-Pentanoic Acid	200	10	30
6	Propionamide	50	--	--
7	n-Hexanoic Acid	250	20	30
8	Butyramide	10	--	--
9	Phenol	10	2100	2100
10	n-Heptanoic Acid	260	--	--
11	o-Cresol	30	670	650
12	m & p-Cresols	20	1800	1800
13	n-Octanoic Acid	250	--	--
14	2,6-Dimethylphenol	--	40	30
15	o-Ethylphenol	--	30	30
16	2,5-Dimethylphenol	--	250	220
17	3,5-Dimethylphenol	--	230	240
18	2,3-Dimethylphenol	--	30	30
19	n-Nonanoic Acid	100	--	--
20	3,4-Dimethylphenol	--	100	900
21	n-Decanoic Acid	50	--	--
22	α-Napthol	--	10	--
23	β-Napthol	--	30	--

¹ Peak number in Figure 3

Table 2

Estimation of Polynuclear Aromatic Hydrocarbons
in Coal and Shale Derived Samples

Peak Number ¹	Co-Chromatographic Identification	Products micrograms per gram (ppm)			Waters micrograms per liter (ppb)	
		Crude	Shale Oil	Synthoil	Shale Retort	Synthane Condensate
1	cis-and trans-Decahydronapthalene					
2	1,4-Dihydronapthalene					
3	Napthalene	200	790	440	86	160
4	2-Methylnapthalene	650	4600	1400	330	1300
5	1-Methylnapthalene	290	2900	430	110	32
6	Azalene					
7	Biphenyl	23	190	84	33	2
8	2,6-Dimethylnapthalene	250	3400	250	46	48
9	1,3 + 1,6-Dimethylnapthalene	670	5100	170	250	66
10	Butylated hydroxytoluene					
	1,5 + 2,3-Dimethylnapthalene	220	2000	180	100	24
	1,2-Dimethylnapthalene	37	1100	IR ²	38	10
11	Acenapthalene					
12	Acenapthene	IR	280	IR	45	IR
13	Fluorene	220	940	345	180	21
14	9,10-Dihydroanthracene	ND ³	26	1700	27	2
15	9-Methylfluorene	ND	410	90	IR	ND
16	9,10-Dihydrophenanthrene					
17	Octanthrene					
18	1-Methylfluorene	140	980	1900	160	ND
19	Phenanthrene + 1,3,6-Trimethylnapthalene }	290	620	1400	260	ND
20	Anthracene					
21	1-Phenylnapthalene					
22	2-Methylanthracene	ND	IR	ND	210	ND
23	1-Methylphenanthrene	330	IR	ND	130	ND
24	2-Phenylnapthalene					
25	9-Methylanthracene	3	ND	IR	35	ND
26	Fluoranthene	68	400	380	18	ND

¹Number of chromatographic peak in Figure 4

²IR = Incomplete resolution

³ND = Not detected

Table 2 (Cont'd)

Peak Number	Co-Chromatographic Identification	Products micrograms per gram (ppm)			Waters micrograms per liter (ppb)		
		Crude	Shale Oil	Synthoil	Shale Retort	Synthane	Condensate
27	Pyrene	IR	170	4300	120		ND
28	1,2-Benzofluorene	22	53	IR	10		ND
29	2,3-Benzofluorene	13	140	IR	5		ND
30	4-Methylpyrene						
31	1-Methylpyrene	36	70	620	61		ND
32	5,12-Dihydrotetracene						
33	1,2-Benzanthracene	IR	IR	270	37		ND
34	Chrysene + Triphenylene	IR	IR	130	5		ND
35	2,3-Benzanthracene						
36	7,12-Dimethylbenz(a)anthracene						
37	1,3,5-Triphenylbenzene }						
38	1,2-+3,4-Benzopyrene	20	180	IR	5		ND
39	Perylene	31	22	ND	ND		ND
40	3-Methylcholanthrene + Unknown	ND	20	200	6		ND
41	1,2,5,6-+1,2,3,4-Dibenzanthracene						
42	o-Phenylene pyrene						
43	Picene	ND	ND	380	ND		ND
44	1,12-Benzoperylene	ND	ND	1500	ND		ND
45	Anthanthracene	ND	ND	80	ND		ND
46	3,4,9,10-Dibenzopyrene + Coronene						

Table 3

ORNL DWG. 76-5075

Fractionation of Coal Liquefaction Products

Fraction	Quantity Found, %		Synthoil
	COED		
	\bar{x}	RSD, %	
1. NaOH Insol.	0.9	34	14.1
2. WA _I	0.2	86	2.0
3. WA _E	1.9	18	6.5
4. SA _I	0.2	41	0.1
5. SA _E	1.1	50	1.9
6. SA _W	1.6	82	1.7
7. B _{Ia}	0.2	29	3.6
8. B _{Ib}	0.2	38	0.3
9. B _E	2.2	16	1.6
10. B _w	7.3	89	0.6
Neutrals			
11. Hexane	74.2	2	27.6
12. Hexane/Benzene	4.9	47	7.4
13. Benzene/Ether	4.7	17	21.7
14. Methanol	2.4	17	11.1
Total Recovery	101.7	9	100.2
n	4		1
Sample wt., g	4.4—11.9		15.2

Table 4

**Determination of n-Alkanes
in Shale Oil and Shale Oil By-Product Water**

Peak	Compound	Concentration	
		Shale Oil (mg/g)	Shale Oil By-Product Water (μ g/l)
1	C ₁₁ H ₂₄	7.55	67
2	C ₁₂ H ₂₆	6.65	66
3	C ₁₃ H ₂₈	6.20	65
4	C ₁₄ H ₃₀	4.80	62
5	C ₁₅ H ₃₂	3.30	45
6	C ₁₆ H ₃₄	4.50	53
7	C ₁₇ H ₃₆	8.60	105
8	C ₁₈ H ₃₈	8.80	106
9	C ₁₉ H ₄₀	5.60	70
10	C ₂₀ H ₄₂	4.80	62
11	C ₂₁ H ₄₄	5.70	73
12	C ₂₂ H ₄₆	4.50	59
13	C ₂₃ H ₄₈	5.00	67
14	C ₂₄ H ₅₀	3.60	48
15	C ₂₅ H ₅₂	3.90	53
16	C ₂₆ H ₅₄	3.45	50
17	C ₂₇ H ₅₆	3.35	51
18	C ₂₈ H ₅₈	2.90	48
19	C ₂₉ H ₆₀	3.20	62
20	C ₃₀ H ₆₂	1.00	30
21	C ₃₁ H ₆₄	0.20	27
22	C ₃₂ H ₆₆	0.10	9
23	C ₃₃ H ₆₈	0.65	12
24	C ₃₄ H ₇₀	0.40	5
25	C ₃₅ H ₇₂	0.15	2

Table 5
Mutagenicity Assays of Fractionated Crude Oils *

Fraction ¹		Syncrude (COED)			Shale Oil			Crude Oil		
		% of Total	Fraction rev/mg ³	Total rev/mg	% of Total	Fraction rev/mg	Total rev/mg	% of Total	Fraction rev/mg	Total rev/mg
NaOH Insoluble		1.0	0	--	1.0	256	3	2.9	0	--
Weak Acids, Insoluble		0.1	0	--	0.1	185	<1	0.2	0	--
Weak Acids, Ether Soluble		1.8	0	--	1.2	52	1	0.8	0	--
Strong Acids, Insoluble		0.1	0	--	0.1	0	--	0.2	0	--
Strong Acids, Ether Soluble		0.9	0	--	0.3	159	<1	0.5	115	<1
Strong Acids, Water Soluble		0.4	0	--	0.6	160	1	0.1	236	<1
Bases, Insoluble (a)		0.2	8300	17	0.2	1377	3	0.4	0	--
Bases, Insoluble (b)		0.2	0	--	0.3	800	2	0.1	0	--
Bases, Ether Soluble		2.6	1500	39	7.1	952	68	0.2	175	<1
Bases, Water Soluble		0.4	0	--	0.3	223	1	0.1	0	--
Neutral		82.3	559	460	86.7	112	97	80.7	90	73
Fractionated Neutrals ²		(%-Neutrals)			(%-Neutrals)			(%-Neutrals)		
Hexane	A	87.1	455	396	58.7	40	23	82.0	92	75
	B	2.6	3100	81	2.1	625	13	2.0	50	1
	C	1.1	760	8	1.3	750	10	0.6	168	1
Hexane/Benzene	A	1.6	2120	34	4.4	238	10	3.6	150	5
	B	0.7	2400	17	1.9	340	6	0.7	254	2
	C	0.6	0	--	1.4	320	4	0.5	70	<1
Benzene/Ether	A	4.1	0	--	12.4	65	8	3.5	32	1
	B	0.4	200	1	2.2	142	3	0.3	53	<1
	C	0.2	160	<1	1.3	253	3	0.2	Not Tested	--
Methanol	A	1.1	1520	18	15.1	179	27	2.4	32	1
	B	0.4	400	2	0.5	684	3	0.4	82	<1
	C	0.1	300	<1	0.9	263	2	0.2	74	<1
MeCl ₂		0.2	200	<1	--	--	--	--	--	--
Subtotal (Neutrals)		100.2	--	559	102.2	--	112	96.4	--	90
Total		91.8	--	516	97.9	--	178	86.2	--	76

Table 5 (Cont'd)

¹Initial sample weights: Syncrude, 11.8862g; Shale Oil, 24.0424g; and Crude Oil, 22.3099g.

²Neutral Fraction chromatographed: Syncrude, 9.7823g; Shale Oil, 10.1314g; and Crude Oil, 10.0130g.

³rev/mg = revertants/milligram: number of histidine from Salmonella strain TA98 using plate assay employing 2×10^8 bacteria per plate; values derived from assumed slope of induction curve extrapolated to milligram value. All assays carried out in the presence of crude liver S-9 from rats induced with Aroclor 1254 (gift of Monsanto).

* Louisiana-Mississippi Sweet Crude. Not necessarily representative of all crude.

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NONPOINT SOURCE WATER EMISSIONS:
ENERGY AND INDUSTRY PROCESSES

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NONPOINT SOURCE WATER EMISSIONS: ENERGY AND INDUSTRY PROCESSES

INTRODUCTION

Since the enactment of P.L. 92-500 (Federal Water Pollution Control Act Amendments of 1972), the U. S. Environmental Protection Agency (EPA) has largely directed its water pollution control program at point sources such as process wastewaters discharged through pipes to receiving bodies of water. Most industries and many municipalities will meet the standards of the interim goal of 1977. However, there are many areas in the United States where water quality has not significantly improved even though point sources have been controlled. In such areas nonpoint source water pollution has a major influence on water quality.

You may ask: what is nonpoint source water pollution? While point sources are defined in P.L. 92-500, nonpoint sources are not defined. However, by inference we can define nonpoint sources as the accumulated pollutants in a receiving body of water from runoff due to snow melt and rain, seepage, and percolation contributing to the degradation of the quality of surface and groundwaters. Some of the characteristics of nonpoint sources are:

- diffuse in nature
- intermittent
- site specific
- not easily monitored at their exact source
- related to uncontrollable climate events
- not usually repetitive in nature from event to event.

Figure 1(1) shows a representation of the runoff cycle.

Since nonpoint source is defined by inference, everyone does not agree on a common definition. Some experts define nonpoint source pollution as water activities not requiring an NPDES (National Pollution Discharge Elimination System) permit. This definition excludes one important class of intermittent, diffuse, site specific sources. That is; raw material pile runoff, process spills resulting in runoff, overflow from impoundments, and any other source caused by rainfall such as solids accumulation, which in several cases have been regulated by the NPDES permit system. This definition also includes a source which may be intermittent, but is not caused by climate conditions, and can be monitored at its source. That is; point sources (which may number in the thousands) which have too low a flow to require an NPDES permit.

Based on these definitions, it is easy to see why the emphasis has been on point source controls. Nonpoint sources by their nature are very difficult and costly to

- predict
- monitor
- control

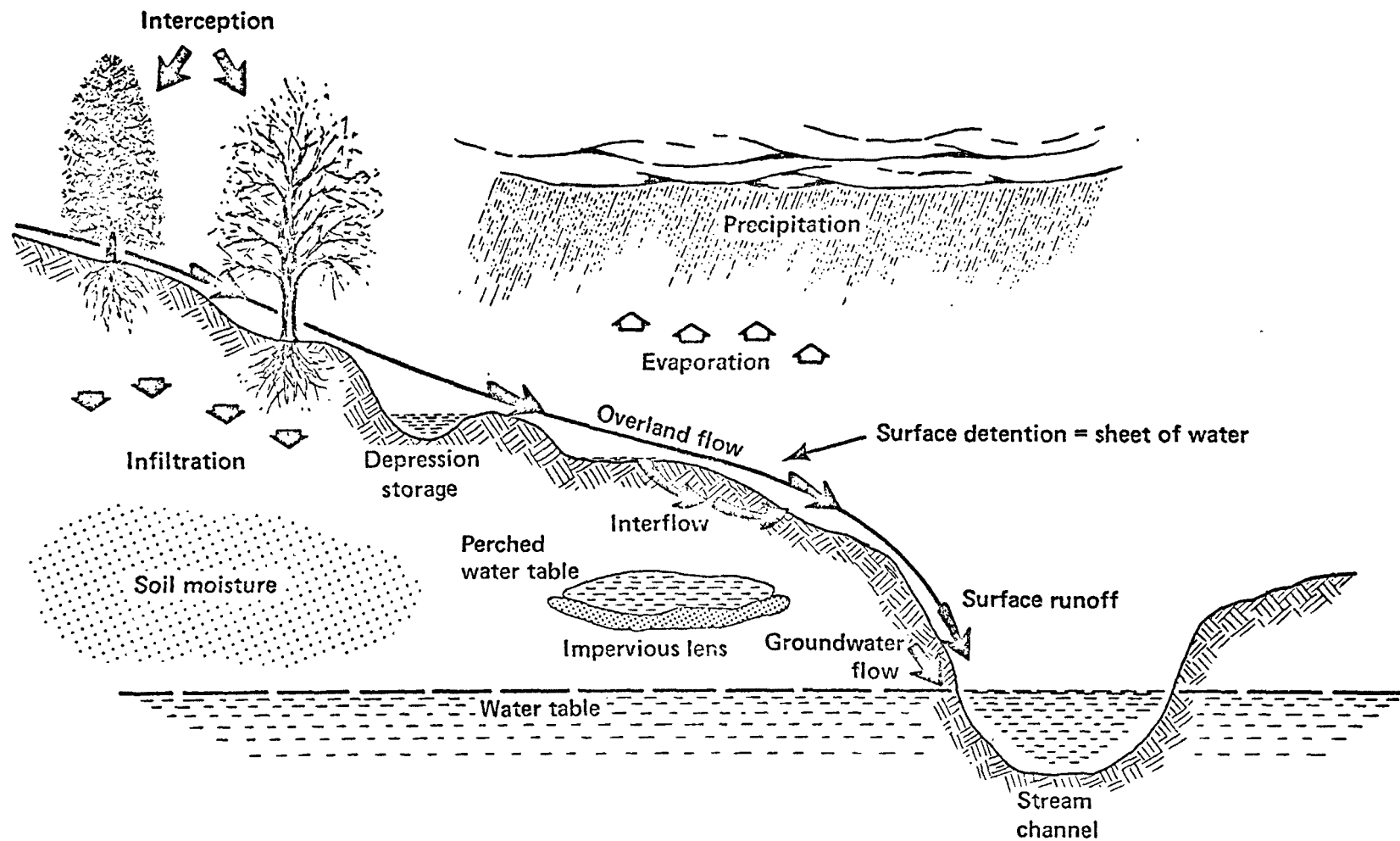


Fig. 1. Simple representation of the runoff cycle.

Now that we have defined nonpoint source water pollution, what are the major sources? Figure 2 is a sketch of potential nonpoint sources which include runoff from urban areas, agricultural and silvicultural activities, construction and recreation areas, wetlands and industrial and mining sites, and salt water intrusion, hydrographic modifications and unregistered point sources. Runoff from urban areas and agricultural activities are known to be of prime concern since much quantification has already been performed. This paper will present a brief overview of a portion of a nonpoint source program performed by TRC for the Industrial Environmental Research Laboratories of EPA. It will address industrial (including mining and energy) processes and look at

- potential nonpoint sources
- measurement methodology
- prediction methodology

Industrial Nonpoint Sources

In looking at potential industrial nonpoint sources, we have selected the following industries:

- energy generation
- timber products
- pulp and paper
- iron and steel
- inorganic chemicals
- petroleum
- fertilizer
- mining (coal, mineral, ore)
- cement
- feedlots
- phosphate manufacturing
- nonferrous metals

Very little work (except acid mine drainage) has been performed in trying to isolate and quantify nonpoint source pollution from industries. Therefore, for each industry we have developed a matrix relating potential sources to categories of pollutants. These pollutants include:

- sediment (suspended and dissolved solids)
- organic materials (including oil and grease)
- metals
- nutrients (nitrogen and phosphorous compounds)
- heat
- sulfates
- acids
- pesticides

Energy Generation

Nonpoint sources depend on the type of power generation, whether fossil fueled (coal, oil, gas) or nuclear powered.

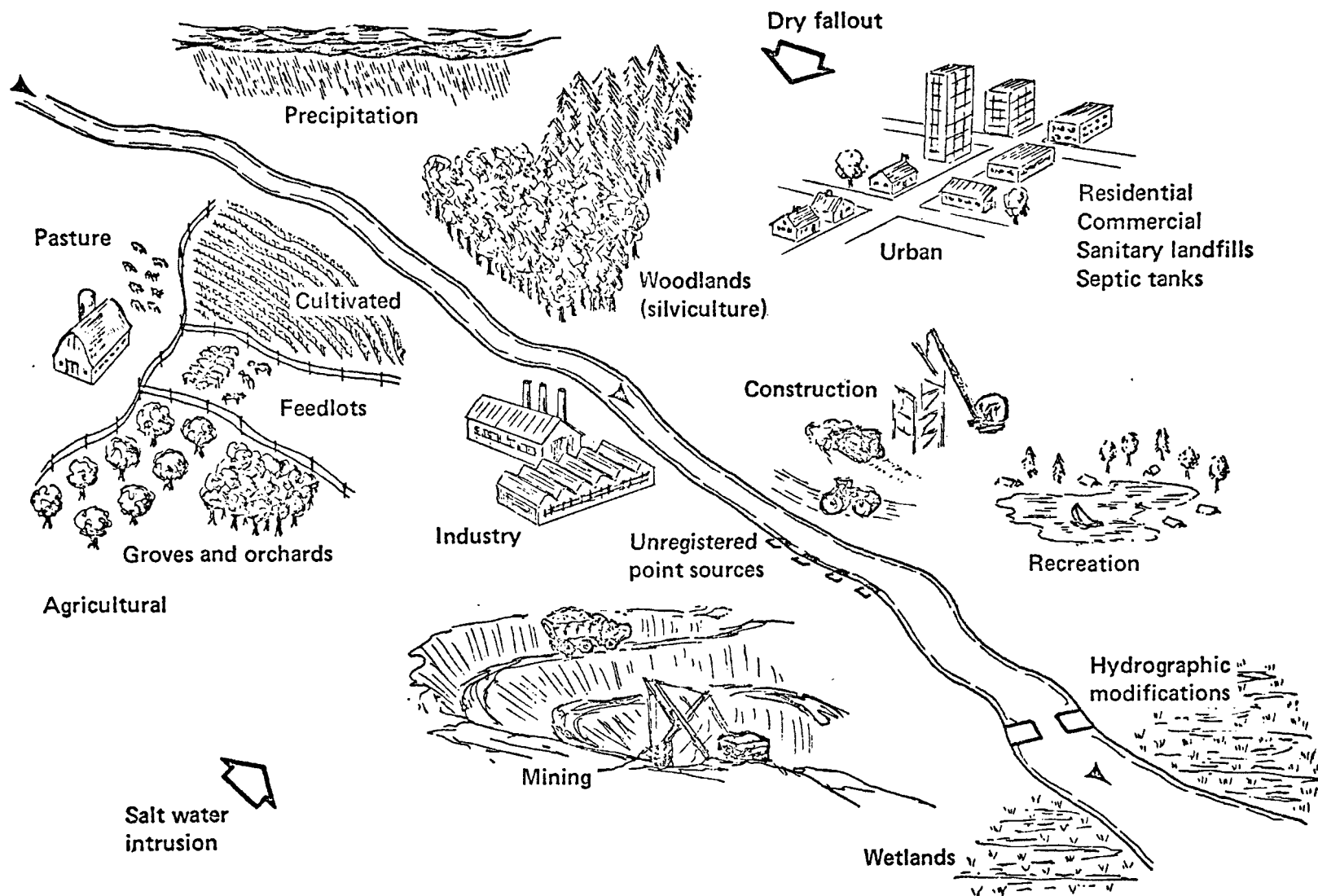


Fig. 2. Nonpoint sources.

Coal fired plants have runoff from coal storage piles and both coal and oil fired plants may have runoff from fly ash storage piles. All fossil fuel plants have the potential of deposited drift from cooling towers and deposited blowdown from steam vents and fallout from fugitive air emissions. Nuclear plants may have deposited drift from cooling towers and deposited blowdown from steam vents. The nonpoint sources from energy generating plants will likely contain sediments (suspended and dissolved solids), pesticides, sulfates, organic materials and acids.

Mining (Coal, Ore & Mineral) - Nonpoint sources include mine drainage, spoil material drainage, runoff from storage piles, runoff from tailing piles, and fallout from fugitive air emissions. Nonpoint source pollutants include metals, organic materials, sediment (suspended and dissolved solids), sulfates and acids.

Iron & Steel - Nonpoint sources include runoff from coal, limestone and iron ore storage piles, process water runoff from ingot and pig casting and process water spills and fugitive and point source air emission fallout. Pollutants include metals, nutrients, acids, organic materials, sediment (suspended and dissolved solids), and sulfates.

Petroleum - Fugitive air emission and air point source emission fallout, leaching from waste ultimate disposal sites, process spills and leaks are the most probable nonpoint sources. These sources contribute organic materials (including oil and grease), pesticides, sediment (suspended and dissolved solids), sulfates, nutrients, and heat.

Timber Products - Erosion from roadways, and timber harvesting fugitive air emission fallout from cutting and transportation and fertilization are potential nonpoint sources. The pollutants emitted include organic materials, acids, pesticides, sediment (suspended and dissolved solids), and heat.

Pulp & Paper - Nonpoint sources are runoff from coal storage piles, log piles, chip piles and bark disposal piles. Sources also include fugitive and point source air emission fallout and erosion from roadways. Organic materials, acids, sediment (suspended and dissolved solids), and sulfates are the most probable contaminants.

Inorganic Chemicals - Runoff from raw material and intermediate storage piles, leaching and runoff from ultimate disposal areas of concentrated wastes and accumulated materials from spills, and fugitive and points source air emission fallout are potential nonpoint sources. Pollutants include sediment (dissolved and suspended solids), organic materials, metals, sulfates, acids and heat.

Fertilizer - Nonpoint sources include runoff from storage piles, accumulated materials from spills and leaks and fallout from fugitive and point source air emissions. Organic materials, nutrients and sediment (suspended and dissolved solids) are the primary pollutants.

Cement - Runoff from storage piles and fugitive and point source fallout are the most probable nonpoint sources. Sediment (suspended and dissolved solids) is the primary contaminant.

Feedlots - Runoff from accumulated materials (waste and feed) is the major nonpoint source. Pollutants include organic materials, nutrients, and sediment (suspended and dissolved solids).

Phosphate Manufacturing - Nonpoint sources are runoff from material piles and fallout from fugitive and point source air emissions. Sediment (suspended and dissolved solids), nutrients and organic materials are principal pollutants.

Nonferrous Metals - Runoff from storage piles and accumulated materials from process water spills, and fallout from fugitive and point source air emissions are most probable nonpoint sources. Contaminants include metals, nutrients and sediment (suspended and dissolved solids).

While regulations now exist for controlling some of these sources, as stated earlier, little has been done in trying to isolate and quantify industrial nonpoint sources. Before regulations imposing controls are to be enforced, there is a need for defining the potential problem. Therefore, there is a need for collecting field data. We would like to discuss the major elements of a field program for sampling industrial nonpoint sources.

Measurement Methodology

As stated earlier in the definition of nonpoint sources, they are not easily monitored at their source. Most of the nonpoint work performed (urban and agricultural), has focused on monitoring changes in the receiving water body. Most of these studies have tested large drainage basins and usually no effort has been made to isolate individual sources. The receiving waters were monitored upstream and downstream of the test areas in both dry and wet conditions. The difference in parameters minus any point source contribution was the nonpoint source loading.

Since industries are usually in urban areas or within industrial parks, it is much more difficult to isolate a particular industry. Sampling the receiving water body input and output and assuming the difference, minus point source, is due only to the particular industry's nonpoint sources could result in gross errors. Therefore, some quantification and qualification of the industrial nonpoint sources must be performed in parallel with receiving body monitoring.

Since nonpoint sources are site specific, in order to design a field sampling program, background information about the test area must be obtained including topography, geology, hydrology, climatology and land use within and around the test area. This information can then be used to locate sampling stations for both runoff and receiving water body sampling. A review of the test industry's operating procedures will also be helpful in developing the test plan.

In designing the test program it will be important to select parameters to be monitored which are associated with the test industry. Table 1 shows some of the common parameters monitored. In addition, it is sometimes helpful to choose a particular organic, such as phenol, as an indicator chemical. The choice of a sampling technique, either manual or automatic, and the type of sample, either composite or discrete, are two more considerations in developing a test plan. Manual sampling has high operating costs but can be an advantage over the high capital cost of automatic sampling if inexpensive labor is available. However, since storm events are intermittent, can occur any time of day and night and can be of short duration, automatic sampling has the overall

TABLE 1

PARAMETERS COMMONLY MONITORED IN NONPOINT SOURCE PROGRAMS

Suspended Solids
Dissolved Solids
Turbidity
Dissolved Oxygen (DO)
pH
Total Organic Carbon (TOC)
Biochemical Oxygen Demand (BOD)
Chemical Oxygen Demand (COD)
Oil & Grease
Metals
Phosphate (Ortho, para)
Total Kjeldahl Nitrogen (TKN)
Ammonia Nitrogen
Sulfates
Pesticides

advantage.

A discrete sample is collected over a certain time interval and is kept separate from other samples. This type of sample allows comparison of runoff water quality over a period of time. The problem with discrete samples is the large number of samples to be collected and analyzed. A composite sample consists of a series of smaller samples which are consolidated to form a larger sample. A composite sample reduces the number of samples to be analyzed, but it does not allow comparison of runoff from specific time intervals. A test program should incorporate both types of samples, discrete samples being used on the parameters of major interest and composite samples on the parameters of minor interest, depending on the objectives of the test program. If interest is in a single storm event then discrete sampling would be required for adequate definition. If interest is in long-term or average storm conditions then composite sampling is adequate.

Samples should be taken in both dry and wet weather conditions. The sampling time is critical during storm events. Because of the "first flush" effect of storms, sampling must begin at the first instance of rainfall and discrete samples should be taken at intervals of 5-10 minutes. Composite samples should be taken at intervals of 15-30 minutes. An automatic sampling system which is electronically signalled to start by increases in water level in a rain gauge is a must if the "first flush" is to be sampled. Figure 3⁽²⁾ is a flow chart of such a system.

Figure 4 shows a sketch of a hypothetical test area located along a river. Once a test area has been selected, upstream and downstream river stations are located during dry weather conditions for sampling and flow measurements. These stations are sampled at various depths and distances across the river during both dry and wet weather conditions. If desired, intermediate river stations can be set up to isolate individual nonpoint sources. In addition, if tributaries flow into the river within the test area, they too must be sampled. River sampling techniques such as those used by Whipple³, DiGiano⁴, Randall⁵ and Colston⁶ have been used for several years.

During storm events, meteorological data including rainfall intensity and duration must be collected.

Topographic plots are developed to divide the test area into drainage basins. (See Figure 4.) Runoff sampling stations are located based on these plots. The major problem with this type of test program is quantification and qualification of runoff. Since we know runoff is diffuse, it is impossible to collect all runoff; therefore total quantification will have to be estimated from representative samples. Systems for collecting these samples are only in an early state of development. In any event a test program of this proportion is costly and since nonpoint sources are site specific and non-repetitive, it is conceivable that it would have to be carried out on several sites and several storm events for a particular industry. Since this is impractical from cost and time standpoints, the use of mathematical models for prediction of nonpoint source pollution as a replacement of a majority of the sampling could be more efficient.

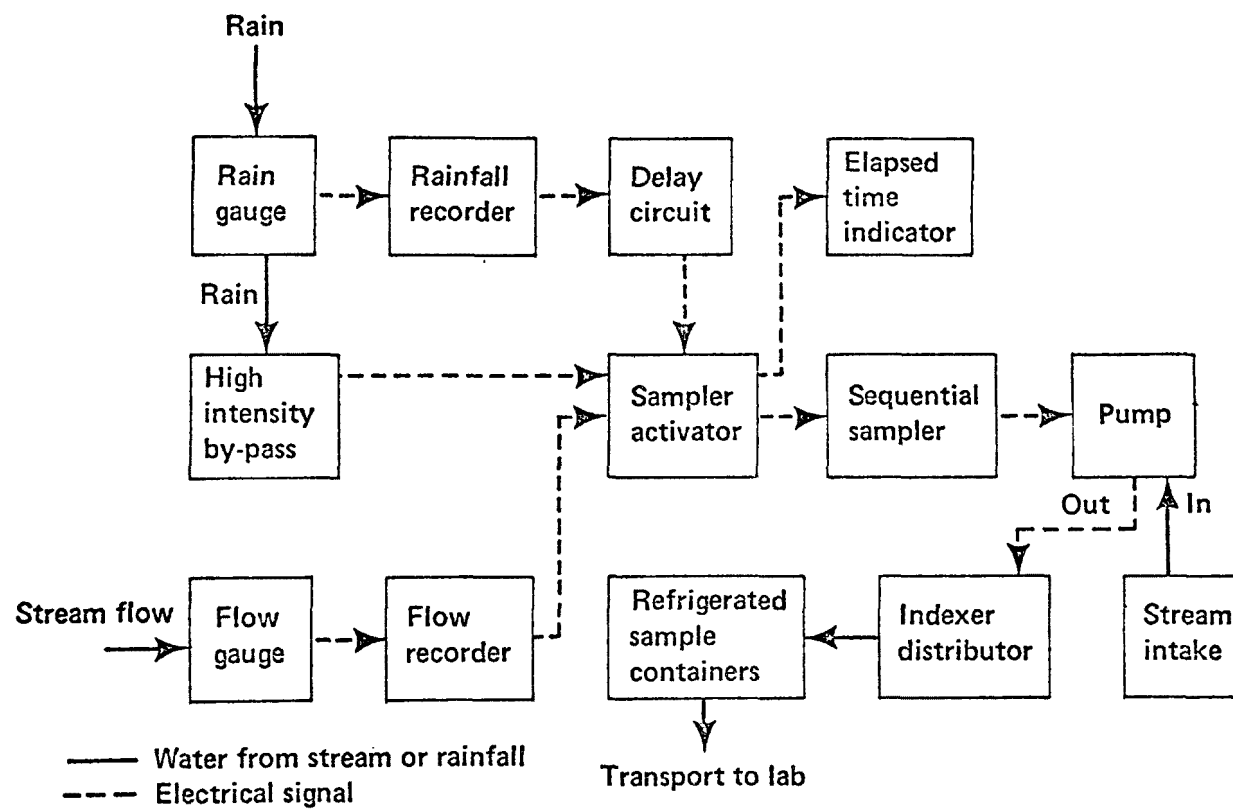


Fig. 3. Flow chart - automatic receiving water sampling during storms.

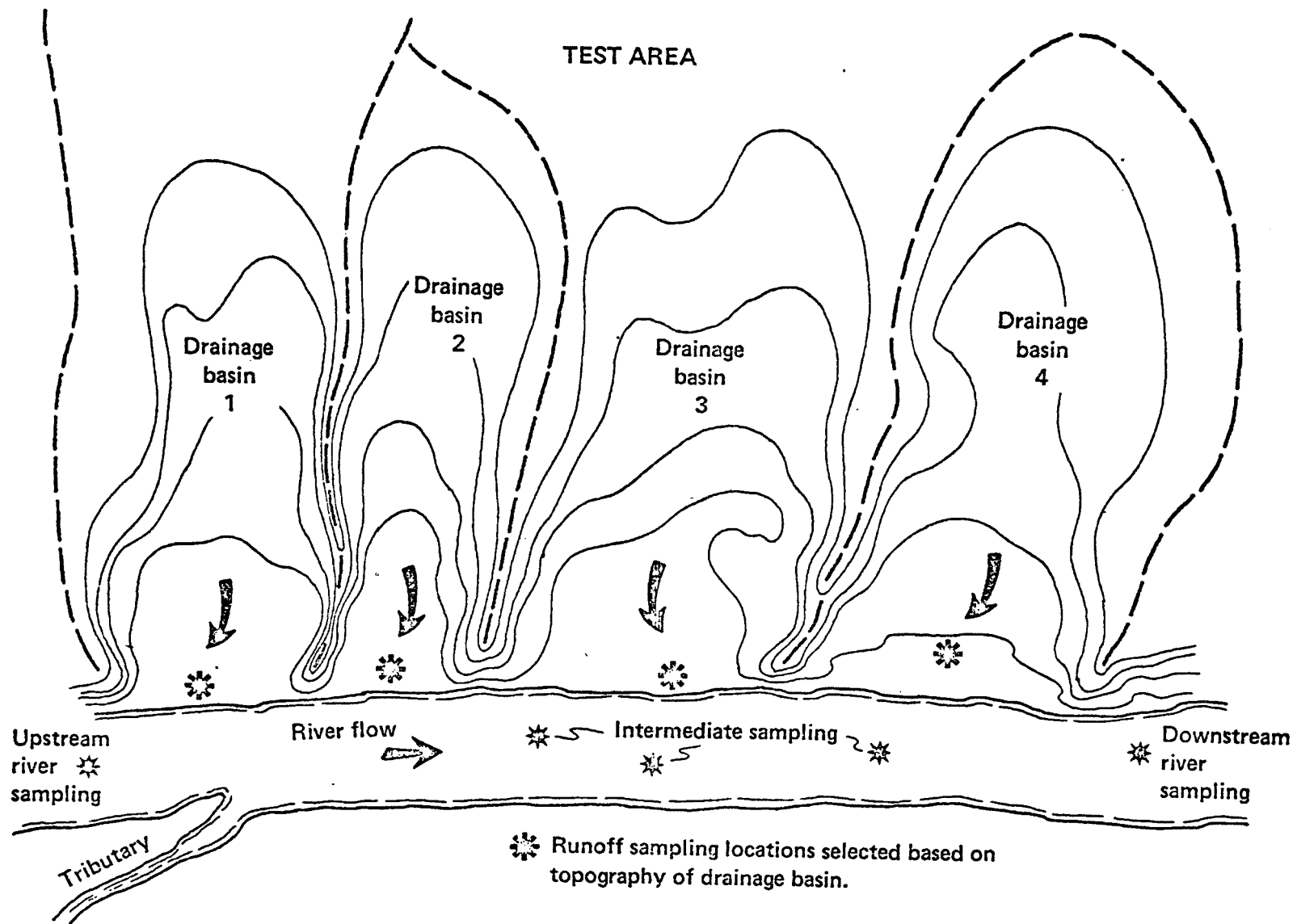


Fig. 4. Field program

Prediction Methodology

Mathematical models properly applied provide a cost effective means of quantifying impacts on water quality resulting from nonpoint source pollution and of evaluating alternatives for the control of nonpoint sources of pollution. In recent years many mathematical models have been developed to simulate the quantity and quality of stormwater runoff and the impact of such runoff on the quality of natural water bodies. Each model, however, was developed to satisfy a different need ranging from the design of municipal storm sewer systems to the assessment of land use as it influences flooding and water quality. There does not yet exist a model developed specifically for industrial runoff (except mining) although some models can be adapted. There are many criteria that can be used when selecting a model. In general, the simplest model which satisfies the project needs should be selected for use since such a model is normally the most economical choice. Figure 5⁽⁷⁾ serves to illustrate one aspect which contributes to model complexity - the choice of parameters to be modeled. For instance, in a relative sense, it is more difficult to model toxicity relationships than to model dissolved oxygen levels.

Once a model has been selected it must be adapted to the specific site or area being studied. A model is so adapted through the processes of calibration and verification. Calibration is achieved by adjusting the model to reflect site specific field data. After the model has been calibrated, it should be tested against a second set of field data. If the second set of field data and the modeled results compare favorably, the model is considered to be verified and ready for application.

For a model to be adaptable to industrial applications it must be capable of predicting the quantity and quality of stormwater runoff, the transport of such runoff to a receiving body of water, and the impact of such runoff on the quantity and quality of the receiving water. Pollutants of primary importance for model simulation include sediment (suspended and dissolved solids), nutrients (compounds of nitrogen and phosphorus), pesticides, acidity (pH and sulfuric acid), organic material (biochemical oxygen demand, chemical oxygen demand, dissolved oxygen), and heat (temperature). In addition, since storm events are dynamic, a model must also be capable of simulating functions in a dynamic, i.e., time dependent fashion.

To predict the quantity and quality of stormwater runoff a model must be capable of simulating the effects of such items as the intensity and the duration of the storm event, infiltration and drainage characteristics, the accumulation of pollutants between storms, and the washoff of such pollutants during storms. For continuous simulation of multiple storms, a model must be capable of simulating dry weather flows as well as storm flows.

To predict the transport of stormwater runoff for industrial land use, a model must be capable of simulating overland flow and routing in man-made systems (channels, sewers, etc.).

To describe the impact of the stormwater runoff on a receiving body of water, a model must be capable of simulating the quantity and quality responses of the receiving water to the runoff impact. Again, for continuous simulation of multiple storms, a model must be capable of simulating dry weather flows as well as storm flows. For increased flexibility a model should be capable of

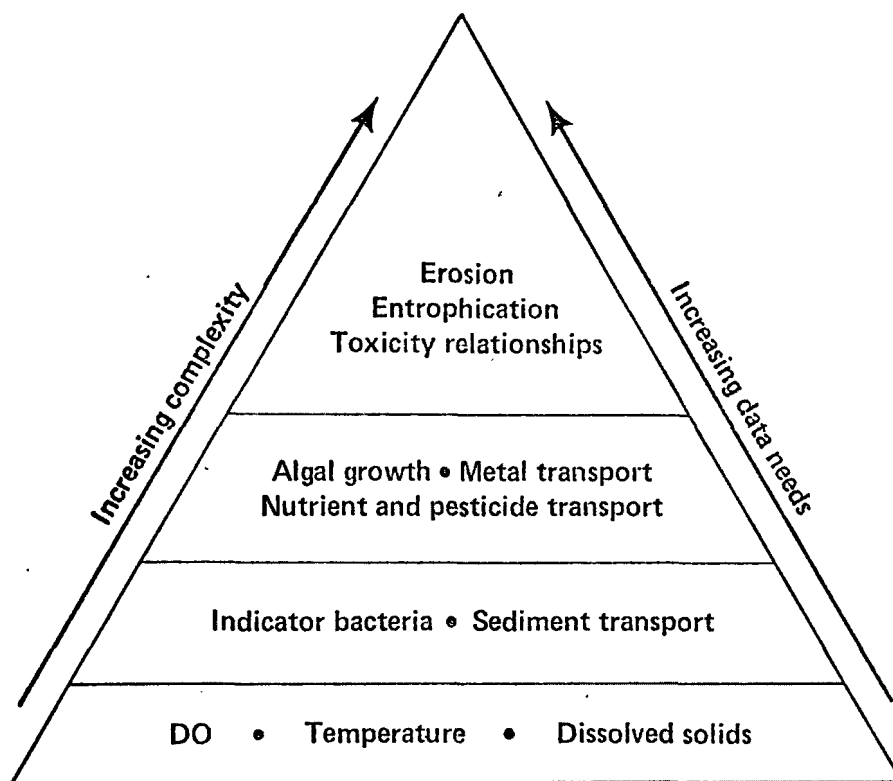


Fig. 5. Relative difficulty of applied modeling.

simulating various types of receiving waters including rivers, lakes and estuaries.

We have reviewed the prominent runoff mathematical models and those which can be adapted to industrial sites are listed in Table 2. The models were evaluated for suitability, adaptability, complexity, cost and availability for predicting the impact of industrial nonpoint sources on receiving body water quality using the following criteria:

Wastewater (Runoff) - quantity, quality, dry weather flows, storm runoff;

Receiving Water - quantity, quality, river, lake, estuary;

Quality Parameters - temperature, suspended solids, total dissolved solids, biochemical oxygen demand (BOD₅), chemical oxygen demand (COD), dissolved oxygen, nitrogen, phosphorus, pH, oil and grease, pesticides;

Simulation of Single Storm

Simulation of Multiple Storms

Computer Program Availability - Public or proprietary;

Complexity - high, moderate, low;

Costs - high, moderate, low.

The last four models listed in Table 2 were eliminated because they can not presently model the receiving water although STORM is currently being adapted to include the receiving water. The WRE, HSP, Dorsch Consult, SWMM - Release II and SSWMM - Receiv II models seem best suited for adapting to industrial applications. However, WRE, HSP and Dorsch Consult models are proprietary. Table 3 shows an evaluation of SWMM - Release II and SSWMM - Receiv II models.

Conclusions and Recommendations

As a result of this overview evaluation of nonpoint source water pollution from industrial activities, we have found the following:

1. Little or no quantification data for any of the 12 industries studied except acid mine drainage.
2. Nonpoint source measurement has been performed for urban and agricultural activities with the monitoring effort centered on the receiving water body.
3. Very little quantification and qualification of runoff has been performed.
4. Several mathematical models are available for predicting impact of runoff on receiving water bodies but none have been applied to industrial activities except mining. However, there are models which can be adapted to industrial activities.

TABLE 2

MATHEMATICAL MODELS ADAPTABLE TO INDUSTRIAL SITES

EPA Stormwater Management Model - Release II (SWMM)
Water Resources Engineers Stormwater Management Model (WRE)
Short Stormwater Management Model - Receiv II (Short SWMM)
Hydrocomp Simulation Program (HSP)
Dorsch Consult Hydrograph Volume Method
Corps of Engineers Storage, Treatment, Overflow and
Runoff Model (STORM)
Battelle Wastewater Management Model
Metcalf & Eddy Simplified Stormwater Management Model
Pyritic Systems: A Mathematical Model

TABLE 3

MODEL COMPARISON

	EPA SWMM Release II	Short SWMM Receiv II
<u>Wastewater</u>		
Quantity	X	X
Quality	X	X
Dry Weather Flow	X	X
Storm Runoff	X	X
<u>Receiving Water</u>		
Quantity	X	X
Quality	X	X
River	X	X
Lake	X	X
Estuary	X	X
<u>Quality Parameters</u>		
Temperature		
Suspended Solids	X	X
Total Dissolved Solids		
BODs	X	X
COD	W	
Dissolved Oxygen	X	X
Nitrogen	W	X
Phosphorus	W	X
pH		
Oil & Grease	W	
Pesticides		
Simulation of Single Storm	X	X
Simulation of Multiple Storms	X	X
Complexity	H	M
Cost	H	M

KEY: X = Yes
W = Wastewater Only
H = High
M = Moderate

Since many industries are faced with meeting regulations regarding non-point source water pollution control, there is a need for determining the extent of the problem so that cost-effective control measures can be taken. The program performed by TRC for the Industrial Environmental Research Laboratory of EPA will be extended to include:

1. The design and performance of a sampling program for runoff and receiving waters for a particular industry.
2. The adaptation of one of the SWMM models to be used in conjunction with the industry test program to model the impact of runoff from an industrial site on a receiving water body.

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16. ABSTRACT The proceedings are a compilation of technical papers presented at a symposium on the measurement and control of fugitive emissions (or non-point sources). They discuss techniques which have been used to measure fugitive emissions, as well as systems which have been used to control the emissions.		
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