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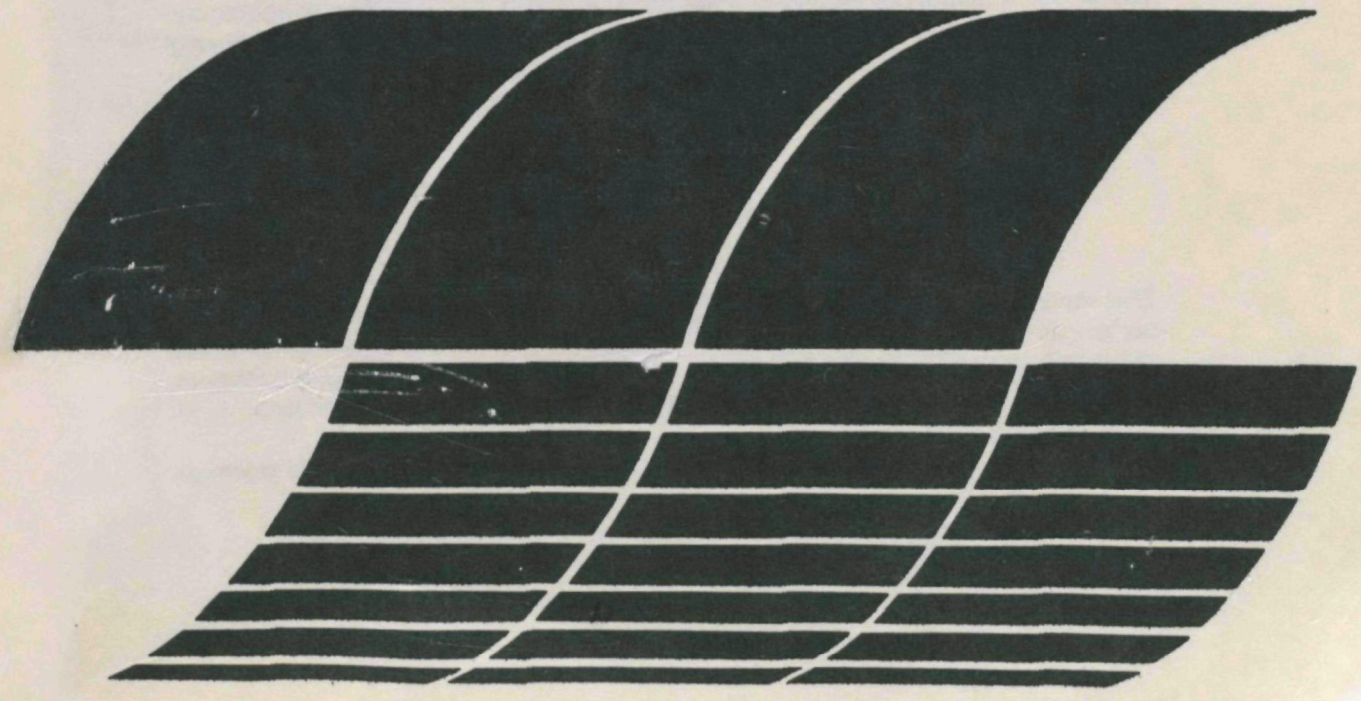
Third Symposium on Fugitive Emissions Measurement and Control (October 1978, San Francisco, CA)

Interagency Energy/Environment R&D Program Report



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Prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Research and Development
Washington, DC 20460

FOREWORD

The technical papers included in this volume were prepared for presentation at the "Third Symposium on Fugitive Emissions: Measurement and Control," held in San Francisco, California on October 23-25, 1978.

The objective of the Symposium was to provide a forum for the exchange of information among concerned representatives of industrial, research and governmental organizations relative to recent developments in industrial and energy-related fugitive emissions measurement and control. The Symposium was sponsored by the Environmental Protection Agency's Industrial Environmental Research Laboratory at Research Triangle Park, North Carolina as part of its continuing effort to develop methods for the measurement and control of airborne and waterborne fugitive emissions.

D. Bruce Harris of the Industrial Environmental Research Laboratory was the Project Officer and General Chairman of the Symposium.

Joanne King of TRC - The Research Corporation of New England was the Symposium Coordinator and Compiler of these Proceedings.

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Sanitary Engineer
EPA/IERL-RTP

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Research Chemical Engineer
EPA/IERL-RTP

OVERVIEW OF THE FUGITIVE EMISSION PROBLEM - 1979 SIP REVISIONS

By

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U. S. ENVIRONMENTAL PROTECTION AGENCY

Presented at

Third Symposium on Fugitive Emissions Measurement and Control

October 23 - 25, 1978

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ABSTRACT

Fugitive emissions and fugitive dust and their associated impacts will be a major consideration in the development of the 1979 State Implementation Plans for those areas which are currently not attaining the National Ambient Air Quality Standards. Fugitive emissions and fugitive dust will also play an important role in obtaining new source permits both under PSD and new source review in nonattainment areas. Specific guidance will be presented on how fugitive emission/dust control will be factored into the State plans for existing, as well as new, sources considering the possible revision to the particulate matter standard.

This paper summarizes some of the more important requirements of the new Clean Air Act of 1977, especially those which relate to the fugitive particulate matter problem--provisions that are already affecting, or likely to affect, everyone in the near future whether they represent a control agency, industry, or the general public.

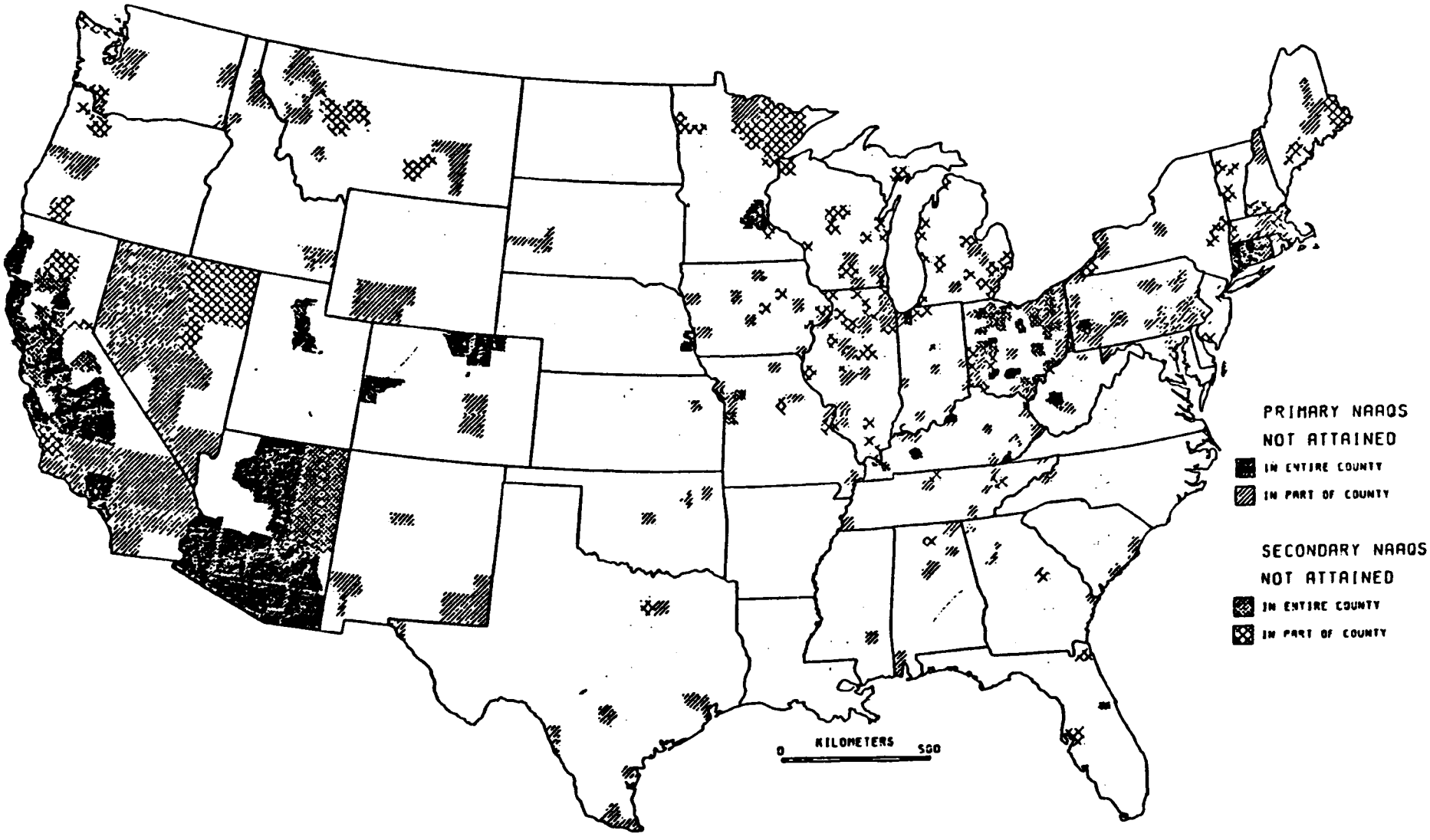
The past seven years have served as a major testing ground for the fundamental goals of the Act. Seven years after passage of the Act, violations of the National Ambient Air Quality Standards for one or more pollutants still exist in more than half of the 247 air quality control regions. As a result of the recent efforts required by Section 107 of the Act, to designate areas as meeting, not meeting, or unclassified with respect to the NAAQS, it was found that of the 3,215 counties in the United States 408 are listed as nonattainment for TSP. (Figure 1)

These standards were to have been attained by 1975; however, substantial progress has been made. For example, the estimated number of people exposed to total suspended particulates in excess of the annual primary standard decreased from 74 million in 1970 to less than 50 million in 1975; in other words, the exposed population has been reduced by more than one-third since the passage of the Clean Air Act of 1970.

Despite the progress that has been made in reducing nationwide emissions of air pollutants, failure to achieve the National Ambient Air Quality Standards in many areas of the country represents the seven-year reality of the implementation of the 1970 Act. The Congress began to address the reality of this situation more than three years ago and initiated the drafting of the first comprehensive amendments to the Act since 1970. During the course of these intense deliberations in Congress, the Agency merely tried to maintain an even keel while we were forced to balance the realities of the non-attainment situation against the demands of the nation's emerging energy problems.

Finally, with the recent enactment of the Clean Air Act Amendments of 1977, the Congress has reaffirmed its commitment to the goals originally established by the 1970 legislation. The new Amendments provide a strong mandate and make clear that Congress does not intend that the nation's energy problem be allowed to compromise environmental quality. The Act strongly reaffirms our goals of attainment and the prevention of significant air quality deterioration.

Figure 1.



PROMULGATED LIST OF TSP NON-ATTAINING COUNTIES - 1978

To a large extent, the hardest phase of our efforts to meet ambient air quality standards is yet to come. The previously mentioned progress has been achieved through the application of controls to sources generally amenable to control requirements. During this next round of control efforts, agencies will be forced to concentrate on the less conventional sources (fugitive dust, fugitive emissions) with which we have far less control experience. In other words, we have done the so-called easy things, and what is left are the more difficult, less tried methods of air pollution control. This is particularly true with respect to the attainment of the TSP standard, as it must be attained by 1982. Conventional stationary sources have generally been controlled; however, in many cases, fugitive emissions constitute a large percentage of total emissions. A recent EPA study on National Assessment of TSP[1] indicates fugitive emissions add 15 to 25 $\mu g/m^3$ to citywide average TSP levels. However, significantly greater impacts in vicinity of industrial sources were noted. Additionally, as part of this study, the contractor visited a number of industrial sites within 14 cities. Of all industrial sites, 32% were greatly impacted by fugitive emissions. Of industrial sites with annual means twice national standard, all were influenced by fugitive emissions. To some extent, the new Act recognizes these difficulties and provides EPA with tools which will enable the Agency to establish new regulatory requirements and new approaches to difficult problems but also provides the flexibility to resolve isolated conflicts between environmental goals and energy, economic, and employment concerns.

The heart of the new Amendments focuses on the problem of attainment in areas where the National Ambient Air Quality Standards continue to be violated. Each State that includes a nonattainment area must submit a plan by January, 1979, for EPA approval by July, 1979, which provides for attainment of the standards as expeditiously as practicable. Congress took a firm position on the issue of attainment. The new State plan requirements will necessitate in most cases the application of reasonable control measures and strict enforcement of these requirements for existing sources. The plan will have to provide sufficient reductions in emissions to allow room for growth, otherwise construction permits for new sources can be granted only when more than offsetting emission reductions are secured on a case-by-case basis prior to the facility start-up date.

Control programs must be developed for each area which has been designated as nonattainment. These control programs must indicate how much control will be needed.

In developing each control program, the control agencies will be considering various alternatives for attaining the standards. Once these control alternatives have been considered, they should be screened to determine what limitations may exist regarding each alternative. However, it should be noted that in most cases, because of the magnitude of the problem, it will take all that one can think of to attain the standards.

Emission reductions for each alternative should be developed to determine the effectiveness of each program. For photochemical oxidants, EPA has developed control techniques guideline (CTG) documents to provide some information on the percent of reduction, etc., available for various hydrocarbon control measures. However, no specific CTG's have been developed for TSP. The Agency, however, has published various documents listed in Table 1 which will be useful in determining the percent control for various particulate matter control measures. However, the most important factor to consider in developing a control strategy is that it be technically feasible.

The given strategy must provide for both attainment of the primary and secondard standards for TSP. The primary standard must be attained as expeditiously as practicable but no later than 1982 and the secondary standard must also be met by 1982. However, if more than RACT is needed to attain the secondary standard, a reasonable time is permitted.

Once the strategy has been selected and it provides for both attainment and maintenance of the NAAQS, the strategy must be adopted in the form of legally enforceable procedures and submitted as a revision to the State Implementation Plan with the Governor's approval. This constitutes a commitment on the part of the State to implement and enforce the plan.

Where adoption by 1979 is not possible (nontraditional controls for particulate matter), a schedule for expeditious development, adoption, submittal, and implementation of these revisions would be acceptable. These schedules must provide for implementation as expeditiously as practicable. Prior to attainment, these measures must be implemented rapidly enough to provide for emission reductions necessary to maintain reasonable further progress as required by the action. Schedules would be a part of the applicable SIP and would represent a commitment on the part of the State to meet the key milestones set forth in the schedule.

Each plan must provide estimates of emission reductions for each adopted or scheduled control measure or for related groups of measures where estimates of individual measures are impractical. The Agency recognizes that estimates may change as measures are more fully analyzed and implemented. As estimates change, these should be revised and included in subsequent plan submissions to ensure the plan remains adequate.

Thus, the 1979 SIP submission for TSP must contain a demonstration of attainment of the primary standard as expeditiously as practicable but no later than 1982, and emission limitations or regulations for the control of traditional, as well as industrial, process fugitive particulate emissions. The plan may also contain schedules for the development, adoption, and submittal of controls of nontraditional sources. These may include demonstration studies, etc., prior to adoption for many nontraditional measures. More information on demonstration studies can be found in "Guidelines for Development of Control Strategy in Areas with Fugitive Dust Problems," OAQPS 1.2-071, pp 6-1 to 6-10.

TABLE 1.

INFORMATION FOR DETERMINING
PERCENT CONTROL FOR TSP

TSP

-- CONTROLLED AND UNCONTROLLED EMISSION RATES AND APPLICABLE LIMITATIONS FOR EIGHTY PROCESSES, SEPTEMBER 1976.

- EXTERNAL COMBUSTION
- SOLID WASTE DISPOSAL
- FOOD AND AGRICULTURE
- METALLURGICAL
- MINERAL PRODUCTS
- WOOD PROCESSING

-- GUIDELINE FOR DEVELOPMENT OF CONTROL STRATEGIES IN AREAS WITH FUGITIVE DUST PROBLEMS, OAQPS #1.2-071, OCTOBER 1977

- UNPAVED ROADS
- ENTRAINED STREET DUST
- CONSTRUCTION AND DEMOLITION
- AGRICULTURE
- TAILING PILES
- PARKING LOTS

-- TECHNICAL GUIDANCE FOR CONTROL OF INDUSTRIAL PROCESS FUGITIVE PARTICULATE EMISSIONS, MARCH 1977, EPA 450/3-77-010

- COMMON DUST SOURCES
- IRON AND STEEL PRODUCTION
- PRIMARY NON-FERROUS SMELTING
- SECONDARY NON-FERROUS SMELTING
- FOUNDRIES
- MATERIALS EXTRACTION AND BENEFICIATION
- GRAIN ELEVATORS
- PORTLAND CEMENT
- LIME
- CONCRETE BATCHING
- ASPHALT CONCRETE PRODUCTION
- LUMBER AND FURNITURE

Impact of Potential Revision to Particulate Matter NAAQS

The Clean Air Act and EPA policy require a periodic reevaluation of each NAAQS. Under requirements, a program has begun to revise the air quality criteria for particulate matter. It is anticipated that a draft revised document will be available for external review in late 1979. Any revisions to the NAAQS for particulate matter that result from revision to the criteria document would probably be proposed in mid-1980 and promulgated in late 1980.

Contrary to the position taken by some, the Agency contends that the current TSP standards are reasonable in light of available evidence. The standards have been under essentially constant review over the past several years, and the Agency believes this position is widely supported by the scientific community. This is not to suggest that the TSP standards could not be improved upon. Many reviews of the standards have expressed concern for the use of TSP as an index to health effects and indicate a need for better characterization of particulate matter by size and chemical composition. The Agency shares the view that any revision of the particulate matter criteria document should consider the possibility of defining a more precise index of particulate matter pollution.

It is possible that the revised criteria document will result in some form of an "inhalable" particulate matter primary standard based on particle sizes of less than 15 micrometers.

Of course, until the criteria document is revised, any estimate of the effect is only speculation and therefore should not be allowed to disrupt the current efforts to develop SIP revisions in areas designated nonattainment for particulate matter. The SIP revisions must still be submitted by January 1, 1979, must demonstrate attainment of the current particulate matter standard by December 31, 1982, and must include emission regulations for conventional sources and programs to subsequently develop controls for unconventional sources as required.

If the reevaluation of the criteria document results in the kind of change to the NAAQS discussed above, however, some controls that would be necessary to attain the current NAAQS may not be necessary to attain a revised primary NAAQS. Generally, the sources in this category are those such as storage piles and materials handling operations that emit relatively large particles.

The 1979 SIP revisions must contain or provide for the development of all measures necessary to attain the current particulate matter standard. However, it is legally permissible for States which adopt new regulations covering sources of predominantly large particles to recognize the possibility of a revision of the current standard in establishing compliance schedules for such sources. As long as compliance is required not later than 1982, such schedules may include dates which are late enough so that the uncertainty over the particulate matter standard can be resolved prior to significant expenditures for control.

The possible revision of the particulate standard should not be a factor which is considered in setting compliance schedule for sources other than those described above. Nor can this possibility affect any existing compliance schedules.

New Source Review and Fugitive Dust

The Agency has set forth a position regarding the impact of industrial process fugitive emissions and fugitive dust emissions on new source review in general in the August 16, 1978, Fugitive Dust Policy, and for PSD in particular in the June 19, 1978, final Regulations for the Prevention of Significant Deterioration.

Briefly, the Fugitive Dust Policy indicates that for new source review in nonattainment urban areas, the current program which includes the emission offset concept remains unchanged. However, since fugitive dust is recognized as a significant air pollution problem in urban areas, it is appropriate to allow sources to minimize either existing fugitive dust sources or particulate stack emissions in order to satisfy their emission requirements.

There has been considerable concern about the location of major new stationary sources in rural areas where fugitive dust has been determined to be the major source. New sources that wish to construct in rural areas with infrequent short-term violations of the TSP standard should be allowed to construct without the need of an emission offset, as long as they comply with the appropriate emission regulation (NSPS, state regulation or BACT for PSD source) and when considering their emissions, plus "non-urban" background and the emissions from other stationary sources in the vicinity of the proposed location, they do not cause violations of the NAAQS or appropriate PSD increments.

The June 19, 1978, regulations for PSD set forth a two-tier review system. Under the second tier, a source must apply best available control technology. In submitting a permit application with regard to BACT, the source must propose BACT, set forth alternative systems, and then defend the BACT selected. BACT can include several technology options. It must cover all emission points, including stack emissions, fugitive process and fugitive dust emissions. It must be at least as stringent as NSPS, NESHAP, and can include design, equipment or operating standards.

Additionally, under the second-tier review, the source must conduct an ambient impact review against both the NAAQS and increments. However, the June 19, 1978, regulations for PSD contained several exemptions regarding ambient impact review. One of those exemptions relates to fugitive dust. Several comments were received regarding the proposed PSD regulations as they related to fugitive dust emissions. As a result of the comments, the Agency will exclude from any air quality impact assessment of a source or modification any fugitive dust emissions. Additional support for this exclusion can be found in the legislative listing. It points to the utilization of "administrative good sense" regarding the treatment of fugitive dust.

Certain aspects of this exclusion for fugitive dust should be noted. First the burden of showing to what extent emissions from the proposed source or modification would be made up of fugitive dust rests with the applicant. Second, the regulations do not exclude fugitive dust from the determination of potential emissions. Any source or modification which, taking into account emissions of fugitive dust, would have potential emissions equal to or greater than 250 tons per year would be subject to the applicable PSD requirements, especially in many instances the BACT requirement. Finally, EPA will treat emissions of fugitive dust as not consuming increment for the purpose of evaluating other sources under PSD.

It should be emphasized that EPA intends to implement the above policy of excluding the fugitive dust only on an interim basis. EPA will reassess the implications of the policy and any possible technical improvements in modeling fugitive dust, and will adjust the policy as appropriate.

The Agency realizes the difficulty of the task that lies ahead to develop adequate SIPs by January, 1979, and to fully implement those SIPs by 1982. The Agency, in developing the requirements for an acceptable SIP, has tried to develop reasonable and achievable goals that still meet the intent of Congress. We have tried to develop requirements that overcome barriers and make it as easy as possible to develop SIPs that will meet the intent of Congress. Between now and January many tough decisions are going to have to be made.

The challenge we are faced with is to achieve the cleanest air possible while maintaining the high standard of living the citizens of America have attained. Fugitive emissions control must begin now--we are beginning to more fully appreciate the problem, and fugitive emissions control must be considered in the current round of SIP revisions. If not, we are only dealing with part of the air pollution control program, and we will fall short of our goal of attaining the NAAQS as expeditiously as practicable.

REFERENCES

1. National Assessment of the Urban Particulate Matter Problem, EPA 450/3-76-025, July, 1976.

REGULATION OF FUGITIVE EMISSIONS

UNDER THE CLEAN AIR ACT'S

PREVENTION OF SIGNIFICANT DETERIORATION

(PSD) AND NONATTAINMENT REQUIREMENTS

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ABSTRACT

The Clean Air Act Amendments of 1977 have forced EPA and the States to come to grips with the fugitive emissions problem. Clearly, two of the most significant forcing functions are the Act's new requirements for preventing significant deterioration (PSD) and the new provisions relating to nonattainment areas. This paper focuses upon the control strategies for fugitive dust and fugitive hydrocarbon emissions which EPA is using to implement the PSD and nonattainment requirements of the Act for total suspended particulate and photochemical oxidants/ozone. These strategies focus upon two basic areas:

1) Control of new fugitive emission sources under EPA's PSD preconstruction review requirements and/or the provisions of EPA's offset policy.

2) Control of new and existing fugitive emission sources under revised State implementation plans called for by the Act's PSD and nonattainment provisions.

A separate issue concerns the extent to which ambient particulate concentrations attributable to fugitive dust should be considered in the ambient air quality analyses required under the PSD and nonattainment procedures for proposed new and modified stationary sources. In areas subject to PSD requirements, the principal difficulty lies in distinguishing between natural and man-induced fugitive dust. In nonattainment areas, distinguishing between rural and urban fugitive dust problems is a major problem issue.

This paper explores the pertinent statutory and regulatory programs under which EPA is attempting to deal with these problems.

1.0 INTRODUCTION

Of the many regulatory consequences of the 1977 Clean Air Act Amendments, one of the most significant is the increased attention they have generated on the fugitive emissions problem. Historically, EPA's implementation of the Clean Air Act has focused on emissions from new automobiles and industrial installations. With respect to non-mobile sources of particulate matter, regulations have been directed primarily at limiting the opacity and mass loading of smokestack emissions from incinerators, fossil fuel-fired boilers and major process industries. Non-stack particulate controls have traditionally been applied only to open burning and mineral extraction operations.

Recent studies indicate that traditional stack-oriented control strategies for particulate matter are insufficient by themselves to achieve the ambitious air quality goals of the Clean Air Act. Control of industrial fugitive emissions and nontraditional sources of fugitive dust will therefore be an essential ingredient in future control strategies for particulate matter. Control of new, and possibly existing, fugitive dust sources will also be required in order to comply with EPA's new regulations on Prevention of Significant Deterioration (PSD). This paper attempts to summarize the federal regulations and policies applicable to fugitive dust sources under the recently amended Clean Air Act. Pertinent provisions of EPA's regulations and guidelines for nonattainment areas and PSD areas will be discussed within the context of the EPA Fugitive Dust Policy.

OVERVIEW OF PSD AND NONATTAINMENT - SPECIAL RULES FOR FUGITIVE DUST

2.1 National Ambient Standards

Any discussion of PSD and nonattainment should be preceded by an overview of the fundamental statutory framework of the Clean Air Act for achieving and maintaining the National Ambient Air Quality Standards (NAAQS). The primary purpose of the Act is to achieve and maintain air quality levels which are protective of public health and welfare. To this end, EPA is required to establish National Ambient Air Quality Standards (NAAQS) for selected pollutants emitted from diverse sources which could endanger public health or welfare (criteria pollutants).[1] (See Table I.) The ambient standards are of two types. "Primary" standards are established at levels which will protect public health while "secondary" standards are designed to protect public welfare. As of October, 1978, EPA has promulgated NAAQS for six of the most ubiquitous air pollutants including particulate matter (PM); sulfur oxides (SO_x); nitrogen oxides (NO_x); carbon monoxide (CO); photochemical oxidants (measured as ozone); and non-methane hydrocarbons (HC)*.[2] An ambient standard for lead was established in the Fall of 1978.[3] It is the particulate matter NAAQS which forms the statutory basis of fugitive dust control requirements under the Clean Air Act.

Attainment and maintenance of the ambient standards for the criteria pollutants is the primary responsibility of the States. Under Section 110 of the Clean Air Act, each State is required to submit a State Implementation Plan (SIP) for EPA's approval. These plans set forth the State's strategy for attaining and maintaining the standards within the time frames established by the Act. The required strategy must satisfy the eleven enumerated requirements contained in Section 110 of the Act, including programs for preventing significant deterioration of clean areas and ensuring timely clean-up of polluted areas. Under the 1970 Clean Air Act Amendments (the foundation of the current statute) all States were to have attained the primary (health-related) standards by May 31, 1975 with the exception of a relatively few areas where an extension to mid-1977 was granted. Secondary standards were to be attained within a "reasonable time," defined by most state plans to coincide with the primary standard attainment date.[4]

2.2 Nonattainment

Unfortunately, the 1970 Act did not specify the consequences of a state's failure to attain the primary standards by the statutory deadline. As the deadline approached, however, at least 160 of the nation's 247 air quality control regions (AQCRs) had monitored violations. Many urban areas failed to attain the particulate standards.

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*The hydrocarbon standard is designed only as a guide for assessing the adequacy of State plans in attaining the photochemical oxidant standard.

TABLE I

NATIONAL AMBIENT AIR QUALITY STANDARDS (NAAQS)
40 CFR 50

NATIONAL PRIMARY AND SECONDARY AMBIENT AIR QUALITY STANDARDS
(Expressed as micrograms per cubic meter [$\mu g/m^3$] at 25°C, 760 mm pressure)

POLLUTANT	PRIMARY STANDARD		SECONDARY STANDARD	
	Annual Mean	Maximum Concentration (Allowed Once Yearly)	Annual Mean	Maximum Concentration (Allowed Once Yearly)
Sulfur Oxides (SO_x) (measured as SO_2)	80	365 (over 24 hours)		1300 (over 3 hours)
Particulates	75	260 (over 24 hours)	60[1]	150 (over 24 hours)
Carbon Monoxide (CO)	--	10 milligrams/m^3 (over 8 hours) 40 milligrams/m^3 (over 1 hour)	Same as Primary Standard	
Photochemical Oxidants[2]	--	157 (over 1 hour)	Same as Primary Standard	
Hydrocarbons (HC)[3]	--	160 (over 3 hours-- 6-9 a.m.)	Same as Primary Standard	
Nitrogen Dioxide (NO_2)[4]	100	--	Same as Primary Standard	

1. The 60 mg/m^3 particulate standard is to be used only as a guide in assessing the adequacy of State plans in achieving the 24-hour standard.

2. EPA has proposed to relax the primary oxidant standard from 157 up to 196 mg/m^3 (i.e., from 0.08 to 0.10 ppm).

3. The hydrocarbon standard is to be used as a guide in assessing the adequacy of oxidant control plans

4. Pursuant to a mandate in the Clean Air Act, EPA is currently investigating the need for a short-term (i.e. 1-3 hour) NOx standard. EPA is currently considering a one-hour standard ranging from 470 to 940 mg/m^3. The agency is expected to make a final decision on this matter in 1979.

The obvious legal dilemma created by these widespread failures concerned the legal "approvability" of new sources which would add to levels of pollution already in violation of the law. Section 110 of the Act, which prescribed the July 1, 1975 attainment deadline, did not provide for the consequences of failure. EPA's regulations implemented this inflexible mandate by prohibiting the construction or modification of any facility which would interfere with the attainment or maintenance of a national ambient standard.[5] Thus, a strict reading of the law would have prohibited new sources from locating in any area which had failed to attain the ambient standard for the pollutant(s) it emitted. The practical effect of such an interpretation was an end to growth in the developed areas of the United States. EPA realized that the American public was unwilling to pay such a price, even for clean air.

EPA responded to this problem with promulgation of its national Offset Policy on December 21, 1976.*[6] The essence of the Offset Policy is that major new growth should be allowed in nonattainment areas only if air quality is improved as a result of that growth. This improvement comes by way of contemporaneous emission reductions from existing sources so as to more than "offset" the emissions which will be added by the proposed new source.

The Offset Policy imposes four substantial conditions on major new or modified sources, including major sources of fugitive dust, seeking permits to expand in and around nonattainment areas. First, the source must reduce its emissions to the lowest achievable emission rate (LAER). Second, it must be able to certify that all sources which it owns or controls in the same state are either im compliance or on a schedule of compliance with the State implementation plan. Third, the new source must obtain emission reductions from existing area sources which more than "offset" the pollution to be added by the new LAER-controlled source. These offsetting emission reductions must exceed the proposed new emissions by enough to represent "reasonable further progress" toward attainment of the ambient standards. The fourth and final condition, closely related to the third, requires the owner to demonstrate that the combination of LAER and offsets will lead to a net air quality benefit in the affected area.

Congress met the nonattainment issue head-on in the 1977 Clean Air Act Amendments by continuing the Offset Policy until mid-1979 and establishing a State implementation plan revision process to deal with the problem after that date. Specifically, Section 129 of the 1977 Amendments provided that EPA's Offset Ruling, as it may be amended, was to remain in effect, except that the baseline for determining emission offset credit was changed and the source applicability was expanded. By January 1, 1979, the States must submit plan revisions to EPA which conform to new Part D of the Act. These revised plans are designed to replace EPA's Offset Policy and must be approved by no later than June 30, 1979. Failure to adopt and receive EPA approval of the revised

*EPA is currently considering several significant changes to the policy. These changes are not, however, expected to revise the basic format of the current policy. Special rules will be proposed for fugitive dust sources, however, as discussed in Section 2.4 of this paper.

CHRONOLOGICAL DEVELOPMENT
OF EPA'S OFFSET POLICY
AND THE ACT'S NONATTAINMENT PROVISIONS

7/1/75: Original Deadline for Attaining the Primary NAAQS

Promulgation of EPA's Offset Policy
12/21/76

The 1977 Clean Air Act Amendments
8/7/77

EPA Administrator's Memorandum Detailing the Criteria for Approving SIP Revisions for Nonattainment Areas
2/24/78

EPA Promulgates List Designating PSD and Nonattainment Areas Pursuant to Section 107 of the Act
3/3/78

EPA Promulgates Major Revisions to Its Offset Ruling
12/78

Revised SIPs Due to EPA
1/1/79

Deadline for EPA Approval of the Revised SIP
(Failure to Obtain Approval by Deadline Triggers the Growth Ban Provision of Section 110(a)(2)(I).)
6/30/79

New Deadline for Attaining the Primary NAAQS
12/31/82

Extended NAAQS Deadline for Areas Unable to Attain the Oxidant and/or Carbon Monoxide Standards
12/31/87

plan by this deadline triggers a statutory ban on major new sources (of the nonattaining pollutant) in the nonattainment area.[7] Until the revised SIPs are approved and before July 1, 1979, the Offset Policy remains in force.

2.3 Prevention of Significant Deterioration

The Act's silence on the nonattainment problem was matched by an equal legislative vacuum on the issue of whether an area with air quality already better than the national standards could allow its air to be "degraded" to the national standards. While the objective of the nonattainment provision is to *achieve and maintain* the ambient standards, the purpose of PSD is to prevent significant deterioration of air already cleaner than the ambient standards.

The PSD controversy began as a result of a seemingly innocuous phrase in the original Act stating that one of the Act's four basic purposes was to "protect and enhance" the quality of the nation's air. Relying primarily upon this provision, environmental groups brought suit against EPA on May 24, 1972 to prevent the agency from approving State plans which failed to prevent significant deterioration of clean air.[8]

This challenge was ultimately successful and on December 5, 1974, EPA promulgated regulations to prevent emissions of sulfur dioxide and particulate matter from significantly deteriorating air quality in areas where concentrations of those pollutants were lower than the applicable national ambient standards. (39 FR 42510, Codified at 40 CFR 52.21). EPA incorporated its PSD regulations into the implementation plan of each State pursuant to Section 110(c) of the Act and established a procedure whereby EPA could delegate its PSD responsibility to States.

The regulations prohibited construction of stationary sources in any of nineteen specified categories unless EPA (or a delegate State) had issued a permit evidencing that the source would apply "best available control technology" (BACT) for SO_2 and for particulate matter and that emissions of those pollutants would not cause significant deterioration of clean air. For determining what levels of deterioration were "significant," the regulations set out an area classification system. Under it, clean air areas could be classified as Class I, II, or III. In Class I areas, small increases of SO_2 and particulate matter would be significant; in Class II areas moderate increases; and in Class III areas, increases up to an ambient standard. The regulations initially classified all clean areas as Class II, but gave States, Indian Governing Bodies, and Federal Land Managers the opportunity to reclassify their lands under specified procedures.

The 1977 Amendments affirm the PSD concept.[9] The new statutory scheme follows the outline of the pre-existing regulations, but is generally more comprehensive and restrictive. Some of the more significant changes introduced by the 1977 Amendments include:

- Formal Designation of PSD Areas - Under Section 107(d) of the new Act, all areas with air quality better than the national standards (or of indeterminate status) must be formally listed by EPA. EPA issued this list on March 3, 1978 (see 43 FR 8962). Several revisions have been made since March 3, 1978. (See 40 CFR 81.)

- More Restrictive "Increments" - The new law reduces the allowable short-term increments for SO_2 and particulates in Class II and III areas. Class I increments remain the same.[10]

- New Mandatory Class I Areas - The new Act retains the 3-class concept of the original PSD regulations and affirms the automatic Class II designation for most PSD areas. However, large national parks above a certain size which were in existence when the Amendments were enacted (8/7/77) are now designated as mandatory Class I areas. These areas may not be redesignated.[11]

- More and Different Sources Subject to PSD Review - The new Act increases the number of source categories subject to PSD preconstruction review from 19 to 28. New and modified sources within one of these 28 categories are subject to PSD if they have the potential* to emit or increase emissions by 100 tons per year of any pollutant regulated under the Act. Furthermore, all new or modified sources, regardless of their category, are covered if they have the potential to emit 250 tons or more per year of any regulated pollutant.[12] Major sources of fugitive dust are covered by the 250-ton criteria.

- More Restrictive Definition of BACT - Under the original PSD regulations, Best Available Control Technology (BACT) could not be more restrictive than the New Source Performance Standards (NSPS) applicable to the source category being proposed. Where NSPS had not yet been established for the applicable category, BACT was a case by case determination which weighed economic, technological, energy and geographic factors. Under the new law, BACT is always a case by case determination. Where New Source Performance Standards (or hazardous emission standards) apply to the proposed source, they represent a minimum, rather than a maximum, level of required control. More importantly, BACT now applies to all pollutants regulated under the Act, not just SO_2 and particulate matter as under the prior regulations.[13]

- Substantially Increased Monitoring and Modeling Requirements - Under the original PSD Regulations, applicants merely had to demonstrate, through atmospheric diffusion modeling, that the proposed emissions would not violate the allowable increments. The new law

*The Amendments define a major source in terms of its "potential to emit" but do not define the term "potential." EPA's final PSD regulations define "Potential to emit" as the capability at maximum capacity to emit a pollutant in the absence of air pollution control equipment.

imposes far more sophisticated modeling requirements. In addition, air quality and meteorologic monitoring requirements may now be required both prior to and after construction.[14]

- o Other Criteria Pollutants to be Regulated in the Future - EPA's original PSD regulations applied only to SO_2 and particulates. While the new law follows this policy on an interim basis, it directs EPA to promulgate PSD regulations for the other criteria pollutants (hydrocarbons, carbon monoxide, photochemical oxidants, and nitrogen oxides) by no later than August 7, 1979. These regulations will take effect one year after promulgation and must ultimately be incorporated into all State implementation plans. PSD regulations for lead must be promulgated in 1980.[15]

On November 3, 1977, EPA took four regulatory actions toward implementing the Act's new PSD requirements.[16] First, the agency promulgated amendments to the pre-existing PSD regulations, conforming them to Section 168(b) of the new Act. That section expressly made certain changes "immediately effective" as of August 7, 1977 (i.e., new mandatory Class I areas; the new increments; and new Class III reclassification procedures). The second action was the proposal of regulations giving guidance for the preparation of SIP revisions called for by the new PSD requirements of the Act. The third action was the proposal of comprehensive changes to EPA's pre-existing PSD regulations, conforming them to the new preconstruction requirements of Section 165. The fourth action was a decision to implement the new preconstruction review and BACT requirements of Section 165 as of March 1, 1978. Environmentalists had argued for an August 7, 1977 effective date while industry contended that the States had sole authority to implement these new requirements through SIP revisions.

EPA formally promulgated its new PSD regulations and SIP revision guidelines on June 19, 1978, three and one-half months after its original self-imposed deadline of March 1, 1978.[17] Due to the widespread public awareness of the March 1st date, however, EPA made the new PSD regulations retroactive to that date. Thus, sources which failed to obtain all necessary air pollution permits prior to March 1, 1978 will have to comply with the new PSD regulations. Furthermore, even those sources which did receive the necessary permits by March 1 will have to commence construction on or before March 19, 1979 in order to be exempt from the new requirements. This is the deadline for the submission of revised SIPs to EPA.

As defined by the new PSD regulations "fugitive dust" consists of native soil particles, uncontaminated by industrial pollutants, which become airborne through the forces of wind or human activities.[18] EPA has formally recognized the greater health impact of fugitive dust in urban as opposed to rural areas. In general, the particulate matter found in rural areas is composed of non-respirable native soil particles. Such particles are usually not exposed to potential contamination by industrial pollutants and, therefore, present an insignificant threat to public health. By contrast, the native soil in industrialized urban areas is typically contaminated by a variety of potentially harmful substances.

CHRONOLOGICAL DEVELOPMENT OF
PREVENTION OF SIGNIFICANT DETERIORATION

Sierra Club vs. Ruckelshaus
6/11/73

EPA's Original PSD Regulations
12/5/74

The 1977 Clean Air Act Amendments
8/7/77

EPA Proposes Its Revised PSD Regulations
and SIP Revision Guidelines
11/3/77

Effective Date of EPA's Revised PSD Regulations
(Sources Receiving Permits After This Date Must Comply)
3/1/78

EPA Promulgates List Designating PSD and Nonattainment
Areas Pursuant to Section 107 of the Act
3/3/78

EPA Formally Promulgates Its Revised PSD Regulations
and SIP Revision Guidelines
(Note: The Revised PSD Regulations
Are Retroactively Effective as of March 1, 1978)
6/19/78

Major Sources with Allowable Emissions in Excess
of 50 Tons Per Year Will Have to Begin Submitting
One Year's Continuous Monitoring Data
7/7/78

Revised SIPs Due to EPA
(Note: All Sources Commencing Construction After This
Date Are Subject to the New PSD Requirements, Regardless
of When They Received Their Permit)
3/19/79

Deadline for EPA Approval or Disapproval of the Revised SIPs
7/19/79

In light of such considerations and in recognition of a substantially higher level of human exposure in urban areas, EPA has decided to focus its fugitive dust control efforts in urban areas. In rural areas, controls are recommended for large man-made sources of fugitive dust such as tailing piles and surface mining operations which themselves are causing or contributing to NAAQS violations.*[19]

For purposes of PSD, EPA will not require applicants to analyze the air quality impacts of emissions which qualify as "fugitive dust." Nor will fugitive dust emissions consume the allowable PSD increments for particulate matter. In order to qualify for this exemption, however, applicants will have to demonstrate that the emissions are indeed composed of uncontaminated native soil. It should also be noted that for purposes of calculating potential emissions, fugitive dust emissions will be included. For example, while a proposed surface mining expansion with a potential fugitive dust emission rate in excess of 250 tons per year would be exempt from the PSD ambient air quality analysis requirements, it would not be exempt from the other requirements of PSD such as BACT.[20]

2.4 Formal Attainment Status Designations

The State Implementation Plan revisions called for by the Act's new PSD and nonattainment provisions are phrased in a way which requires the respective boundaries of a State's PSD and nonattainment areas be accurately defined. Specifically, Section 107(d) directs each State to submit a list of the NAAQS attainment status of all State areas to EPA. EPA promulgation of the formal list, with any necessary modifications, was required within 60 days of the submittal of the State lists. This promulgation appears in the Federal Register of March 3, 1978 (43 FR 8962, Codified at 40 CFR 81, Subpart C).

Section 107(d) specified that designations should be based upon air quality levels on the date of enactment of the Amendments (August 7, 1977). States were required by EPA guidance to consider the most recent four quarters of monitored ambient air quality data available. If this data showed no NAAQS violations, then the previous four quarters of monitoring data were to be examined to assure that the indication of attainment was not the result of unrepresentative meteorologic conditions. In the absence of sufficient monitored air quality data, other evaluation methods were used, including air quality dispersion modeling.[21]

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*In general, a new major source to be located in a rural area with infrequent short-term violations of the particulate NAAQS will be allowed to construct after applying the required controls provided that the dust in question is uncontaminated by pollutants from industrial activity and the emissions of the source in conjunction with emissions from other sources in the vicinity (excluding such dust) would not cause a violation of the applicable increment(s) or the applicable NAAQS, assuming as to the NAAQS an appropriate "non-urban" background concentration.

These Section 107(d) designations are subject to revision under Section 107(d)(5) whenever sufficient data is available to warrant a redesignation.[22] Both the State and EPA can initiate changes to these designations, but any State redesignation must be submitted to EPA for concurrence. Private parties must, therefore, work through the State or EPA to initiate a change in the designations.

The major significance of the 107 designations is not their impact on new source preconstruction review, but rather on the SIP revision process.* Specifically, the formal designations of 107(d)(1)(A)-(E) are incorporated into the SIP revision mandates of Part C (PSD) and Part D (Nonattainment) of the Act. Section 161 of Part C requires SIP revisions to prevent significant deterioration in each area identified pursuant to Section 107(d)(1)(D) or (E). Section 171(2) in Part D defines the term "nonattainment area" to include any area identified under subparagraph 107(d)(1)(A) through (C), while giving EPA authority to add other areas based upon monitoring or modeling data. Designation as a "nonattainment area" triggers the SIP revision process of Part D and a general ban on growth for nonattainment areas lacking an adequate SIP after June 30, 1979. Designation as a "PSD area" triggers the SIP revision process of Part C.

As noted in the preamble to the March 3rd Federal Register promulgation, the designation of an area as attainment or nonattainment must be considered only a point of departure and not as a final end in itself.[23] Indeed, the Act makes it clear that the designations are to be revised as appropriate to reflect more current or accurate data. It must also be noted that the designations will have only limited significance for new source revisions since under both the PSD and nonattainment preconstruction review procedures, major new and modified sources must undergo a preliminary modeling analysis to determine their impact upon all nearby areas as well as the area in which they will be located. Thus, case by case impact analysis is required to determine the impact on all neighboring areas and also to account for the possibility that an area with a particular designation may encompass "pockets" which do not fit that designation.

For designations of total suspended particulates (TSP), the localized nature of the violations precluded the use of general area-size criteria. States are therefore advised that designations along political boundaries such as city or county lines were practical from an air quality management standpoint. In the case of particulates, however, one very difficult problem was the appropriate designation of rural areas with significant levels of fugitive dust. As noted earlier, EPA's Fugitive Dust Policy recognizes the generally greater health impact and toxicity of urban as opposed to rural fugitive dust. Therefore, for the purposes of these designations, rural areas experiencing TSP violations which could be attributed to fugitive dust could claim attainment of the particulate NAAQS. Rural areas for this purpose are defined as those which have: (1) A lack of major industrial development or the absence of significant industrial particulate emissions, and (2) low urbanized population densities.[24]

The attainment status designations are very important with respect to major new fugitive dust sources.* Specifically, under EPA's soon to be revised Offset Ruling, major fugitive dust sources locating in clean portions of formally designated nonattainment areas or in attainment or unclassified areas shall be subject only to applicable requirements for preventing significant deterioration of air quality (see 40 CFR 52.21). Thus, only those major new fugitive dust sources locating in actual nonattainment areas will be subject to the Offset Ruling.**

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*Under both the Offset Policy and the PSD regulations the term "fugitive dust" refers to particulate emissions composed of soil which becomes suspended either by the forces of the wind or man's activity. This would include emissions from unpaved haul roads, wind erosion of exposed soil surfaces and soil storage piles and other activities in which soil is either removed, stored, transported, or redistributed.

**Even where the Offset Ruling is applicable to a major new or modified fugitive dust source, the soon to be revised Offset Policy exempts these sources from the requirement of demonstrating a "net air quality benefit" (Condition 4).

3.0 SIP REVISIONS FOR NONATTAINMENT AREAS - NEW FOCUS ON FUGITIVE DUST

3.1 Geographic Considerations

In analyzing particulate pollution, it is necessary to distinguish between rural and urban nonattainment areas. The sources and characteristics of particulate pollution in urban areas are fundamentally different from the rural counterparts. The most important distinction is the generally greater human health impact of urban particulate pollution. This is true in both a quantitative and qualitative sense in that urban particulate not only affects more people, but also is more hazardous than rural particulate matter. Indeed, contaminated urban fugitive dust, coupled with potentially hazardous direct industrial particulate emissions in urbanized areas, is one of the most serious air pollution problems facing the nation from a public health standpoint.

By contrast, rural particulates frequently consist of windblown native soil particles. Although these rural fugitive dust emissions are often the result of human activities such as agriculture, surface mining and quarrying operations, the actual particles are typically in the non-respirable size range and are uncontaminated by industrial pollutants. EPA's recent Fugitive Dust Policy recognizes the fundamental distinctions between rural and urban nonattainment problems for suspended particulates. Specifically, the policy provides that major emphasis for fugitive dust control in nonattainment SIP revisions should center on urban areas. It should be noted, however, that this general exemption for rural nonattainment does not extend to rural areas where traditional industrial particulate sources are causing or contributing to the NAAQS violations. These areas are treated the same as urban nonattainment areas.

With these geographic considerations as a backdrop, it becomes obvious that TSP nonattainment strategies will focus primarily on major metropolitan areas. Nearly 70% of the Nation's urbanized areas with populations exceeding 200,000 have been designated nonattainment for TSP.* Thus, the remainder of this section will focus on the issues inherent in developing acceptable TSP strategies for urban nonattainment areas.

3.2 Sources of Urban Particulate

For purposes of regulatory control strategy development, urban particulate sources are categorized either as traditional or nontraditional. The distinction is significant in terms of the types of control strategies required in the 1979 SIPs. Traditional sources, as the name implies, refer to those industrial sources which have been regulated under earlier SIPs. Both stack and fugitive emissions associated with such sources must, to the extent

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*Some suprising and noteworthy exceptions to the general nonattainment status include New York-NE New Jersey, Philadelphia, and Detroit.

necessary, be controlled through formally-adopted regulations in the 1979 SIP submittals.[25] Nontraditional sources are those which generally have not been controlled in the past. These include emissions from construction activities, demolition operations and resuspended street dust. By their nature, nontraditional sources are not amenable to the kind of straightforward emission limitations or design standards typically applied to traditional sources. The relationship between these nontraditional emissions and resultant ambient air quality concentrations has never been adequately defined. The development of emissions factors, modeling techniques, and effective control strategies for these complex sources will take time and study.

The SIP revision memorandum of February 4, 1978 recognizes these problems and authorizes states to submit control program *schedules* (as opposed to formal regulations) for control of nontraditional particulate sources.[26] These schedules must call for the expeditious development, adoption, submittal and implementation of the necessary control measures. For nontraditional particulates, field studies and demonstration projects to define the problem and investigate alternative controls should begin as soon as possible. If these studies reveal that major nontraditional control efforts are needed to meet the 1982 NAAQS deadlines, implementation of the required controls prior to the deadline will be necessary.

The significance of nontraditional sources to the TSP nonattainment problem has become apparent as traditional source control programs have failed to achieve ambient standards. In Greater Pittsburgh, for example, modeling of inventoried traditional sources is able to account for only 15% of measured TSP concentrations at certain monitors. Assuming the reliability of the model estimates and the accuracy of the traditional source emissions inventory, this means that fully 85% of ambient particulates are of nontraditional origins. One of the most significant findings of the 1976 National Particulate Assessment is that particulates from nontraditional sources contribute 25 to 30 $\mu g/m^3$ to citywide TSP levels, thereby preventing many urban areas from attaining the ambient standards.[27]

These nontraditional particulate emissions are the product of everyday urban activities. Five major nontraditional source categories have been isolated in urban areas. These include reentrained street dust, construction and demolition operations, exhaust (and tire) particles from automobiles, dust from unpaved roads and secondary particulates. The impact of these sources depends upon factors such as meteorology, topography, soil characteristics, road patterns and many other variables which control the impact of wind or traffic on surrounding particles. As a general rule, however, their air quality impact is more localized than stack emissions. As such, SIP revisions relating to nontraditional sources will have to be rather site and source specific.

Recent evidence indicates that wind and vehicle-induced reentrainment of street dust is probably the largest source of particulate matter in many downtown areas. Depending upon the amount and type of road dust, as well as the speed and level of traffic and wind conditions, reentrainment may contribute from 14-20 $\mu g/m^3$ to ambient TSP levels.[28] The potential toxicity of many of these particles, coupled with the high level of human exposure to them, would seem to demand heightened regulatory attention to this problem.

Automotive-related particulate emissions from tail pipes and tire wear are closely related to the reentrainment problem since much reentrained particulate originates from the automobile. The states' failure to address this problem creates a serious regulatory deficiency in that such emissions have potentially harmful health effects. For example, lead emissions from automotive exhaust contribute as much as 4 $\mu g/m^3$ in certain cities.[29] Such concentrations are believed to produce nervous system damage in children.[30]

The air quality impact of construction and demolition activities is also substantial in terms of total particulate loadings. These emissions are similar to those from unpaved areas generally in that they result from both wind erosion over exposed soil surfaces and man's physical activities on the surface. Heavy construction projects such as roadway construction and residential, commercial or industrial developments are clearly the largest contributors in this category. Field studies indicate an average dust generation rate of 1.2 tons of particulate per acre per month.[31] Applying this emission factor to a 150-acre residential construction project, the 100-ton major source threshold is exceeded in less than 3 weeks.

Another TSP source not addressed in earlier SIP strategies is "secondary particulate." Secondary particulates are the products of chemical reactions occurring in the atmosphere through forces of sunlight, temperature and meteorology in the presence of water vapor and gaseous atmospheric pollutants such as SO_2 and NO_x. Found in both rural and urban areas, secondary particulates are composed primarily of sulfates, organics and nitrates. They are a prime component of urban smog and are a major contributor to ambient TSP levels.

3.3 Control Strategy Development

From a conceptual standpoint, development of revised control strategies for TSP is a seven step process beginning with problem definition and ending with an adopted strategy demonstrating attainment of the 75 $\mu g/m^3$ primary standard before 1983. These seven steps include:

- o ANALYSIS OF EXISTING AIR QUALITY DATA
- o DEFINITION OF THE CONTROL AREA
- o PREPARATION OF AN EMISSION INVENTORY
- o ESTABLISHMENT OF SOURCE/AIR QUALITY RELATIONSHIPS
- o EVALUATION OF ALTERNATIVE CONTROL STRATEGIES
- o ANALYSIS OF ESTIMATED AIR QUALITY IMPROVEMENT
- o SUBMITTAL OF AN APPROVABLE SIP REVISION

STEP 1 - Analysis Of Existing Air Quality Data

Determination of the attainment status of an area is based upon measured TSP values at selected monitoring locations. High-volume samplers measure TSP concentrations at these monitoring sites by drawing ambient air through a particulate filter at a specified flow rate. The collected particles are then weighed and the results expressed in terms of micrograms per cubic meter of sampled air.

The purpose of the ambient air quality analysis is to determine the validity and representativeness of measured TSP concentrations. Assessing the validity of measured TSP levels is essentially a matter of determining that appropriate measurement techniques and quality control procedures have been employed. Assuming the data is valid, procedures must then be employed to determine whether it is representative of either a broad or confined area. Once the relationship between monitored data and contributing sources is established, it can then be used in a meaningful way to determine the actual origins of the nonattainment problem.

STEP 2 - Definition Of The Control Area

Based upon the results of the air quality analysis, the next step in the SIP revision process is to determine the boundaries of the area to be controlled. This determination is based upon the geographic scope of the nonattainment problem, and the location of the sources causing that problem. Both of these factors should have been defined in the air quality analysis.

STEP 3 - Preparation Of An Emission Inventory

Having defined the origins and geographic scope of the nonattainment problem, it is necessary to quantify the problem. This is accomplished by an emission inventory. This inventory allows strategists to assess the relative contributions to various sources and to determine the emission reduction impact of alternative control options.

As a practical matter, many states simply will not be able to submit "comprehensive, accurate and current emission inventories" by the Act's January 1, 1979 deadline. As an interim measure, therefore, these States will have to use emission estimates to help supplement their data base until more reliable emissions information becomes available. For traditional sources, EPA Document AP-42[32] and the 1976 TRC analysis of Emission Rates For Eighty Processes[33] are the primary references recommended by EPA for making these estimates. For nontraditional sources, emission estimates are less precise and reliance upon EPA's Guidelines for Development of Control Strategies in Area with Fugitive Dust Problems is required.[34]

STEP 4 - Establishment Of Source/Air Quality Relationships

Having defined and quantified the problem, it becomes necessary to quantify the relationship between area emissions and resultant ambient concentrations. Atmospheric diffusion models are employed for this purpose.

The selection of an appropriate diffusion model is a highly technical determination requiring the services of trained professionals. EPA has published numerous guideline documents on atmospheric modeling, the most recent being its 1978 Guideline on Air Quality Models.[35] This guideline specifies the models to be used in assessing control strategies for TSP and other criteria pollutants. Approval of the Regional EPA Administrator is required before other modeling approaches may be employed.

One of the major gaps in current air pollution knowledge concerns the modeling of fugitive emissions and fugitive dust sources. EPA's modeling guidelines deal primarily with stack emissions. Modeling techniques are relatively well developed for stack emissions. Due to the complex configuration and emission characteristics of fugitive sources, however, traditional modeling techniques are not adequate. At the present time, therefore, modeling the impact of fugitive control strategies will be reviewed by EPA on a case by case basis. EPA is now studying alternative modeling techniques for both fugitive process emissions and fugitive dust sources. It is doubtful that these studies will produce satisfactory fugitive modeling techniques in the near future.

STEP 5 - Evaluation Of Alternative Control Strategies

The prior steps in the strategy development process lead naturally to the evaluation of control alternatives. The objective of this step is to identify the most cost-effective means of ensuring timely attainment and maintenance of the ambient standards.

The first task in this assessment is to identify the sources (traditional and/or nontraditional) which will receive priority attention for control. Many considerations guide this determination, including economic costs, administrative and social impacts, technological difficulties, and the air quality benefits associated with imposition or tightening of controls on a particular source. A comprehensive analysis of these policy considerations is beyond the scope of this paper. Suffice it to say that this task forms the cornerstone of the control strategy and should be treated accordingly by State and local air quality strategists. The ultimate objective is to achieve the greatest level of emissions reduction over time at the lowest per capita cost. The selection of target sources sets the stage for realizing that objective.

Once the target sources have been identified, it becomes necessary to estimate the degree of emission reduction that will occur from full compliance with adopted controls. Such control measures may be in the form of increased or expedited enforcement of existing regulations, or entirely new regulations and programs. These emissions reduction estimates comprise the key variable in the modeling exercise through which reasonable progress toward timely NAAQS attainment is demonstrated.

For existing sources, control strategies will have to impose reasonably available controls (RACT) to the extent necessary for NAAQS attainment. This is particularly true for major industrial centers where traditional stationary sources have not been subject to such controls under prior SIP's. RACT may represent a relatively stringent level of control for these sources and may involve forcing technological innovation.* For all sources, however,

*Various efforts have either been completed or are underway to provide guidance for determining RACT. Technical support documents for new source performance standards are a valuable information source. In addition, EPA is currently in the process of issuing documents which describe RACT for numerous particulate sources covering both stack and fugitive emissions.

RACT will be determined on a case by case basis taking into account such factors as retrofit feasbility, economic and energy costs and environmental impacts.[36]

In urban areas where fugitive dust is causing high TSP levels, nontraditional source controls for reentrained dust, construction activities, automotive particulates and secondary particulates will have to be considered. While the impact of these sources are generally localized in nature, they are typically found throughout a given area and may, therefore, create widespread TSP violations. The emission inventory should provide the spatial segregation and emissions quantification necessary for evaluating these sources.

Control of reentrained street dust is one of the most important nontraditional source programs which should be considered. Unfortunately, it is also one of the most poorly understood particulate problems. Field studies indicate that reentrained street dust consists primarily of sand and soil, and automotive exhaust and debris from automotive wear and tear of brakes, bearings and tires. Findings from these studies suggest two basic methods of control:[37]

(1) Reducing the amount of material being deposited onto roadway surfaces through curbing, gutter and sidewalk improvements, enforcement of visible emission regulations for automotive vehicles, improvements in sanding operations for snow and ice control and increased controls on street dust loadings from construction vehicles.

(2) Street cleaning through sweeping and/or water flushing operations. Common sense would indicate that street cleaning should reduce reentrained dust, but it is still unproven as an effective method for reducing ambient TSP concentrations.

Urban roadway improvements anticipated in the next several years are expected to result in significant reductions of re-entrained dust. As older vehicles are phased out and automotive particulate contributions decrease, TSP levels can be expected to decline in high traffic areas. State or local agencies seeking to accomplish additional control over re-entrained dust should refer to EPA's recent guideline document on Control of Re-entrained Dust From Paved Streets.[38] This guide provides more detailed information on street cleaning, construction site control and the costs associated with these controls. With respect to construction-related emissions, wetting or soil stabilization of exposed surfaces and access roads appear to be the most feasible control alternatives. Consideration should also be given to limiting the length of time that any cleared area may remain exposed before construction activities commence.

Another nontraditional control which will have to be considered in some areas concerns secondary particulates. Although the relationships between the precursors of secondary particulates (SO_x, NO_x and organics) are not well understood, it is suspected that these pollutants contribute from 5-15 $\mu g/m^3$ of the particulate matter in many urban areas, much of it in the respirable size range.[39] To achieve the primary NAAQS for TSP, efforts will therefore have to focus on controlling SO_x, NO_x and organic emissions so as to prevent the formation of sulfate, nitrate and organic aerosal particulates. While

additional understanding of aerosol formation is needed, urban TSP control strategies should at least consider reduction of secondary particulate precursors as a possible alternative.

STEP 6 - Analysis Of Estimated Air Quality Improvement

Having identified alternative control strategies, the next step in the process is to estimate the air quality impact of each alternative. This is accomplished through application of the atmospheric diffusion models already selected under Step 4. This model translates estimated emission reductions over time into estimated resulting air quality. Alternative strategies are then manipulated until timely attainment of the ambient standards can be demonstrated.

STEP 7 - Adoption And Submittal

The SIP Revision Memorandum, as noted earlier, distinguishes between traditional and nontraditional source controls in terms of how they must be addressed in the 1979 SIP submissions. All necessary stack and fugitive emission limitations for traditional sources must be adopted and submitted by January 1, 1979. Where nontraditional source control is part of the attainment plan, however, formal regulations need not address them. For these sources the plan must instead contain:

(1) an assessment of the impact of nontraditional sources, and

(2) a commitment by the state to develop, submit and implement the appropriate procedures and programs. Obviously, these schedules must include milestones for evaluating progress toward timely attainment of the standards. Recognizing that the development of these programs will take considerable time, the memorandum advises states to initiate the necessary studies and demonstration projects for controlling nontraditional sources as soon as possible.

3.4 Conclusions and Recommendations

Defining and evaluating control options for TSP nonattainment will obviously take a great deal of study and effort. The development of an effective control strategy requires an accurate identification of the sources of the problem, a quantification of emissions from those sources and an understanding of the relationship between these emissions and air quality. Once this information has been assembled, a control strategy can be developed which reflects the relative contribution of traditional, nontraditional and background sources to known NAAQS violations.

Particulate control strategies have traditionally been oriented toward fuel combustion, process emissions and incineration. Control programs for particulates have resulted in substantial emission reductions and resulting air quality improvements in many urban areas. Yet, further reduction in these emissions can still be achieved. Several specific recommendations were made in the National Particulate Assessment, with regard to traditional sources. These included:

- Increased regulatory and enforcement attention directed to the primary metals and minerals industries;

- Increased inspection, source sampling and enforcement activity to obtain compliance by major sources with existing regulations;

- Promulgation and enforcement of numerical or visible "property-line" fugitive emission standards; operating and maintenance standards for specific processes such as storage piles and material handling equipment; and increased paving of industrial roadways and parking areas;

- Increased regulatory attention to residential and commercial space heating boilers where they are shown to be a problem;

- Considerable tightening of emission limits for fuel-burning installations, especially oil-fired operations;

- Regulation of industrial process emissions on an industry-specific basis. Under existing regulatory practice, general process weight standards are established for all processes, regardless of how difficult it is for any particular industry to meet them;

- Consideration of extending regulatory coverage to smaller industrial point sources where the contribution of such sources to NAAQS violations is found to be significant.

Even with the traditional control measures discussed above, it is apparent that many urban areas will not be able to attain the ambient TSP standards before 1983. This is the single most important conclusion of the National Particulate Assessment. It presents difficult planning problems for urban air quality strategists since emission reduction efficiencies are lower (and less certain) for nontraditional sources than for traditional sources. The difficulties lie not so much in the technical feasibility of nontraditional controls as in their cost and political implications.

Still, much can be done to control particulate emission from automobiles, and construction activities and from re-entrainment of particulates from urban streets.

Finally, it is important to remember that there are a number of gaps in current data and understanding which will have to be overcome before TSP control strategies can be evaluated with the degree of precision necessary. Two of the most critical areas of needed investigation are siting criteria for TSP monitors and the relationship between meteorology/climatology/topography and ambient TSP concentrations. Improved emissions/air quality data bases and expanded research into techniques for modeling and controlling fugitive emissions and fugitive dust will also be important to the control effort.

FOOTNOTES

1. Clean Air Act (hereafter "CAA") §109.
2. 40 CFR 50.
3. 43 FR 46246.
4. CAA §110(a)(2)(A).
5. 40 CFR §51.13 and 51.18.
6. 41 FR 55524.
7. CAA §110(a)(2)(I).
8. Sierra Club vs. Ruckelshaus, 4 ERC 1205.
9. CAA, Subchapter I, Part C, Subpart 1.
10. CAA §162.
11. CAA §162.
12. CAA §165 and §169(1).
13. CAA §165(a)(4) and §169(3).
14. CAA §165(c).
15. CAA §166.
16. 42 FR 57459.
17. 43 FR 26380, 40 CFR 51.24, 52.21.
18. 40 CFR 52.21(b)(b).
19. 43 FR 26395.
20. Id.
21. 43 FR 8962.
22. CAA §107(d)(5).
23. 43 FR 8963.
24. Id.

25. 43 FR 21676.

26. Id.

27. EPA-450/3-76-025, p. 30.

28. Id. at p. 78.

29. Id. at p. 70.

30. 43 FR 46247.

31. EPA Guideline for Development of Control Strategies in Areas With Fugitive Dust Problems, EPA-450/2-77-029 at p. 3-10.

32. This document establishes emissions factors for many air pollution sources.

33. EPA Contract Report No. 68-02-1382 T.O. #12.

34. Supra, note 31.

35. EPA-450/2-78/027, May 1978.

36. December 9, 1976 EPA memorandum from Roger Strelow to EPA Regional Administrators.

37. Supra, note 27 at pl 128.

38. EPA-907/9-77-007.

39. Supra, note 27 at p. 48.

THE IMPACT OF FUGITIVE EMISSIONS ON TSP CONCENTRATIONS IN URBAN AREAS

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ABSTRACT*

The primary cause of total suspended particulate (TSP) violations in urban areas such as Chicago, Phoenix, Philadelphia, St. Louis, Miami, Denver and many others, is due to fugitive emissions. These fugitive emissions, contributing from 40% to 70% of the TSP concentrations, are principally from a single source - suspension of roadway dust and pavement aggregate by vehicles. This roadway dust is mainly pavement aggregate but also includes rubber tire fragments and other deposited dusts such as soil minerals. The contributions from various entrainment mechanisms such as direct tire erosion or tire and vehicle turbulence is unknown but have a direct impact on control strategies.

Microscopical analysis supplemented with chemical analysis has proven to be the most useful method of determining the sources of fugitive emissions. This analytical protocol will be described in detail along with the techniques for source assignments.

* This paper was not available at the time of the Proceedings' publication.

EMISSIONS AND EFFLUENTS FROM COAL STORAGE PILES

by

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ABSTRACT

Coal storage piles are sources of atmospheric emissions of fugitive dust and nonpoint effluent discharges.

Pile geometry, coal erodability (dustiness), wind, humidity, precipitation and temperature are the major parameters known to contribute to the emissions from coal storage piles. From the literature and a source-oriented sampling program, an equation was derived describing the mass emission rate from a given coal pile. This program also showed that the variability of the emissions from a specific coal pile are much greater than that between piles. This is due to the large influence that wind speed and the precipitation-evaporation index have on emissions. Emissions were analyzed utilizing x-ray fluorescence and atomic absorption. Particle size analyses were conducted on two samples. Over 97 percent of the particles, by number, were smaller than 10 microns. Eighty-eight percent of the particles from a composite Brink® sampler taken over two days of sampling were smaller than 5 microns in size.

The water pollution potential of coal stockpiles maintained outdoors at production and user sites was also studied. These storage piles are sources of effluents due to the drainage and runoff of wastewater which occurs during and after precipitation. The runoff usually flows from the drainage area into the nearest waterway. In this study, the effluent levels from these sources were quantified by examining coals (both freshly mined and aged) from six coal regions of the United States. Coals were placed under a rainfall simulator and grab samples of the drainage were collected. The samples were analyzed for organic and inorganic substances and for water quality indicators.

The coal leachate and air samples were also analyzed for selected polycyclic organic materials, and only small quantities of POM's were detected.

INTRODUCTION

Water effluents and air emissions exist at coal stockpiles maintained outdoors. Water effluents form from the drainage and runoff of wastewater which occurs during and after precipitation. Air emissions form from the action of wind forces on the exposed coal surface.

Effluents were evaluated from six major coal regions of the United States (1). Data were obtained by placing several coals beneath a rainfall simulator and collecting integrated samples of the drainage. The samples were analyzed for chemical compositions and water quality indicators. Hydrologic relationships were used to estimate the runoff concentrations of full size stockpiles from the simulator concentrations.

Emissions were evaluated using field sampling of typical stockpiles and wind tunnel studies of the erosion process (2). Air emissions were analyzed for chemical composition and air quality indicators.

SOURCE DESCRIPTION

Three-fourths of all coal produced is consumed at electric utilities. In 1975 there were approximately 950 coal stockpiles containing 124×10^6 metric tons of coal at user facilities throughout the United States. These stock piles are maintained outdoors and exposed to a variety of atmospheric conditions. Rainfall leaches pollutants from the stockpile which drain into waterways. Aquatic life forms in the waterways are thus exposed to the pollutants. Drainage in coal mining operations produces high sulfate concentrations and low pH values in nearby streams. Hence, the potential for this same problem exists at coal stockpiles because pyrites, the prime factor in acid mine drainage, exist within the coal pile.

Wind blows the loose and fine coal away from the stockpile. The combined leaching and drying of the coal causes fracturing which releases more fine material that can become airborne. Mechanical movement of the stockpile during usage also releases or fractures more coal.

In addition, coal contains inorganic substances in the "ash" (extraneous mineral matter) and in the coal structure; these substances are released. Because coal is primarily organic, organic contaminants are also released.

AIR EMISSIONS

Carbon monoxide (CO) hydrocarbons and particulate matter are the criteria pollutants emitted. Concentrations of carbon monoxide and hydrocarbons are barely detectable (<20 ppm CO, and <5 ppm hydrocarbons); these can be expected to be three orders of magnitude below ambient air quality criteria at a distance of 50 meters from the pile. The average emission factor for respirable particulates (those smaller than seven microns in diameter) is 6.4 milligrams/kilogram of coal per year. The composition of the emissions from coal piles is essentially that of coal dust. There is no significant increase in the level of any known hazardous species other than the normal increase found when coal is fractured to produce dust. This increase is shown in Table 1.

TABLE 1. COMPOSITION OF COAL AND COAL DUST

Element	Amount in coal, ppm	Amount in coal dust, ppm
Arsenic	0.3	26
Cadmium	0.2	3.8
Copper	25	868
Iron	1,600	79,200
Nickel	2.7	755
Vanadium	12	166

Pile geometry, coal erodability (dustiness), wind, humidity, precipitation and temperature are the major factors known to contribute to the emissions from coal storage piles. Coal erosion has been studied in a wind tunnel, and the effects of most of these factors, or their equivalents, have been evaluated. Equation 1 describes the mass emission rate from a given coal pile in terms of these factors.

$$Q = (336)\frac{u^3 \rho_b{}^2 s^{0.345}}{(P-E)^2} \qquad (1)$$

Q = emission rate, mg/s

u = wind speed, m/s

ρ_b = coal density, g/m^3

s = surface area, m^2

$(P-E)$ = precipitation-evaporation index

During this study, variability of the emissions from a specific coal pile were determined to be much greater than the variation of emissions from one coal pile to another. This is due to the large influence that wind speed and precipitation-evaporation rate have on emissions. The location of a specific coal pile will have a major influence on the emission rate. One storage pile sampled one day apart showed emission rates of 14 and 41 milligrams per second. The emission rate may also be essentially zero, as it was during some days of the sampling program. One of three sampling tests showed undetectable contributions due to the storage pile.

Because about 90 percent of the coal stored in the United States is stored in P-E index regions of 90 to 130 (Figure 1), the effect of the P-E index is reduced substantially for the average coal pile. In addition, the distribution of coal piles shows that a tremendous quanity of small piles exist. The most frequently occurring coal pile size is 49,000 metric tons; the average size is 95,000 metric tons (Figure 2).

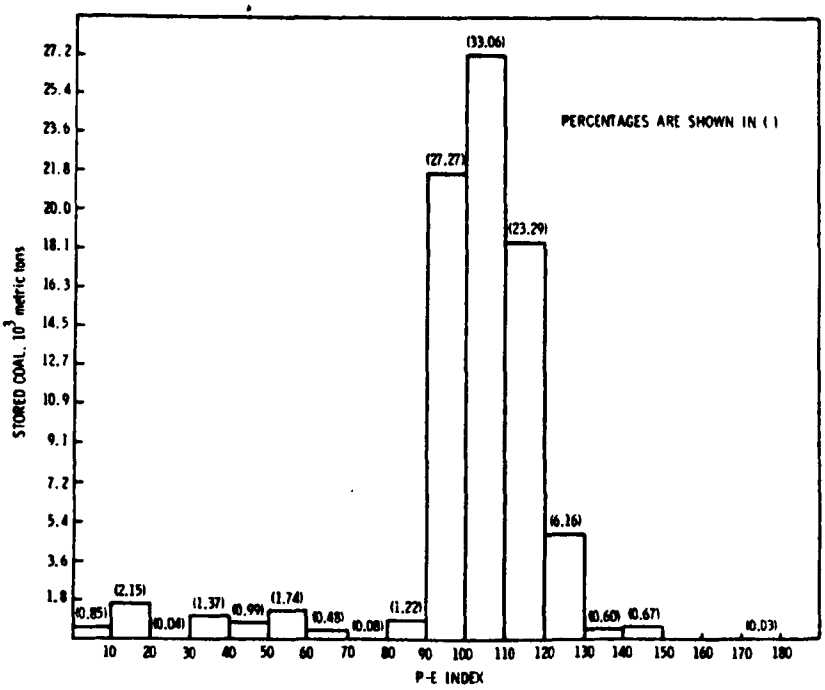

Figure 1. Quantity of coal stored as a function of P-E index.

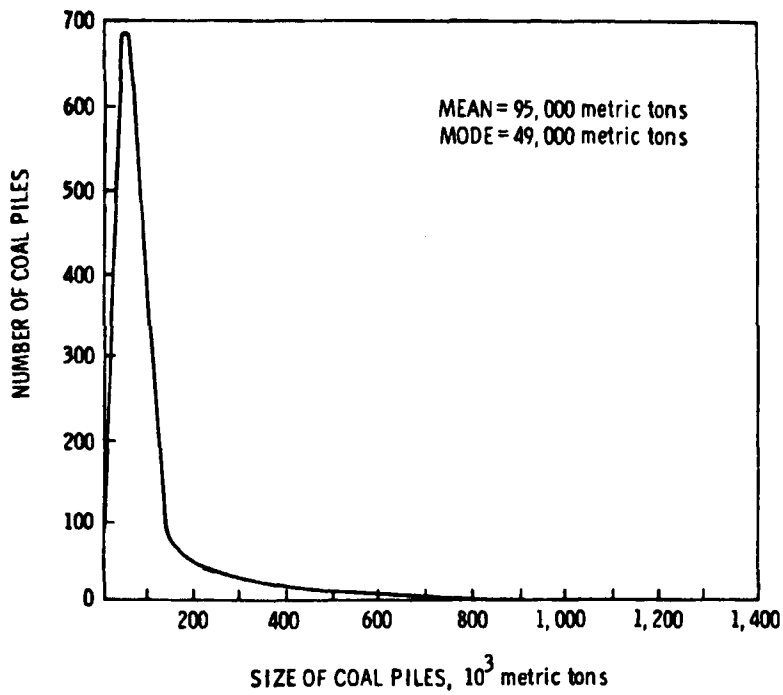

Figure 2. Distribution of coal storage pile sizes.

A coal storage pile of 112,000 metric tons was sampled at two seasons of the year: late March and mid-August. Only small variations in the emissions were observed between these two sampling sets. The average emission factor was 6.4 milligrams per kilogram of coal per year; the estimated population standard deviation was 2.9 milligrams per kilogram of coal per year. The emissions were analyzed utilizing x-ray fluorescence and atomic absorption; their compositions were essentially that of coal dust. Particle size analyses were conducted on two samples. Over 97 percent of the particles, by number, were smaller than 10 microns. Eighty-eight percent of the particles from a composite Brink® sampler taken over two days of sampling were smaller than 5 microns in size. Based upon these results, it is concluded that essentially all of the emissions are in the respirable range. No fibers were detected in any of the samples. The coal samples were also extracted with pentane and analyzed for selected polycyclic organic materials using chemical ionization mass spectroscopy. Although small quantities (<0.5 ppm) of benzo(c)phenanthrene, benzo(a)pyrene and 3-methylcholanthrene were detected, the quantity identified was insignificant compared to the quantity of coal dust.

In order to evaluate the impact of coal storage on the environment, a representative coal pile was defined from average parameters. This representative stockpile would contain 95,000 metric tons of bituminous coal piled to an average height of 5.8 meters. The pile would be located in a P-E region of 91, be exposed to an average annual wind speed of 4.5 meters per second, and be located 86 meters from the nearest plant boundary. Assuming that this coal pile is round, Figure 3 shows the approximate relationship between the coal pile, the plant boundary and a typical emissions radius. The ambient concentration of particulate at different distances is given in Table 2 based on a particulate emission rate of 19 milligrams per second or 610 kilograms per year from the average pile. These ground level concentrations were calculated using dispersion methodology as described by Turner (3). The coal pile which was sampled in this study had parameters very similar to those of the representative stockpile.

TABLE 2. AMBIENT CONCENTRATIONS OF PARTICULATE

Distance from coal pile,	Ground level concentration, $\mu g/m^3$	
	Instant	24-hr average
86	18	6.4
528	1.9	0.67

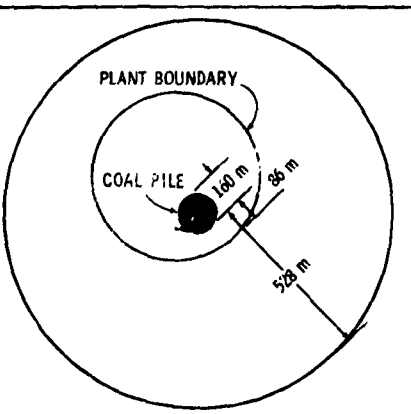

Figure 3. Emissions radius of coal pile.

WATER EFFLUENTS

Because coal is a complex aggregate capable of discharging a vast range of compounds, the study (1) was limited to compounds listed on the EPA Toxic Substances List (4), pollutants with effluent limitations for coal storage areas, and other water quality criteria used to indicate the presence of classes of compounds. Due to the diversity of stockpiles and coals, several samples were collected from each of the six major coal regions of the United States. Aged coals and fresh coals were collected. Samples were selected to represent the range of inorganic element content in coal and to obtain coals that were representative of each coal region.

The apparatus shown in Figure 4 was used to simulate rainfall of representative intensity and duration. Coal samples were placed under this apparatus for exposure to simulated rainfall. Drainage seeped through the coal, out the bottom of the pans, and into collection bottles. A background sample was also obtained of the rainfall water used. Three simulation runs were completed over a period of 30 days. The time between runs was varied to observe its effect. The average effluent concentration found for each coal region is presented in Table 3.

The effluent levels from a representative stock pile were computed as coal-production-per-region weighted averages. Table 4 presents these levels at the source. However, the pollutant concentration levels in the stream that enters the nearest waterway result from the dilution of pile drainage by runoff waters in the entire coal storage area.

The representative storage pile was assumed to be located 86 meters from the receiving waterway. The representative rainfall rate is 0.7 centimeter per hour over a stockpile area of 18,792 square meters. This rainfall occurs 139 days per year (i.e., every 2.6 days) and lasts for approximately one hour. However, only 15 percent of the rainfall volume on the pile appears as direct runoff used in the rational method of hydrology (5). Volumetric flow from the pile was computed as 21 cubic meters per hour.

Runoff from the entire coal storage area was obtained from a survey of coal storage sites; the average runoff was 610 cubic meters per hour (6).

The concentration levels shown in Table 4 are diluted by the drainage area volumetric flow to obtain the effluent concentrations shown in Table 5. Thorough mixing of runoff waters with pile drainage is assumed. The coal aggregate retards the runoff flow for a time period sufficient to enable mixing of upstream runoff.

CONCLUSION

Pollution from coal stockpiles is a site-specific problem. Large, aged coal stockpiles located in areas of frequent and light rainfall will generate much higher effluent concentrations than that of the representative pile used in this study. The variability of the emissions is also dependent on the site, although local meterology will be the dominating factor. Air emissions will not be a major problem because of the small contribution to ambient air quality at typical exposure distances, even for coal stockpiles of several million tons. Water effluents from Interior Western (Iowa to Texas and Arkansas to Nebraska) and possibly the Great Northern Plains (Montana, North and South Dakota) could

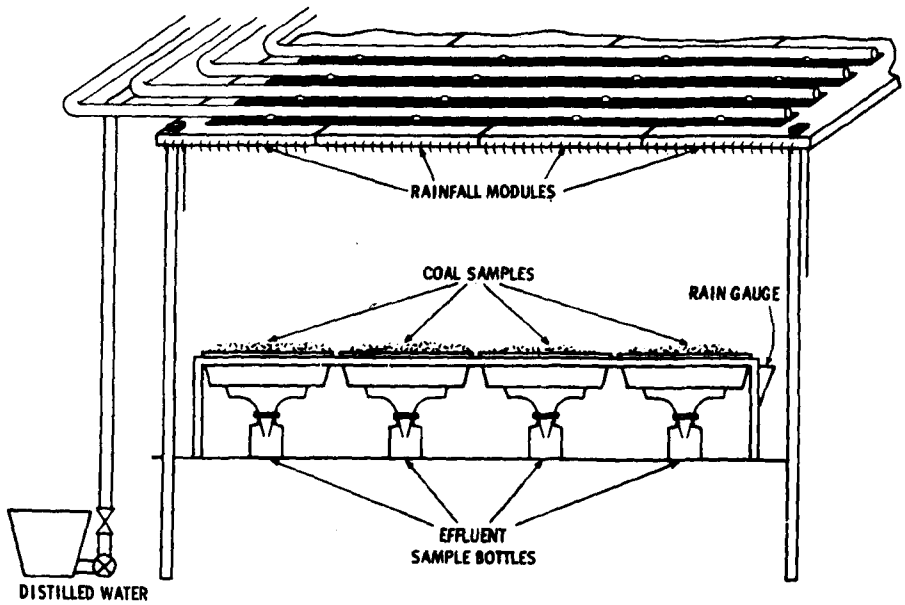

Figure 4. Rainfall simulation apparatus.

TABLE 3. AVERAGE EFFLUENT DRAINAGE CONCENTRATION FOR EACH COAL REGION

	Effluent concentration, g/m³					
Effluent parameter	Appalachian	Great Northern Plains	Interior Eastern	Interior Western	Western	Southwestern
Total suspended solids	1,521	1,282	1,264	1,853	2,486	1,538
Total dissolved solids	259	430	1,136	5,539	1,900	356
Sulfate	66	1,598	648	4,860	240	190
Iron	3.1	1.5	9.1	1,131	8.2	5.5
Manganese	0.03	0.14	0.44	17.9	0.4	0.04
Free silica	12.3	NDL[a]	0.8	86.3	NDL	NDL
Cyanide	<0.001	NDL	0.002	NDL	NDL	NDL
BOD_5	<5.0	<7.5	NDL	<1.2	<2.5	<7.5
COD	1,407	1,324	1,556	1,053	1,826	769
Nitrate	0.12	0.14	0.33	0.09	1.8	0.16
Total phosphate	NDL	NDL	NDL	NDL	NDL	NDL
Antimony	2.1	NDL	7.5	10.3	14.0	6.5
Arsenic	23	1.8	4.1	10.1	5.6	4.1
Beryllium	NDL	NDL	NDL	NDL	NDL	NDL
Cadmium	NDL	NDL	NDL	0.05	0.005	NDL
Chromium	NDL	NDL	NDL	0.03	0.04	NDL
Copper	0.02	NDL	NDL	2.2	NDL	0.02
Lead	0.05	0.05	0.06	0.33	0.07	0.05
Nickel	0.06	0.02	0.09	10.2	0.05	0.03
Selenium	23.8	NDL	12.5	25.2	15.0	21.5
Silver	NDL	NDL	NDL	NDL	NDL	NDL
Zinc	0.008	0.17	0.14	25.0	0.15	0.04
Mercury	<0.001	0.003	NDL	0.004	0.005	0.002
Thallium	NDL	NDL	NDL	NDL	NDL	NDL
pH[b]	6.28	6.93	7.62	2.81	7.24	6.60
Chloride	0.33	NDL	NDL	2.3	NDL	NDL
Total organic carbon	251.7	373.2	380.1	90.5	318.4	158.7

[a]No detectable level. [b]Negative logarithm of hydrogen ion concentration.

TABLE 4. COAL PRODUCTION-WEIGHTED EFFLUENT CONCENTRATION

Effluent parameter	Concentration, g/m^3
Total suspended solids	1,551
Total dissolved solids	754
Sulfate	401
Iron	39
Manganese	0.69
Free silica	10.1
Cyanide	<0.001
BOD_5	<3.8
COD	1,436
Nitrate	0.31
Total phosphate	NDL[a]
Antimony	4.6
Arsenic	15.7
Beryllium	NDL
Cadmium	0.002
Chromium	0.004
Copper	0.08
Lead	0.06
Nickel	3.1
Selenium	19.9
Silver	NDL
Zinc	0.80
Mercury	<0.001
Thallium	NDL
pH	6.78[b]
Chloride	0.27
Total organic carbon	280

[a] No detectable level.

[b] Negative logarithm of hydrogen ion concentration.

TABLE 5. CALCULATED RUNOFF CONCENTRATIONS FROM THE REPRESENTATIVE COAL STORAGE FILE

Effluent parameter	Concentration entering waterways, g/m^3
Total suspended solids	7.8
Total dissolved solids	3.8
Sulfate	2.0
Iron	0.2
Manganese	3.4×10^{-4}
Free silica	0.05
Cyanide	$<5 \times 10^{-6}$
Nitrate	1.5×10^{-3}
Total phosphate	NDL[a]
Antimony	0.02
Arsenic	0.08
Beryllium	NDL
Cadmium	1×10^{-5}
Chromium	2×10^{-5}
Copper	4×10^{-5}
Lead	3×10^{-4}
Nickel	0.02
Selenium	0.1
Silver	NDL
Zinc	4×10^{-4}
Mercury	5×10^{-6}
Thallium	NDL
Chloride	1.3×10^{-3}
Total organic carbon	1.4
2-Chloronapthalene	7×10^{-5}
Acenapthene	7×10^{-5}
Fluorene	7×10^{-5}
Fluoranthene	8×10^{-5}
Benzidine	7×10^{-5}
Benzo(ghi)perylene	2×10^{-4}

[a] No detectable level.

present a major effluent problem which would require neutralization and sedimentation of the runoff (primarily for control of acid).

ACKNOWLEDGMENT

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URANIUM MILL TAILINGS AREA

FUGITIVE EMISSION

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ABSTRACT FOR THE THIRD SYMPOSIUM ON FUGITIVE EMISSIONS:

MEASUREMENT AND CONTROL

Uranium Mill Tailings Area Fugitive Emissions

For the past year and a half, an environmental assessment of the expansion of Canada's largest uranium mining area has been underway. Investigation of the air quality aspects has involved an extensive study of tailings area fugitive dust emissions by field monitoring and emission and dispersion modelling.

The hi-vol field program resulted in approximately 500 samples of 24-hour suspended particulate concentrations at locations in the immediate vicinity of seven tailings areas. Micrometeorological stations located on the tailings areas, provided wind speeds, wind directions, rainfall and other meteorological data at the source during some of the dust sampling periods. There were very large day to day changes in suspended particulate levels next to tailings areas. The majority of the readings obtained were less than 60 $\mu g/m^3$ with an occasional value above 1000 $\mu g/m^3$ and a number of values below 20 $\mu g/m^3$. It was ascertained that suspended particulate levels are highly dependent on tailings moisture content and surface windspeed.

A computer model for the emissions and dispersion of dust from the tailings areas was developed based on the concepts of particle saltation, suspension, and deposition and atmospheric transport using a Gaussan tilted plume model. The selection of a suspension factor of 10^{-7} per meter resulted in good agreement between the model and the field program results. The ratio of suspended particulate concentrations calculated from the tailings to actual measured suspended particulate ranged from 0.07 to 2.91, with an average ratio of 1.1.

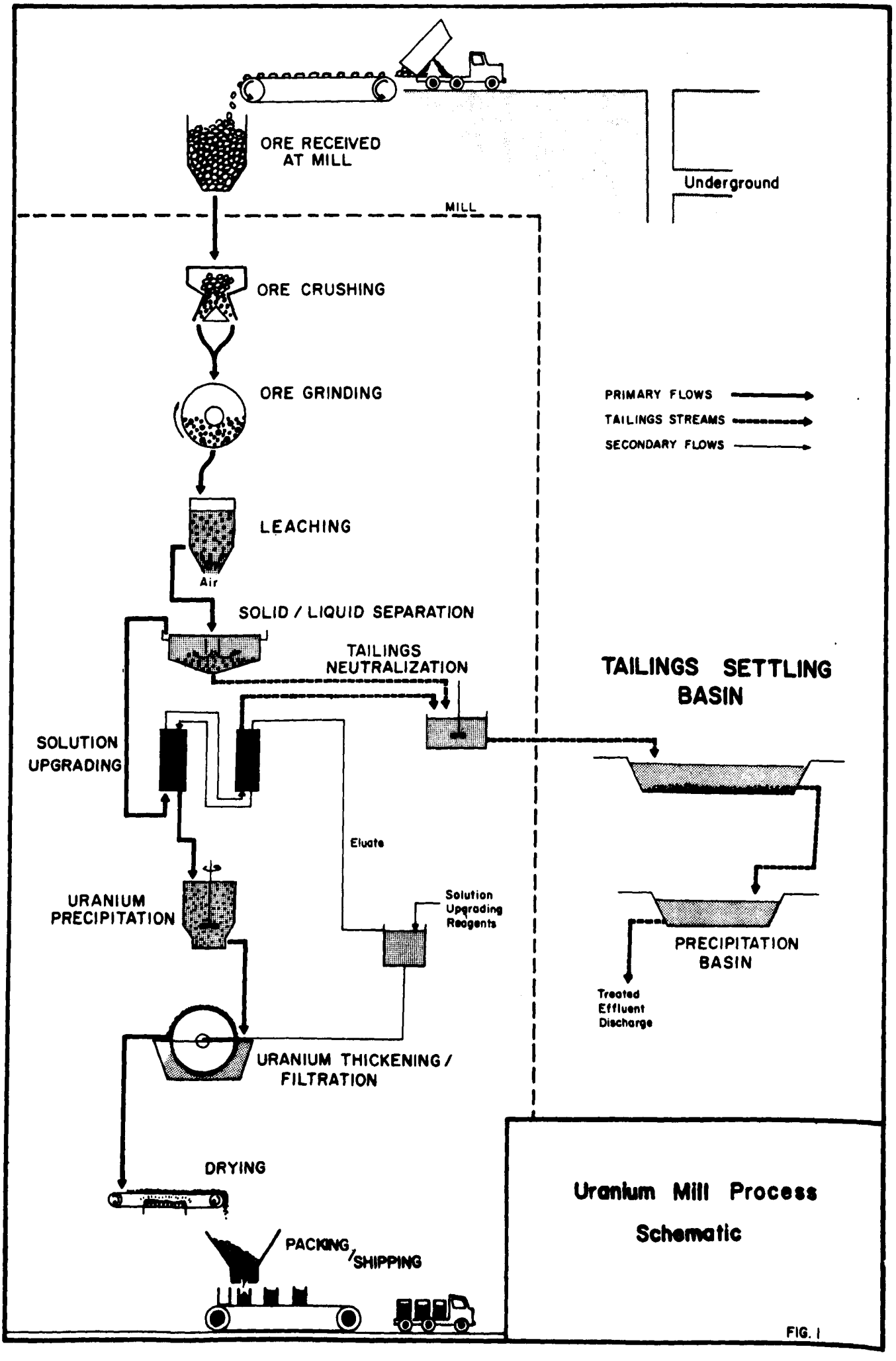
URANIUM MILL TAILINGS AREA FUGITIVE EMISSIONS

Since the fall of 1976, an environmental assessment of the expansion of Canada's largest uranium mining area has been underway. Investigation of the air quality aspects has involved an extensive study of tailings area fugitive dust emissions by field monitoring and emission and dispersion modelling, with the intent of predicting the potential impact of tailings emissions after mine expansions.

In Elliot Lake region of Canada, underground mining of uranium deposits in quartz-pebble conglomerates has been conducted since the 1950's and is expected to continue until the turn of the century. In underground mining, the ore is drilled, blasted, transported to underground primary crushing, hoisted to the surface, crushed again and conveyed to the mill for uranium extraction. A "typical" mill illustrated in the schematic shown (Fig. 1) consists of ten primary steps:

Ore Crushing
Ore Grinding
Leaching
Solid/Liquid Separation
Solution Upgrading
Uranium Precipitation
Uranium Thickening/Filtration
Drying
Packaging
Tailings Treatment/Discharge

After tailings neutralization, the tailings slurry is disposed of in containment areas. Within the Elliot Lake region, the tailings areas are usually natural basins often containing swamps and small lakes with waste rock used for containment dams. The uranium mining industry has been active in this area since the 1950's and there are presently both active and inactive mines and mills. There are a total of seven majcr tailings areas of which two are


ORE RECEIVED AT MILL
Underground
MILL
ORE CRUSHING
ORE GRINDING
PRIMARY FLOWS
TAILINGS STREAMS
SECONDARY FLOWS
LEACHING
Air
SOLID / LIQUID SEPARATION
TAILINGS NEUTRALIZATION
TAILINGS SETTLING BASIN
SOLUTION UPGRADING
Eluate
URANIUM PRECIPITATION
Solution Upgrading Reagents
PRECIPITATION BASIN
Treated Effluent Discharge
URANIUM THICKENING / FILTRATION
DRYING
PACKING / SHIPPING
Uranium Mill Process Schematic
FIG. 1

now active and five inactive.

Very little information is presently available about windblown particulate from tailings areas. Dust emissions from tailings areas share many similarities with the common fugitive sources such as unpaved roads, agricultural tilling operations, aggregate storage piles and heavy construction operations.

To gain more information, a field programme of measuring suspended particulate and dustfall was undertaken in the immediate vicinity of the tailings areas. Suspended particulate was collected with high-volume samplers and particulate deposition was collected in dustfall jars. One hi-vol sampler and one dustfall jar constituted a sampling station.

Wind data for the region showed that although no wind direction predominates, north and west winds were expected to occur most frequently. Consequently, upwind-downwind sampling was used in assessing the tailings dust emissions. In addition to the hi-vol samplers, micro-meteorological stations were located directly on the tailings areas to obtain data on conditions which affect the wind generation of the dust.

This approach was modified to account for topographic constraints, access to sites and the proximity of interferring sources such as roads and construction. Within these general siting criteria, the precise locations for the sampling stations were chosen in areas generally clear of dense bush with a visible line of sight between the tailings and the sampling station. If locations were in close proximity to roads, calcium chloride solution was used to limit road dust generation.

There were very large day-to-day changes in suspended particulate levels next to the tailings areas. The majority of the readings were less than 60 $\mu g/m^3$ with an occasional value above 1000 $\mu g/m^3$ and a number of values below 20 $\mu g/m^3$. A summary of the results is presented in Table 1.

Further interpretation of the data confirmed some beliefs on the affect of wind, rain and snow-cover on the degree of dust generated. Figure 2 illustrates the results from a portion of the 1977 summer particulate

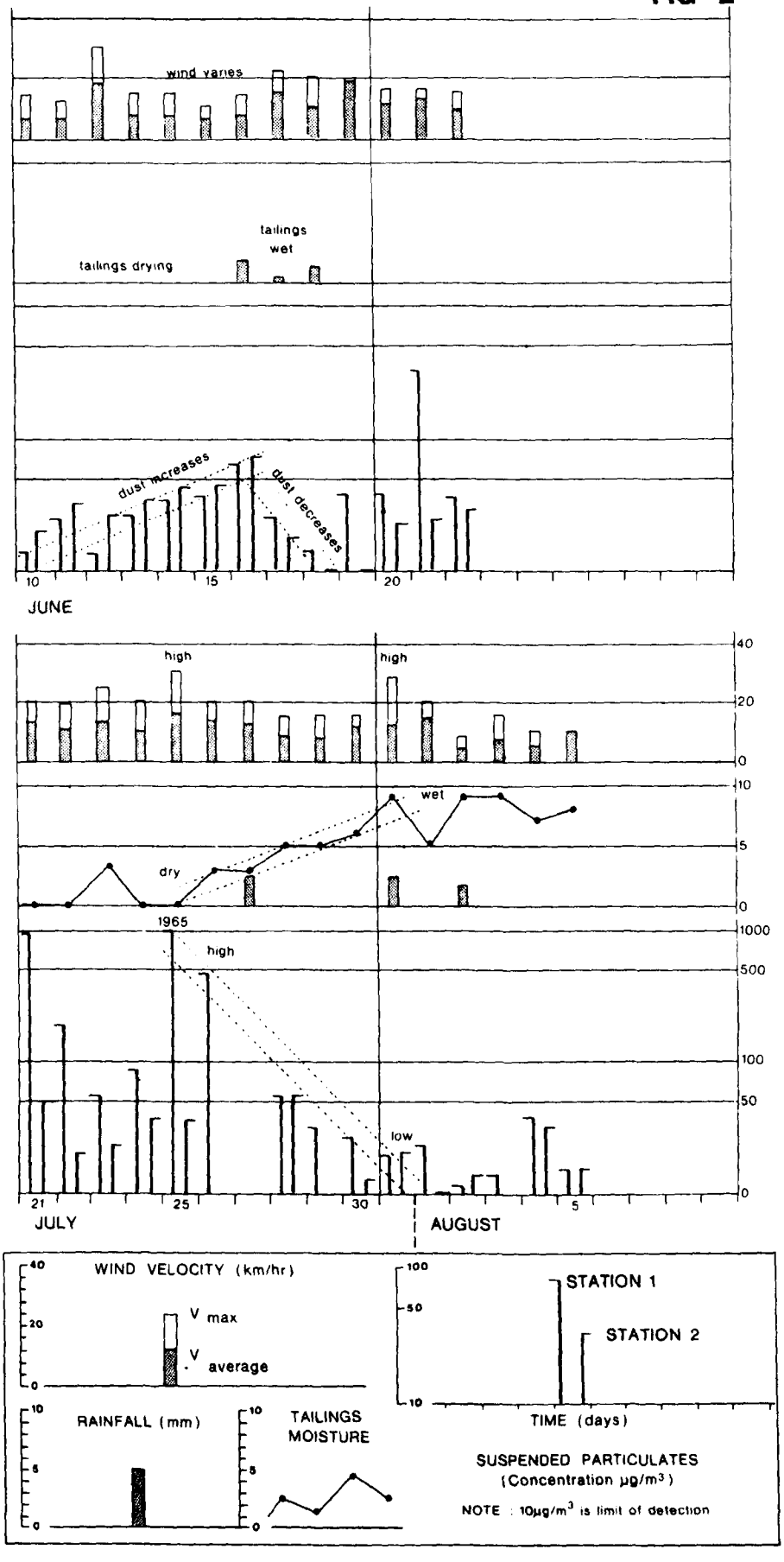
TABLE 1

SUMMARY OF TAILINGS SUSPENDED PARTICULATE SURVEY

Tailings Monitoring Site	Number of Readings	Distance To Dry Tailings (m)	Average of Readings ($\mu g/m^3$)	Geometric Mean of Readings ($\mu g/m^3$)	Highest Reading ($\mu g/m^3$)	Lowest Reading ($\mu g/m^3$)	% of Readings Above 120 $\mu g/m^3$ (%)	% of Readings Above 400 $\mu g/m^3$ (%)
Stanrock 1	31	100	115	45	1051	14	16	6
2	30	100	91	55	575	9	17	7
3	29	250	22	19	60	6	0	0
Long Lake 1	13	60	44	26	240	4	8	0
2	13	60	15	13	49	6	0	0
3	15	1000	38	19	262	1	7	0
Nordic 1	73	100	94	32	1965	0	10	5
Lab	66	200	24	20	65	4	0	0
Crotch Lake 1	38	20	54	27	341	4	13	0
2	35	20	43	22	300	4	9	0
3	9	60	15	13	34	5	0	0
Panel 1	5	10	10	7	24	1	0	0
2	2	20	44	30	75	12	0	0
Quirke 1	48	120	43	33	147	7	2	0
2	28	30	48	23	441	1	7	4
3	8	1000	67	43	276	16	13	0
Sheriff Lake	61	1000	21	18	48	5	0	0

EFFECTS OF WIND AND RAIN ON SUSPENDED PARTICULATE

FIG 2



survey selected to demonstrate the effects of tailings moisture content and windspeed on suspended particulate levels. Between June 12 and June 16, the tailings were drying out following rain and the suspended dust levels rose quite steadily day by day from 20 to 70 $\mu g/m^3$ at Station 1 and from 13 to 62 $\mu g/m^3$ at Station 2. During this time wind speeds averaged about 6 km/h (4 mi./h). The rainfall on June 16, 17 and 18 caused dust levels to drop to 0 at Station 1 and 13 and Station 2 by June 18, even though the wind speeds over this period were higher.

The role of high wind speeds is clearly shown by the July 25 data. On July 25, average winds of 18 km/h (11 mi./h) with gusts to 30 km/h (19 mi./h) combined with dry tailings resulted in the highest dust level recorded at Station 2. However, on July 31, comparable wind gusts and only a slightly lower average wind speed showed low dust levels due to an increase in tailings moisture content.

The principle survey was conducted during the summer of 1977 and a lesser continuing programme is still underway. Annual data, Table 2, indicates the effects of seasonal climatic changes on airborne dust released from tailings areas. The geometric means and highest values recorded during the winter months are very low. During the spring period, the values increase somewhat, but it is obvious from the data that summer conditions result in higher values. Clearly, the tailings are not prone to becoming airborne during the winter months, and are less likely to drift in the spring than summer.

Sufficient data was available from two active tailings areas to perform a upwind-downwind analysis. The results do confirm the belief that tailings dust is carried downwind and will result in elevated suspended particulate levels immediately adjacent to the tailings area. They do not, however, identify the effect of tailings fugitive emissions on the areas of concern, the townsites and residential areas.

For this reason, a theoretical model, based on sand physics, was used to predict air emissions, and classical atmospheric dispersion techniques were used to transport and deposit the airborne particulate. The results of the monitoring program were used to calibrate the model.

TABLE 2

SEASONAL SUSPENDED PARTICULATE RESULTS

JUNE 1977 - AUGUST 1977

MONITORING SITE	NUMBER OF READINGS	AVERAGE ($\mu g/m^3$)	GEOMETRIC MEAN ($\mu g/m^3$)	HIGHEST READING ($\mu g/m^3$)
NORDIC 1	73	94	32	1965
NORDIC LAB	66	24	20	65

NOVEMBER 1977 - MARCH 1978

NORDIC 1	45	13	6	81
NORDIC LAB	44	13	10	33

APRIL 1978 - JUNE 1978

NORDIC 1	27	35	21	236
NORDIC LAB	28	31	20	106

The air pollution effect of fugitive emission sources depends on the quantity and drift potential of the dust particles entrained into the air. In addition to large particles which tend to settle out near the source, fine particles can be entrained and will disperse from the source.

Studies on sand movement and desert dunes have shown that the transport of materials similar to tailings is affected by particle size distribution, local wind velocities (including wind gusts), and other variables. Exposed surface area and the physical condition of the tailings also have a pronounced effect on dust entrainment.

As part of the overall programme to expand the data base for parameters which affect tailings emi-sions and provide information for the model exposed tailings areas were examined and a series of tailings particle size analyses were initiated.

Aerial photographs and field observations were combined in an effort to delineate the extent of wet and dry tailings and develop the dry exposed surface areas. Size sampling programmes were undertaken at two non-operating tailings areas and at two operating areas. Samples were taken from tailings areas which differed in terms of texture and appearance, and these samples were aggregated and dried prior to sizing analysis.

A model for dispersion of dust from a uranium mill tailings area was developed by James F. MacLaren Limited for use at the Elliot Lake properties. The model is based on a concept developed by the Oak Ridge National Laboratory and includes particle saltation, suspension and deposition and atmospheric transport using a Gaussian plume model.

The suspension of dust at the source is related to the process of saltation as described by Bagnold and an emplirical estimate of the particles which remain airborne near the tailings surface, referred to as suspension.

The saltation rate equation presented here is influenced by the windspeed, V, and the theshold wind velocity, V_t, required to initiate particle motion.

TABLE 3

TAILINGS PARTICLE SIZE DISTRIBUTIONS

PARTICLE SIZE	TYLER SCREENS FOR SAMPLES	PREDOMINANT EMISSION PROPERTIES	PREDOMINANT DISPERSION PROPERTIES
µm	mesh		
300-150	to 100	CREEP AND SALTATION	DUSTFALL
150-106	100 to 150		
106-80)) 80-75)	150 to 200		
75-44)) 44-38)	200 to 400	SUSPENSION	
<38	<400		SUSPENDED PARTICULATE

$$\text{Saltation Rate, } q = \left(\frac{k}{\ln Z/Z_o}\right)^3 .C. \left(\frac{d}{D}\right)^{1/2} \cdot \rho \left(\frac{v-v_t}{g}\right)^3$$

The material remaining in suspension per unit area, S, has been shown to be direct function of the saltation rate:

$$\text{Suspension, } S = kq$$

where the value of K, the suspension factor, varies from 10^{-5} to 10^{-9} per metre dpending upon the particle size and mass.

The dust emission rate or source strength is the material remaining in suspension per unit area multiplied by the source area:

$$\text{Emission rate, } Q = SA$$

The dispersion and deposition of the suspended materials was based on the "titled plume" method described by Pasquill:

$$\chi\ (x,0,0) = \frac{Q_i}{2\ \pi\ \delta_y \delta_z u} \exp\left(-\frac{(H - v_{ti} \cdot x/u)^2}{2\delta_z^2}\right)$$

This equation assumes that material reaching the ground will not be reflected or entrained. Some percentage of smaller particles will be reflected at the ground and so this equation overestimates deposition and underestimates suspended particulate concentrations.

To ensure that the model predictions resulted in conservative values for both deposition and suspension for small particles separate models were used to calculate suspended particulate and deposition.

The previous equation was used to model deposition and modified for suspended particulate as follows:

$$\chi\ (x,0,0) = \frac{Q_i}{2\ \pi\ \delta_y \delta_z u} \exp\left(-\frac{(H-v_s \cdot x/u)^2}{2\delta_z^2}\right) + \exp\left(\frac{-(H-v_s \cdot x/u)^2}{2\delta_z^2}\right)$$

The principal difficulty in using the model to quantify tailings dust emissions and dispersion is the selection of the suspension factor, K. As noted, K values can range from 10^{-5} to 10^{-9} per metre, which covers a ten thousand fold variation.

Value of 10^{-7} per metre was compared with the arithmetic average values for the tailings hi-vol stations as shown in Table 4. As shown in the table, the ratio of suspended particulate concentrations calculated from the tailings to actual measured total suspended particulate matter (Sm/Ss) ranged from 0.07 to 2.91, with an average ratio of 1.1.

At two stations, gravel pit operations add appreciably to the total particulate loadings. If these stations are not considered, the average ratio is increased to 1.25.

Since the tailings hi-vol stations were all in close proximity to tailings and, except where other known sources are also in proximity, most of the particulate measured is from tailings. However, there will in all cases be some background contribution due to natural and man-made sources. On the assumption that this background would not be less than 10 $\mu g/m^3$ the average ratio (Sm/Ss) is increased to 1.65.

The selection of the K value at 10^{-7} per metre appears to result in good overall agreement between the model and the field results. More detailed calibration did not appear to be reasonable.

Annual contours of suspended particulate and dustfall were predicted by the model to determine the extent of the effect of the emissions and to assess the calibration of the model. The model prediction of the present mining and milling is presented in Figure 3.

A comparison of the suspended particulate model to the air quality, Table 5, results indicates that the model is reasonable. The measured overall air quality at town monitoring stations was approximately 40 $\mu g/m^3$ (arithmetic average) while the tailings area contribution predicted by the model is less, ranging from 8 to 17 $\mu g/m^3$ at locations in the general vicinity of the

TABLE 4

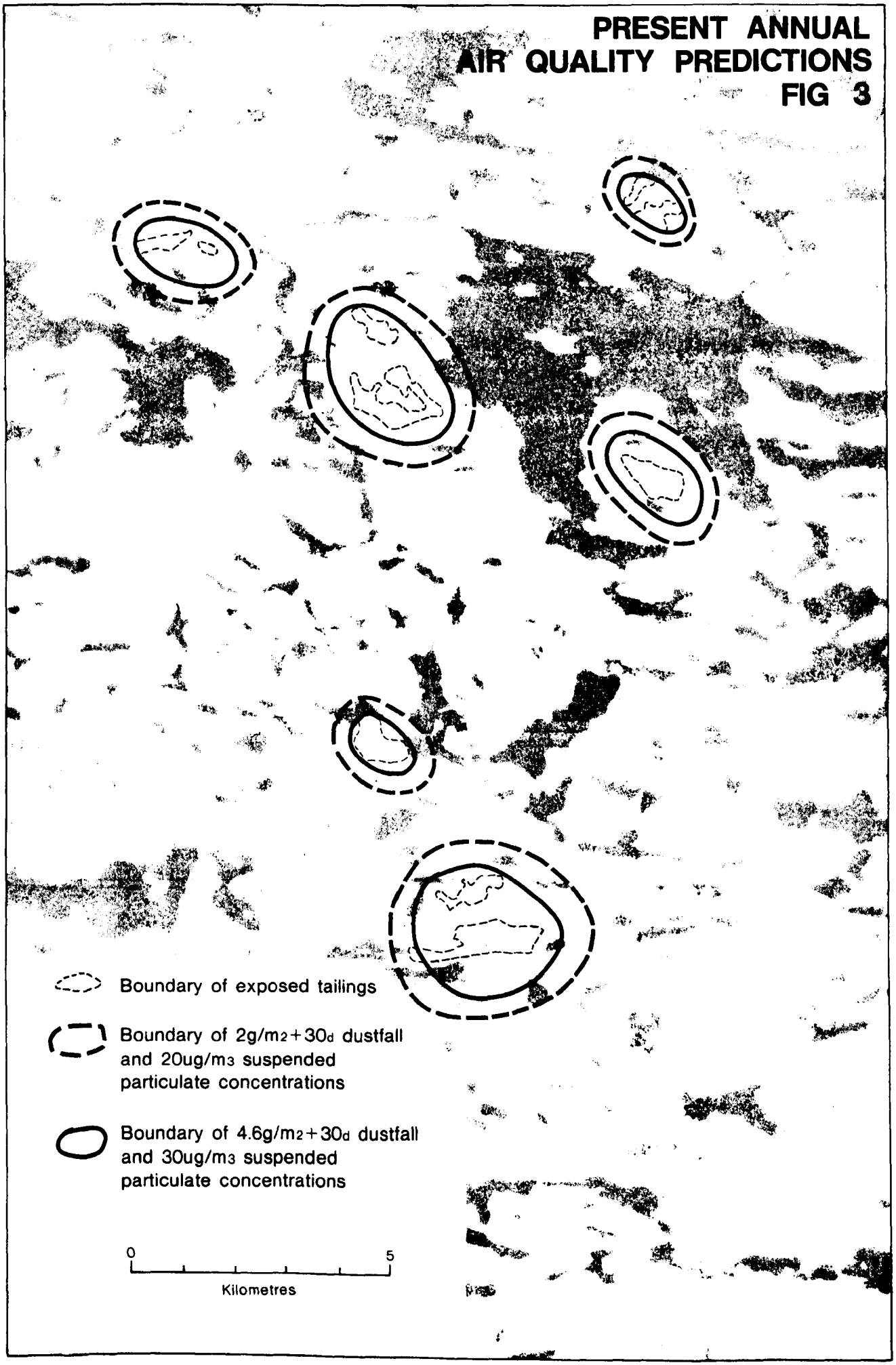
COMPARISON OF MODEL WITH SUSPENDED PARTICULATE FIELD STUDY

MONITORING SITE	SURVEY RESULTS		MODEL		DISTANCE FROM DRY TAILINGS (m)
	Number of Readings	Arithmetic Average (Ss) ($\mu g/m^3$)	Prediction (Sm) ($\mu g/m^3$)	Evaluation Sm/Ss	
STANROCK 1	31	115.0	62	0.54	90
2	30	92.8	58	0.62	120
3	29	22.3	32	1.43	240
NORDIC 1	73	93.9	35	0.37	200
LAB	66	24.3	43	1.77	200
SHERIFF LAKE	51	20.7	10	0.42	1000
CROTCH LAKE 1	38	53.8	67	1.25	20
2	35	43.1	79	1.83	20
3	9	15.2	38	2.50	50
PANEL 1	5*	(10.4)	55		20
2	2*	(43.5)	37		20
QUIRKE 1	48	42.4	33	0.78	120
2	28	48.2	25	0.52	30
3	8	67.0	5	0.07 (1)	1000
LONG LAKE 1	13	44.3	59	1.33	20
2	13	14.8	43	2.91	50
3	15	38.4	9	0.23 (1)	1000
AVERAGE OF ALL STATIONS				1.1	

*Not enough readings for meaningful results

(1) Stations were near other known dust sources

PRESENT ANNUAL AIR QUALITY PREDICTIONS FIG 3



tailings and 4 $\mu g/m^3$ in the more removed town of Elliot Lake. Background contributions include other fugitive sources such as gravel pits, unpaved roads, and construction operation which also have variable emission characteristics. From the results of the particulate survey, these could make up 20 $\pm$ 10 $\mu g/m^3$ at any one location at any one time.

If the model predictions were increased by a factor of two, they would approach the measured air quality and the amount left for other background sources would be unreasonably low.

The same approach can be applied to the dustfall results, Table 6, and the same conclusion reached.

As previously mentioned, the uranium mining industry on Elliot Lake is undergoing an expension and the effect of the increase in tailings areas is of concern. The ultimate use of the model in using its ability to predict trends and extent of the tailings dust on air quality was to determine the future air quality. Overall air quality patterns only changed slightly with the isopleths around the sources expanding, Figure 4.

Specific results at the critical areas of the townsites indicates that the expansion of tailings will not have a significant effect on the existing air quality, Table 7.

TABLE 5

COMPARISON OF TAILINGS SUSPENDED PARTICULATE MODEL TO AIR QUALITY RESULTS

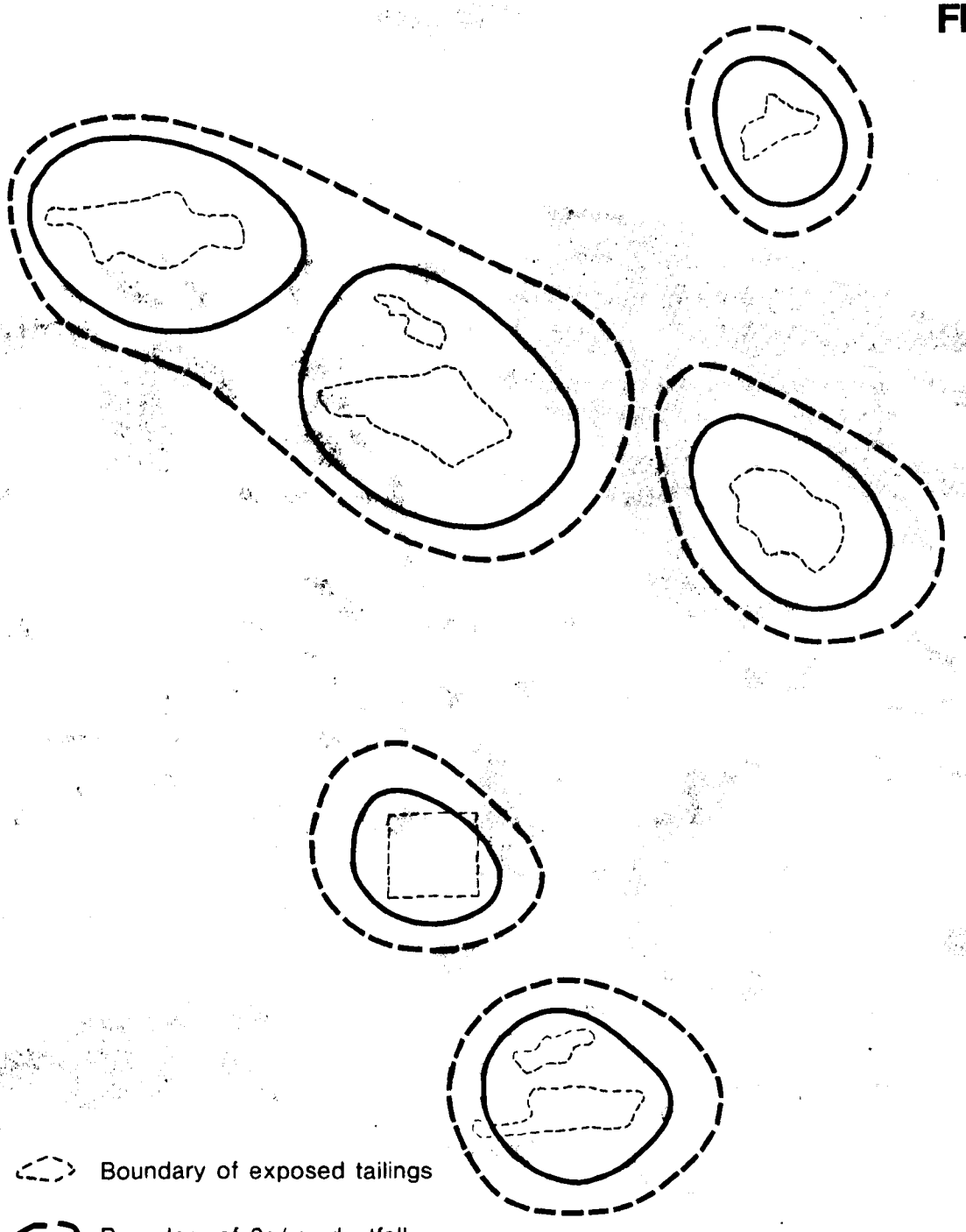
MONITORING STATION	FIELD SURVEY RESULTS					MODEL RESULTS (TAILINGS ONLY)	
	Number of 24 h Measurements	Highest Reading $\mu g/m^3$	Lowest Reading $\mu g/m^3$	Geometric Mean $\mu g/m^3$	Arithmetic Average (Xs) $\mu g/m^3$	Annual Average (Xm) $\mu g/m^3$	Xm/Xs x 100
Elliot Lake	55	136	5	35	42	3.5	10
Nordic Townsite	29	121	6	30	39	17	45
Denison Townsite	27	108	1	30	40	13	35
Quirke Townsite	29	85	6	32	38	8	20

TABLE 6

COMPARISON OF TAILINGS DEPOSITION MODEL TO AIR QUALITY RESULTS

STATION	SURVEY RESULTS Number of 30 d Measurements	Highest Reading g/m^2 30 d	Lowest Reading g/m^2 30 d	Arithmetic Average (Ds) g/m^2 30 d	MODEL RESULTS (TAILINGS ONLY) Monthly Average (Dm) g/m^2 30 d	Dm/Ds x 100
Hillside Drive	11	7.5	0.8	3.8		
Kilborn Way	10	6.5	1.0	3.6	0.2	10%
Roman Avenue	8	17.7	0.5	4.1		
Denison Townsite	11	24.1	0.9	8.4	1.8	20%
Quirke Townsite	11	4.3	0.4	2.2	0.9	40%

FUTURE ANNUAL AIR QUALITY PREDICTIONS FIG 4



Boundary of exposed tailings

Boundary of 2g/m2 dustfall and 20 ug/m3 suspended particulate concentrations

Boundary oh4.6g/m2+390d d dustfall and 30ug/m3 suspended particulate concentrations

0 5

Kilometres

TABLE 7

COMPARISON OF EXPANSION CASE AIR QUALITY ESTIMATES

IN RESIDENTIAL AREAS WITH PRESENT MEASUREMENTS

Station	SUSPENDED PARTICULATE ($\mu g/m^3$) (geometric mean)		DUSTFALL (g/m^2. 30 d)	
	Existing	Expansion Case Approximation	Existing	Expansion Case Approximation
ELLIOT LAKE				
Hillside Drive	35	37	3.8	3.9
Kilborn Way			3.6	3.6
Roman Avenue			4.1	4.2
DENISON TOWNSITE	30	45	8.4	9.6
QUIRKE TOWNSITE	32	44	2.2	3.7
CRITERION	60	60	7	7

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IDENTIFYING SOURCES AND QUANTITIES OF FUGITIVE EMISSIONS IN BALTIMORE

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ABSTRACT

Violations of Federal primary standards for total suspended particulate (TSP) matter have been observed in the Baltimore area for the past several years and, based on modeling results, are expected to continue to occur through 1985. A study was undertaken to estimate fugitive emissions from nonstandard sources and to analyze hi-vol samples to determine the types of sources which are contributing to the concentrations which are presently exceeding standards. The results of a survey of fugitive source characteristics in the designated TSP nonattainment area in the metropolitan Baltimore area were used to estimate fugitive dust emissions. Emissions from gravel and dirt roads were found to account for 74 percent of fugitive emissions. Fugitive emissions in the nonattaintment area were estimated to be at an annual average rate of 10,000 lb/hr. A labortory analysis of 18 hi-vol filters based on optical microscopy and a battery of additional chemical and physical tests were used to characterize the filter sample into six source categories. The analysis was made on half-filters and was repeated for three samples. The source type attributions for sources consisting of over 15 percent of the sample were reproduced within 12 percent of the original estimate. The hi-vol filter analysis showed that 50 percent of the sample materials are associated with fugitive sources. Possible fugitive emission control strategies were identified based on these results which will reduce fugitive emissions by 70 percent. At locations with the highest measurements, the emissions controls may reduce TSP concentrations by 50 percent. Future study of the air quality impact of possible control alternatives is recommended.

INTRODUCTION

A study of suspended particulate matter in a selected portion of the Metropolitan Baltimore area has characterized the types of particles which contribute to exceedances of the National Ambient Air Quality Standards. A survey of the nonattainment area was made to identify fugitive sources and estimate emissions. Potential control techniques for reducing fugitive emissions were identified.

The total suspended particulate (TSP) air quality problem in the Baltimore area is shown by the data in Tables 1 and 2. Monitoring stations which exceeded Federal primary TSP standards in 1975 or 1976 are listed for annual geometric means in Table 1 and for second highest 24-hour concentrations in Table 2. The locations of these monitoring sites are shown in Figure 1.

Table 1. Stations Exceeding Annual Primary TSP Standard* in the Metropolitan Baltimore AQCR in 1975 or 1976

Station	Annual Geometric Mean (g/m3)	
	1975	1976
Fire Department HQ	86	74
Fire Department #10	128	164
Ft. McHenry	86	105
Fire Department #22	86	82
Patapsco STP	150	144
S.E. Police Station	78	77
AAI	(not operated)	90

* Annual Standard = 75 g/m^3 geometric mean.

Table 2. Stations Exceeding 24-Hour Primary TSP Standard* in the Metropolitan Baltimore AQCR in 1975 or 1976

Station	Second Highest TSP Concentration ($\mu g/m_3$)	
	1975	1976
Riviera Beach	357	174
Fire Department #10	358	559
Patapsco STP	398	509
Lansdowne High School	101	271

* Primary Standard = 250 $\mu g/m^3$ geometric mean.

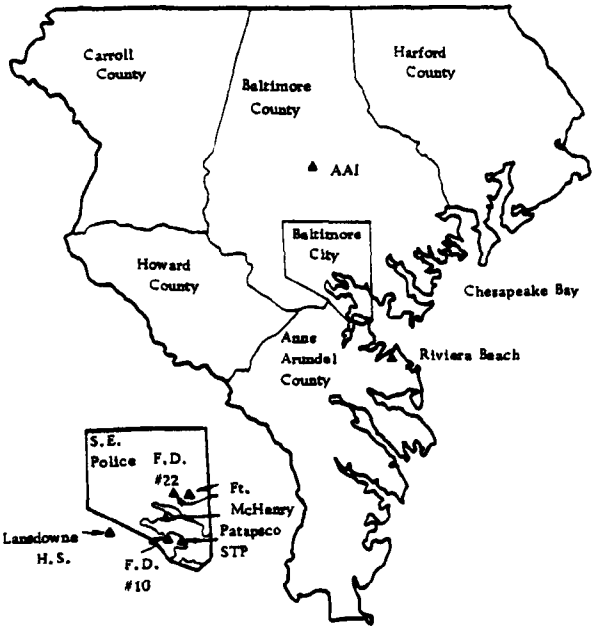

Figure 1. Location of TSP monitoring stations exceeding primary NAAQS in 1975 and 1976.

A nonattainment area for TSP was designated for Metropolitan Baltimore as shown in Figure 2. This study was conducted to determine the causes of the high concentrations of TSP in this area and to suggest alternative methods of controlling the contributions to the high concentrations from fugitive emissions, which were suspected to be a major cause of the exceedances.

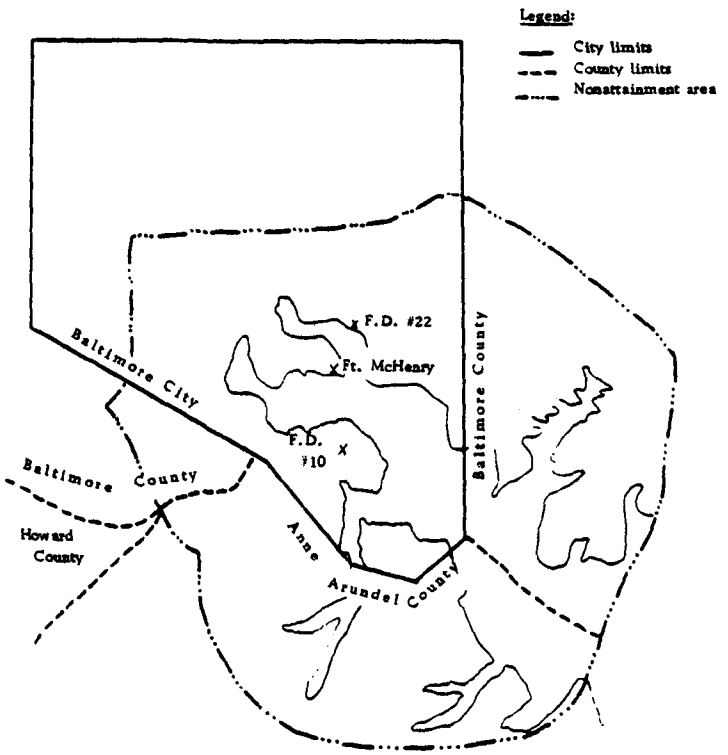

Figure 2. Metropolitan Baltimore nonattainment area for TSP, including the three hi-vol stations used in the project.

The Maryland Bureau of Air Quality and Noise Control (MBAQNC) collected 24-hour hi-vol samples at three sites in the nonattainment area (see Figure 2) during the 7-month period from June to December 1977. Eighteen filters were selected for physical and chemical analysis to determine generic types of particles collected. The particle types, their relative mass on each filter, and their frequency of occurrence at each site and for different days helped determine significant sources or classes of sources.

A ground-area, "windshield" survey was performed by MBAQNC on the location and characteristics of fugitive sources of TSP which were not included in the state emission inventory. Types of surface material, e.g., grass, asphalt, dirt and gravel, were identified along with quantitative factors affecting emission rates, including size, amount of vehicle activity, and types of industrial activity. The survey results and other data relating to mobile sources (i.e., cars, trucks and trains, etc.) were used to estimate fugitive emissions in each square mile of the city. The estimates will be used to update the state emission inventory.

A review of control strategies found in the literature was made to recommend methods to implement for each of six types of fugitive particulate sources, including paved roads, dirt and gravel roads, construction and waste disposal sites, wind erosion, railroads and storage piles.

FUGITIVE EMISSION ESTIMATES

The data collected by MBAQNC in its survey of fugitive emission sources covered an area of 83 square miles within the City limits of Baltimore and 284 squares measuring 1,000 feet on a side in parts of the surrounding Baltimore and Anne Arundel Counties. An inspector visited each area and characterized each open lot or other area by the type of surface, its use (e.g., parking, recreation, construction, unused, railroad, storage, etc.), the type of terrain (e.g., open, enclosed, steep slopes, etc.), the types of emissions (e.g., vehicles, stockpiles, material handlers, natural occurrences, etc.) and location identification data. Special characteristics, reflecting mobile activities were also noted, including the number of various vehicles, speed of vehicles, and frequency of stockpile turnover.

The Maryland emission inventory was updated to include fugitive particulate emissions in two ways: (1) by adding fugitive emissions from new sources identified in the survey, and (2) by adding fugitive emissions from sources in the present inventory. Both the amount of fugitive particulate emissions and information on particle sizes were included in the inventory data. Fugitive emission factors applicable to each source were developed from the available literature and from data obtained by direct contact with investigators whose work had not been published. The factors were adapted to the meteorological conditions, the materials and the types of activities found in the Baltimore area.

Paved Roads

The following emission factor for paved roads was derived from work done primarily by Bohn and Cowherd (1978):

$$EF = 0.45\ (s/10)(L/5000)(W/3) \qquad (1)$$

where EF = emission factor (lb/vehicle mile traveled)

s = silt content of road surface material (%)

W = average vehicle weight (tons)

L = surface dust loading on traveled portion of road (lb/mile).

There was not sufficient information available to assign detailed spatial and temporal variations to the parameters in this relationship. Values estimated to be representative for Baltimore were selected. Street runoff measurements by Sartor and Boyd (1972) were used to estimate that 12 percent is a reasonable estimate of the silt content. The average weight of vehicles was estimated to be 1.5 tons corresponding primarily to light-duty vehicles. The dust loading of streets was assumed to be 17.7 lb/mile based on informal discussions with investigators in EPA Region III and Midwest Research Institute. The resulting emission factor for Baltimore paved roads is 0.43 g(9.6×10^{-4} lb) per vehicle mile traveled (VMT).

Unpaved Roads

Emission factors for unpaved roads, including both gravel and dirt roads, were derived from the EPA publication AP42 (1977). The annual number of days per year with precipitation greater than or equal to 0.01 inches was found to be 112 days for Baltimore. The silt content of gravel roads was estimated to be 12 percent. For dirt roads, information from local sources was used to estimate the silt content. The following relationship was used to determine emission factors:

$$EF = 0.6(0.81)s(S/30)(1 - (P/365)) \qquad (2)$$

where EF = emission factor (lb/VMT)

s = silt content (%)

S = average vehicle speed, mph

P = days per year with 0.01 inch or more precipitation or with reported snow cover.

To apply equation (2) it is necessary to know the number of vehicle miles traveled, the average vehicle speed, and (for dirt roads) the silt content of the roadway. If a better estimate was not available, vehicle speeds were assumed to be 25 miles per hour. When applied to parking lot traffic, vehicle speeds were assumed to be 10 miles per hour. The silt content of dirt roads was estimated for each quadrant of the city and found to vary from 62 to 75 percent.

In order to be compatible with other emission factors used in this study, the EPA equation was multiplied by 0.6 to estimate the fraction of the total particulate emissions with aerodynamic diameters less than 30 μm.

Construction Sites

An emission factor for construction sites was developed from work by Cowherd and others (1974). Emissions of 1.2 tons/acre/month (269 g/m^2/month) are representative of average construction activity and are assumed to represent solid waste disposal sites which use similar types of operations. Variations in activity levels may result in emissions varying by a factor of two over a period of a month. Activity levels are difficult to estimate without precise information on the nature and intensity of construction or disposal operations. Since observations from the southwest United States were used to develop the emission factor, it may be high for east coast areas.

Wind Erosion

For fugitive emissions due to wind erosion, the following emission factor was used based on an analysis by Bohn and Cowherd (1978):

$$EF = 381\ (e/50)\ (s/15)\ (f/25)\ (PE/50)^{-2}$$

where EF = emission factor, $g/m^2/yr$

e = surface erodibility, ton/acre/yr

s = silt content of surface material, %

f = frequency that wind exeeds 12 mph, %

PE = Thornthwaite's precipitation-evaporation index.

For the Baltimore area, the wind exceeds 12 miles per hour 21 percent of the time and the Thornthwaite precipitation-evaporation index is 108. For surfaces common in the Baltimore area, the surface erodibility varies from 40 to 200 tons/acre/year. For most clay soils the factor is 85 tons/acre/year. A list of soil erodibility factors for various soil textural classes was compiled by Cowherd et al. (1974).

Railroads

An emission factor for railroad traffic was developed to treat the large amount of railroad volume much of which connects with the Baltimore Harbor. Based on observations of rail traffic, conversations with railyard managers and considerations of emission factors for other mobile sources, the emission factor was set at 10 percent of the emission rate for unpaved roads. The silt content of railroad loads was estimated at 10 percent and railroad car speeds were estimated to be 15 miles per hour in the city limits and 40 miles per hour outside the city limits. Railroad data were used to estimate the railroad car miles traveled in city and county areas.

Storage Piles

An emission factor for aggregate storage piles developed from work by Cowherd et al. (1974) is as follows:

$$EF = \frac{1650}{(PE)^2}$$

where EF = emission factor, g/kg (placed in storage)

PE = Thornthwaite's precipitation-evaporation index.

To apply this factor, it is necessary to know the amount of material moved into the storage pile over the course of a year. This emission factor was applied in the Baltimore area to large piles with a material throughput on the order of 100,000 tons per year or more.

Summary of Fugitive Emissions

There are many uncertainties associated with the emission factors. Based on a review of the variability reflected in the data used to develop the factors, considering the uncertainty of other parameter estimates used to apply the factors, and considering the uncertainty of the applicability of the factors to the Baltimore area, it is estimated that over the course of the year actual fugitive emissions will be within a factor of 2 of the calculated fugitive emissions. On any given day, the emissions may be off by a greater amount. The validity of the emission factors for determining maximum 24-hour concentrations for verifying compliance with National Ambient Air Quality Standards needs further study. A summary of the emission factors used for the Baltimore area is presented in Table 3.

Table 3. Summary of Fugitive Emission Factors and Required Source Data

Type of Source	Emission Factor	Required Source Characteristics
Paved road	0.43 (g/VMT) 0.96×10^{-3} (lb/VMT)	VMT (vehicle miles traveled)
Gravel road	61 S (g/VMT) 0.13 S (lb/VMT)	VMT S (vehicle speed, mph)
Dirt road	5.1 sS (g/VMT) 0.0112 sS (lb/VMT)	VMT S s (silt content of road material, %)
Construction sites	9.0 (g/m^2/day) 79 (lb/acre/day)	Site area
Wind erosion	2.6×10^{-4} e s (g/m^2/day) 2.3×10^{-3} e s (lb/acre/day)	Area s e (surface erodibility, ton/acre/year)
Railroads	6.1 S (g/VMT) 0.013 S (lb/VMT)	VMT S
Storage piles	0.14 (g/kg) 0.28 (lb/ton)	Quantity placed in storage

When the emission factors and the required source data are applied to the designated nonattainment area of Metropolitan Baltimore, the fugitive emissions presented in Table 4 are obtained for the portion within the city limits, the portion outside the city limits and the total for the nonattainment area. From these results, it can be seen that, of the seven nontraditional sources, dirt roads, gravel roads and construction operations contribute the greatest amount of fugitive particulate emissions. The emissions for unpaved roads (dirt and gravel) amount to 74 percent of the total. This only includes material kicked up from the pavement and does not include tire wear or tailpipe emissions.

Table 4. Baltimore TSP Nonattainment Area Fugitive Particulates (Tons/Day)

Type	Within City	Outside City	Total
Paved roads	-	-	-
Gravel roads	26	12	38
Dirt roads	45	5	50
Construction	10	9	19
Wind erosion	2	4	6
Railroads	-	-	-
Storage piles	4	2	6
Total	87	32	119

HI-VOL FILTER ANALYSIS

The State of Maryland collected hi-vol filters at 3 sites in the non-attainment area where noncompliance with Federal standards had been reported. A total of 18 filters collected over a period of 7 months between June and December 1977 were selected for microscopy and chemical analysis. The location of the sampling sites is shown in Figure 2. The selected filters represent different wind directions, wind speeds, and particulate loadings as shown in Table 5. On 3 days filters were selected from all 3 sites; on another 3 days filters were obtained from 2 sites; and the remaining 3 filters were selected on 3 separate days. The wind speeds were slower than normal on the selected days, varying from 2 to 6.5 as a daily average, comapred to the climatological average for Baltimore of 10 miles per hour.

Microscopy and chemical analysis were performed on the selected filters by IIT Research Institute. One-half of each selected filter was submitted for analysis. At the conclusion of the study, three additional halves, selected at random from the original 18, were submittted for duplicate analysis.

The microscopy was performed by removing the particles from a piece of the filter half and viewing the particles optically under a high-powered microscope which is illuminated by polarized light. The optical properties observed to classify particles by different types of sources include transparency, color, refractive index, birefringence, reflectance, pleochroism and fluorescence. The physical properties observed include size, shape, surface texture, magnetism, solubility, melting point and density. To identify particle types, the microscopist relies on reference collections of particle data which include atlases of photomicrographs, handbooks of optical properties, the microscopist's previous experience and actual source samples.

Table 5. Hi-Vol Filters Selected for Analysis

Date	Day of Week	Measured Concentrations (µg/m)			Rainfall (in)		Wind Speed (mph)	Prevailing Wind Direction
		Fort McHenry	Fire Dept.#10	Fire Dept.#22	Day of Sample	Day before Sample		
6/9	Th	59	63	-	1-36	0	6.2	S to SSW
6/21	T	136*	145*	60	0	0.36	6.3	WNW
7/15	F	114*	221	149	0	0	2.0	ESE to SE
8/2	T	154	142*	93*	0	0.03	2.8	WNW
9/19	M	69*	165*	108	0	0	3.5	SW
9/25	S	39	51	38	T	T	6.5	ENE
10/25	T	118	306	-	0	0	3.2	E
11/24	Th	41	42	52	0.01	0.31	4.3	W
12/12	M	85	169	129	T	0	4.6	S

* Sample not analyzed.

A simple particle counting and sizing procedure is used to determine the weight percentages of specific particle types present in each hi-vol sample. A mass per unit area of the filter is determined for each type of identified particle. The sizing and counting procedure was used to calculate mass concentrations of silicate, calcite, hematite, rubber tire fragments, flyash, coal and cornstarch. Other types of particulates (e.g., ammonium sulfate, auto exhaust, pollen and magnetic fragments) were determined by nonmicroscopical procedures, including low temperature plasma ashing, plasma emission spectroscopy, scanning electron microscopy and sulfate analysis.

Low-temperature ashing determined the concentration of combustible matter such as starch, pollens, and carbonaceous vehicle exhausts. It generally divided the samples between organic and inorganic materials. A chemical analysis procedure was used to identify the sulfate concentration of the filter. Lead and vanadium concentrations were analyzed by plasma emission spectroscopy to determine the concentrations of auto exhaust and oil soot particles in the sample. The auto exhaust content is taken as 1.5 times the lead concentration, and oil soot is taken as 39 times the vanadium content. The scanning electron microscope was used to estimate the concentrations of 18 elements. The results were helpful in substantiating estimates obtained by other methods. Unfortunately, the variation in elemental content of the filters themselves made the estimates of doubtful value as an independent estimate.

The analysis of hi-vol vilters revealed that six types of particles accounted for most of the material (from 70 to 96 percent) collected on 18 filters. The percentages of each type, including silicates, sulfates, rubber, calcite, cornstarch and slag are shown in Table 6. A summary of the partciles classified by six types of sources using results from both microscopy and other analyses is presented in Table 7. The mineral category

Table 6. Predominant Materials Found on Hi-Vol Filters

Station	6/9/77		6/21		7/15		8/2		9/19		9/25		10/25		11/24		12/12	
FD #10	Si	41%			Si	40%					Su	36%	Si	31%	Su	35%	Ca	40%
	Su	21			Ca	30					Si	35	Ca	25	Si	32	Si	30
	Ru	12			Su	13					Ru	11	Si	17	Ru	11	Su	13
	Ca	13			Ru	7					Ca	10	Su	13	Total	78	Ru	8
	Total	87			Total	90					Total	92	Total	96			Total	91
Ft. McHenry	Si	44%					Co	41%			Su	50%	Si	44%	Su	45%	Si	33%
	Su	30					Si	23			Si	24	Su	18	Si	32	Su	25
	Co	11					Su	15			Total	74	Co	8	Ru	14	Ru	12
	Total	85					Total	79					Total	70	Total	91	Ca	14
																	Total	84
FD #22			Si	52%	Si	54%			Si	45%	Su	40%			Si	35%	Si	37%
			Su	24	Su	16			Su	22	Si	37			Su	32	Su	17
			Ru	14	Ru	9			Ru	11	Ru	12			Ru	24	Ca	14
			Total	90	Total	79			Total	78	Total	89			Total	91	Ru	10
																	Total	78

Note: Si = silicates
Su = sulfates
Ru = rubber
Ca = calcite
Co = cornstarch
Sl = slag

Includes all material with % by weight $\geq$ 10 only.

Table 7. Summary of Particles Classified by Source Type

Date	Site	TSP Concentration ($\mu g/m^3$)	Low Temperature Ashing (% Removed)	Source Type					
				Minerals %	Mobile Emissions %	Combustion Sources %	Non-Specific Combustion %	Corn Starch %	Biologicals %
6/9/77	FD #10	63	41%	57	13	3	23	2	2
	Ft. Mc	59	51%	49	3	4	31	12	<0.5
6/21/77	FD #22	60	44%	54	15	3	24	1	4
7/15/77	FD #10	221	29%	71	8	4	16	1	1
	FD #22	149	40%	58	10	7	20	4	1
8/2/77	Ft. Mc	154	65%	30	5	2	16	43	1
9/19/77	FD #22	108	42%	56	12	6	24	3	1
9/25/77	FD #10	51	39%	46	12	5	36	<0.5	<0.5
	FD #22	38	50%	39	13	5	40	2	2
	Ft. Mc	39	51%	29	7	10	50	2	1
10/25/77	FD #10	306	23%	44	4	4	16	31	<0.5
	Ft. Mc	118	48%	50	8	9	23	9	<0.5
11/24/77	FD #10	42	45%	40	13	8	37	3	<0.5
	FD #22	52	47%	40	26	3	33	<0.5	<0.5
	Ft. Mc	41	60%	35	16	4	47	1	<0.5
12/12/77	FD #10	169	31%	70	9	2	19	1	<0.5
	FD #22	129	40%	56	11	4	22	3	<0.5
	Ft. Mc	85	51%	48	13	7	28	5	<0.5

includes both soil and road surfaces. Every sample contains a high percentage of mineral content, but the percentage content is generally highest when the total particle loading (as indicated by the TSP concentration) is highest.

As a verification of the validity of the microscopy and supporting analyses, 3 filter halves which were matches to the original 18 were submitted for analysis. The comparative results are shown in Table 8. The mean difference between independent analyses of the same filter divided by the mean of all measurements of a given particulate component is shown in the last column. This ratio varies from 0 to 0.85 for the 17 components listed with an overall mean of 0.25. These results are very encouraging regarding the reproducibility of the analysis.

In summary, the results in Tables 6 and 7 show that mineral sources, such as roadway kickup, constuction operations and wind erosion, are the major contributors to TSP loadings in the Baltimore nonattainment area. Another important source is nonspecific combustion sources which are primarily evident by the presence of sulfates. Particulates from mobile source tailpipes are generally the third highest component of particulate loadings. A fourth type of particulate, which is generally small, but is occasionally the largest type, is cornstarch.

SUGGESTED CONTROL STRATEGIES

Based on the inventory of fugitive sources of TSP and the analysis of hi-vol filters, it appears that dirt and gravel roads and construction operations are the major local sources which contribute to the highest ambient TSP concentrations. There is also a unique and highly specific component of TSP due to cornstarch, which is identified with a few very specific sources. Controls for cornstarch can be specific to these sources. Alternative control strategies for dirt and gravel roads and an estimate of their effectiveness in reducing emissions were developed from various sources of emission measurements. The following measures are cited as effective, although their costs have not been evaluated:

1. Paving will reduce emissions by 99 percent.
2. Applying water or a chemical stabilizer will reduce emissions by 50 percent.
3. Oiling and a double chip surface will reduce emissions by 85 percent.
4. Reduce vehicle speeds.
5. Decrease multiwheeled truck activity on dirt and gravel roads.

The following suggested control strategies for construction sites have been compiled, but useful estimates of their effectiveness have not been determined:

1. Regularly water exposed soil.
2. Keep handled materials at solid waste disposal sites wet.
3. Cover hauling operations.
4. Reduce the free fall of materials handled.
5. Revegetate waste disposal sites.

Table 8. Results of Duplicate Microscopy Analysis

Component	% by Weight						Mean Difference Divided by Mean
	Set 1		Set 2		Set 3*		
	1st	2nd	1st	2nd	1st	2nd	
Silicates	23	23	47	47			0 (2)**
Calcite	5	13	1	2			0.85 (2)
Mica	<0.5	<0.5	< 0.5	<0.5	< 0.5		0 (2)
Clays, humus	2	1	1	1	2		0.4 (2)
Hematite	2	1	1	0.5			0.67 (2)
Carbonaceous tailpipe exhaust	0.5	0.5	1	1	1	1	0
Rubber tire fragments	5	2	4	4	3		0.4 (2)
Glassy flyash	1	0.5	1	2			0.43 (2)
Coal fragments	0.5	0.5	1	0.4			0.25 (2)
Oil soot	1	1	2	3	1	1	0.22
Fine carbonaceous particles	1	1	< 0.5	< 0.5	2		0 (2)
Recrystallized sulfates	15	9	40	38	13	12.6	0.13
Cornstarch	41	46	< 0.5	< 0.5	<0.5	<0.5	0.11
Pollens, spores, conidia	<0.5	1	1	2	<0.5	<0.5	0.37
Plant parts	<0.5	<0.5	< 0.5	< 0.5	<0.5	<0.5	0
Insect parts	1	<0.5	1	< 0.5	<0.5	<0.5	0.5
Magnetic fragments	<0.5	<0.5	< 0.5	< 0.5	<0.5	<0.5	0
Total	98.0+	99.5+	101.0+	100.9+			0.25 (mean)

*Results are incomplete for this comparison.

**A two in parentheses (2) indicates result is based on two comparisons.

CONCLUSIONS

About 50 percent of the sampled TSP from hi-vols in the Baltimore non-attainment area is mineral matter from fugitive emission sources. Vehicle-generated emissions and, to a lesser extent, wind-generated emissions from dirt and gravel road surfaces are the most likely sources of this material.

Cornstarch from harbor grain transfer operations and nearby processing operations is a significant source within the critical area of high TSP concentrations. Slag was also found to be a significant pollutant on some days. Its source is easily associated with nearby steel production operations.

Sulfates are an important source of TSP, but this component is most likely due to emissions from distant combustion of sulfur-bearing fuels outside of the Metropolitan Baltimore area.

About 70 percent of the fugitive emissions in the Baltimore area can be eliminated by local area controls. This would reduce overall TSP levels by 35 percent in areas where fugitive emissions constitute 50 percent of the total TSP loading. In areas with high TSP loadings, it is likely that fugitive and other locally controllable emissions (e.g., corn starch and slag) constitute over 70 percent of the TSP levels. Therefore, the highest TSP levels can be reduced by more than 50 percent.

A relatively large degree of uncertainty exists concerning fugitive emission factor estimates which are only weakly supported by measurements. Questions concerning how representative the particulates caught on hi-vols are of the particulates in the atmosphere necessarily raise some doubts regarding how representative these samples are of what really makes up ambient TSP matter. The number of measurements used to derive emission factors is small. More data representing many variations in roadway emissions are needed to better estimate the impact of this important source of TSP loading.

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FUGITIVE EMISSIONS FROM THE BY-PRODUCT COKE OVEN PUSHING OPERATION

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ABSTRACT

An intensive sampling program at the by-product coking facility of a major midwestern steel company has quantified the release of trace metals and other particulate to the atmosphere resulting from the "pushing" operation. Emission factors and emission rates were obtained from "clean" (completely coked) and "green" (incompletely coked) pushes. A number of coke oven push plumes were sampled isokinetically at a rate of approximately 30 SCFM. The sampling head with associated temperature and velocity sensors was suspended over the quench car by means of a tower/boom arrangement mounted on a mobile laboratory. The cross sectional area of the plume in the horizontal plane of the sampling was determined by measurements on motion picture films taken from two vantage points. The mass emission rate of total particulate was calculated from the particulate concentration, plume velocity and cross sectional area. Trace metal emissions were quantified by atomic absorption analysis of all particulate samples for Cd, Pb, Zn, and Cu.

INTRODUCTION

A total of 66 coke oven pushing plumes have been sampled using a specially designed mobile laboratory for characterizing the plume parameters of temperature, velocity, shape, and particulate emission. An earlier publication by this author[1] reported on the first 15 samples while this work concentrates on the complete sample set of 66. This paper will be helpful to those involved in the quantification and control of coke oven pushing emissions and the design of fugitive emission sampling techniques.

MEASUREMENT METHODOLOGY

The pushing emission can be classified as a "fugitive emission" of sorts in that it does not emanate from a confined conduit or stack. As such, quantification of the particulate emissions has a "high degree of difficulty" and requires a special experimental design. The basic approach in this study was to place sampling nozzles and sensors for temperature and velocity in a stationary plane in the plume over the duration of the push. Meandering of the plume around the sampling nozzle caused by atmospheric motion reduced the biasing effect of large variations in plume parameters. Figure 1 contains a cross-section view of the sampling apparatus aligned adjacent to the coke oven. Note the mobility of the apparatus and freedom of any supports from the coke oven structure itself. This provided for unrestricted movement of the sampling laboratory and no interference with the coke oven production schedule. The sampling nozzle and instrumentation were at the elevation of the top of the coke guide and positioned approximately over the center of the quench car.

Temperatures were measured with two iron/constant and thermocouples at the end of the boom. A cup anemometer provided plume velocities and particulate samples were taken at a rate of approximately 43 scfm through 8 x 10 inch glass fiber filters.

Cross-sectional plume shape was determined by taking 16 mm black and white motion pictures in two planes over the duration of each push. Subsequent stop-action projector analysis yielded plume cross-sectional dimensions. The camera set-up is shown in Figure 2. Since the motion picture camera frame rate was calibrated, plume velocities could be checked by use of a stop watch. In addition, the gas sample volume was easily corrected for those instances where the sampling nozzle was not in the plume by examination of the film.

The particulate concentration was determined by drawing a gas sample at an approximate rate of 43 scfm through an 8 x 10 inch glass fiber filter. The gas sample volume was determined with an orifice plate and the sampling rate was controlled from within the vehicle to achieve an isokinetic rate as a function of the anemometer readout. The reader is referred to the primary author for specific hardware details. Knowing the weight of coal charged to each furnace, an emission factor was subsequently computed for each push.

RESULTS AND DISCUSSION

Plume Temperature

Fifteen of the 66 pushes sampled were taken in December and the remaining in April. Since the steel mill was located in Northwest Indiana, ambient temperatures were significantly different in each of these seasons. The December ambient temperatures were close to $0^{o}F$ and the April temperatures were close to $40^{o}F$. This difference in temperature is partially reflected in average plume temperatures seen in Table 1.

TABLE 1
Coke Oven Push Plume Temperatures ^{o}F

	No. of Samples	Average	Range	One Standard Deviation
DECEMBER				
Green	7	157	81-309	77
Clean	8	89	77-108	11
Overall	15	121	77-309	62
APRIL				
Green	33	232	109-534	101
Clean	18	117	71-167	25
Overall	51	191	71-534	99

Note from Table 1 that the green push plume temperatures are about twice that of the clean pushes regardless of the season. This is a result of the incomplete combustion of the remaining volatiles in the green push coke and the attendant flames. Also, the range of plume temperatures is quite wide from 71 F to 534 F as is probably expected but it is interesting to note that the clean pushes have a significantly lower standard deviation value as compared to green pushes. The December and April standard deviations were 11 and 25 F for the clean pushes respectively and 77 and 101 for the green pushes. Apparently, the clean push plume temperature is more closely related to the coke temperature which is relatively constant from push to push as compared to a green push whose temperature is probably more closely related to the amount of flame in the plume which can vary greatly from one green push to another.

The temperature-time history characteristic curve would be of interest to those designing capture hoods or other control devices. Figure 3 contains a plot of the three temperatures recorded over the 66 pushes sampled. Note that the maximum temperature was 534 F and corresponds to a green push. The average rate of increase in temperature for the three temperature curves in Figure 3 was $4^{o}F$/sec with the maximum occurring about 40% of the total time into the push.

Plume Velocity

The buoyancy of the push plumes result in a vertical velocity which was measured with a cup anemometer whose cups were located in a vertical plane. Table 2 contains the average statistics.

TABLE 2
Push Plume Velocity, ft/sec

	No. of Samples	Plume Average	Velocity Range	One Standard Deviation
Green	40	15.7	6.8-21.3	3.9
Clean	26	14.1	6.2-20.9	3.3
Overall	66	15.0	6.2-21.3	3.7

From Table 2 it is seen that the average and range of plume velocities are not greatly different for the clean or green pushes. Note also that one standard deviation about the average value is not exceptionally large and reflects the relatively consistent plume velocity from push to push. The overall range, however, is quite wide from 6.2 to 21.3 ft/sec; a 3.5 to 1 change.

Figure 4 shows a typical green push velocity-time history plot. Note the erratic nature of the trace reflecting the billowing nature of the plume. Also, the peak velocities occur about 15 seconds in the push, are sustained for 15 seconds and decay in about one-half the time it took to reach peak velocity.

Plume Cross-Sectional Shape

Analysis of the two motion picture film records which were shot at approximately 90^{0} to one another (Refer once again to Figure 2) allowed the plume cross-sectional shape to be defined. The procedure involved the use of a stop action projector and measuring the plume width frame-by-frame for each 90^{0} view. Table 3 shows the resulting plume dimensions.

TABLE 3
Plume Cross-Sectional Shape

	Plume Shape, ft. Average	Range	One Standard Deviation
Length "A"	18.2	10-29	3.8
Length "B"	17.8	10-31	5.6
Overall	18.0	10-31	4.7
Ratio of "A"/"B"	1.02	1.0-0.94	0.3

Note that the two diameters are very nearly equal with a ratio of 1.02. This indicates that a circular plume cross-sectional area is a reasonable model to use for estimation of the emission parameters.

From Tables 2 and 3 the plume volumetric flow rate can be estimated on an average basis. Table 4 shows the plume actual volumetric flow rate as calculated from the statistics in Tables 2 and 3.

TABLE 4
Plume Actual Volumetric Flow Rate

	ft^3/min
Average	229,000
One Standard Deviation about the average	94,000-454;000
Observed overall range	61,000-920,000

Table 4 indicates the average volumetric flow rate to be 229,000 actual ft^3/min. Using one standard deviation value to both reduce and increase the plume diameter and velocity a one standard deviation range on volumetric flow rate was calculated. This range as seen in Table 4 is 94,000 to 454,000 actual ft^3/min. The observed minimum and maximum values are also shown in Table 4.

These volumetric flow rates are of interest to those involved in the design of control hardware and yield an estimate of gas flow rate that must be handled. However, keep in mind that these measurements were made in the open atmosphere where gaseous and particulate diffusion is relatively unlimited. If the control hardware confines the push plume, significantly lower plume volumes will probably result. One reason for this is the restriction on plume diffusion and dilution. For green pushes, a confined duct limits the amount of oxygen available to the remaining coal volatiles thus inhibiting combustion and therefore temperatures which results in lower volumetric flow rates.

Particulate Emissions

The concentration of particulates measured in the push plume is seen in Table 5.

TABLE 5
Push Plume Total Particulate Concentration*

	No. of Samples	Range $grams/m^3$	Average $grams/m^3$	One Standard Deviation $grams/m^3$
Green	39	0.22-16.0	3.3	2.9
Clean	25	0.07-5.0	1.8	1.2
Overall	64	0.07-16.0	2.7	2.5

*per standard conditions of 70 F, 1 atm

As expected, the green pushes have a significantly higher particulate concentration relative to a clean push. The green pushes were 3.3 $grams/m^3$ as compared to 1.8 for clean. Note the large standard deviation for both the green and clean pushes. The coefficient of variation (average ÷ std. dev. x 100) for green and clean pushes is 88% and 67% respectively.

Emission rates and factors of total particulates are shown in Table 6. The emission factors are based on steel mill records of coal charged for each push sampled and indicated an average oven charge of 30,000 lb-coal.

TABLE 6
Summary of Mass Emission Rates and
Emission Factors for Total Particulates

	No. of Samples	Range	Average	One Standard Deviation
Mass Emission Rate (grams/sec)				
Green	39	21-1495	382	335
Clean	25	11-377	138	79
Overall	64	11-1495	287	291
Emission Factor (lb/ton-coal)				
Green	39	0.09-9.0	2.0	1.9
Clean	25	0.05-2.0	0.7	0.4
Overall	64	0.05-9.0	1.5	1.6

In an earlier paper[1], the average mass emission rate of the first 15 December samples was reported. The values were 407 grams/sec for green pushes and 147 grams/sec for clean pushes. It is interesting to note that these values compare favorably with those reported for the total sample set of 64. In other words, the initial 15 samples when reported were reasonably representative of the larger sample mean composed of many more samples.

The emission factors expressed as lb-part./ton-coal charged range from 0.05 to 9.0, a change of 180 to 1. This is a relatively wide variation and is reflected in the large standard deviation values. The resulting "coefficient of variation" for green and clean pushes is 95% and 57% respectively. This indicates the clean push emission factors are not quite as variable as the green pushes and reflects the degree of flame and remaining volatiles in green push.

The average emission factors are of interest and were 0.7 lb/ton for the clean pushes and 2.0 lb/ton for the green pushes. The overall value was 1.5 lb/ton. The emission factor reported in "AP-42" for total particulates[2] for coke "discharging" is 0.6 lb-part./ton-coal charged.

The distinguishing feature of the emission factors is the wide variability. However, whether or not one will experience a green or clean push appears to be directly related to the individual coke oven. In this sampling program, the same ovens were repeatedly sampled. Upon analysis of the data it appeared that a specific oven would either yield a clean push or a green push. Table 7 contains a listing of the ovens that were sampled three or more times and how many of the samples were classified clean or green.

TABLE 7
Characterization of Coke Push Plumes by Individual Ovens

Oven Number	No. of Times Sampled	No. of Green Plumes	No. of Clean Plumes
49	5	0	5
52	3	0	3
54	4	1	3
57	5	4	1
59	5	5	0
62	4	4	0
64	4	0	4
67	3	3	0
69	5	5	0
72	4	4	0

It is interesting to note that ovens #49, 52, 54 and 64 were sampled 16 times and all were clean pushes except one. This contrasts to the record of ovens #57, 59, 62, 67, 69 and 72 which were sampled a total of 26 times. Of these, all were green pushes except one. These results suggest that a clean or green push is a function of the oven from which it came. It may be related to the condition of the internal heat transfer surfaces and to the time between rebuilds. It should be stated that no correlation whatsoever was found between the coking times and the particulate emission factors.

SUMMARY AND CONCLUSIONS

For the 66 coke pushing plumes sampled the overall average parameters observed were:

1)	temperature	176 F
2)	velocity	15 ft/sec
3)	shape	circular w/ratio front to side = 1.02
4)	volumetric flow rate	229,000 ACFM
5)	particulate concentration	2.7 grams/m^3
6)	particulate emission rate	287 grams/sec
7)	particulate emission factor	1.5 lb/ton coal charged

Specific significant differences were measured for green and clean pushes. Generally, green pushes were greater in all of the above parameters by at least 2 to 1 except the velocity which was approximately the same. The one dominant facit regarding all of the pushes sampled was the wide variability in plume parameters not only between green and clean but within each of these categories. One standard deviation values expressed as a percentage of the mean value approached 100% in some cases.

The data suggested that a green or clean push is a strong function of the oven from which it came. Repeated sampling of the same ovens showed a trend that some ovens produce consistently clean pushes while others yield green pushes. Oven maintenance and condition may be the important factor here.

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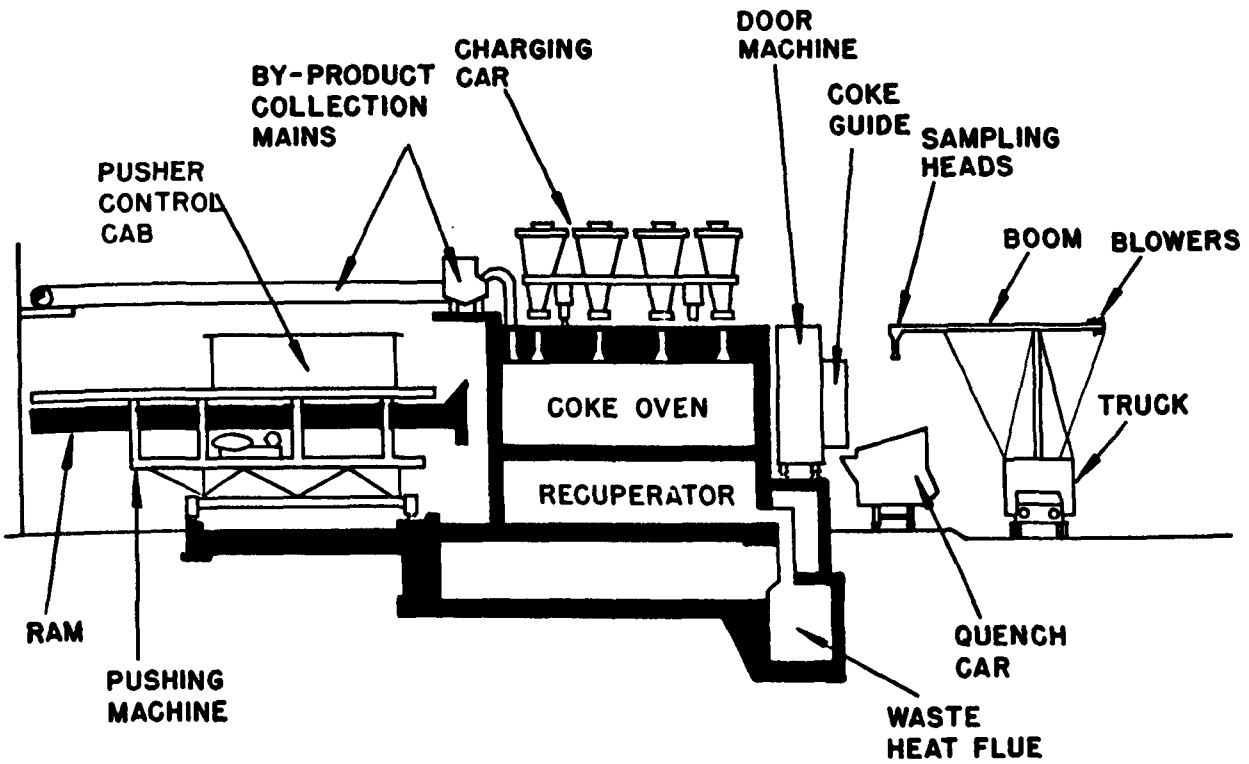

Figure 1 VIEW OF COKE OVEN AND SAMPLING VEHICLE PLACEMENT

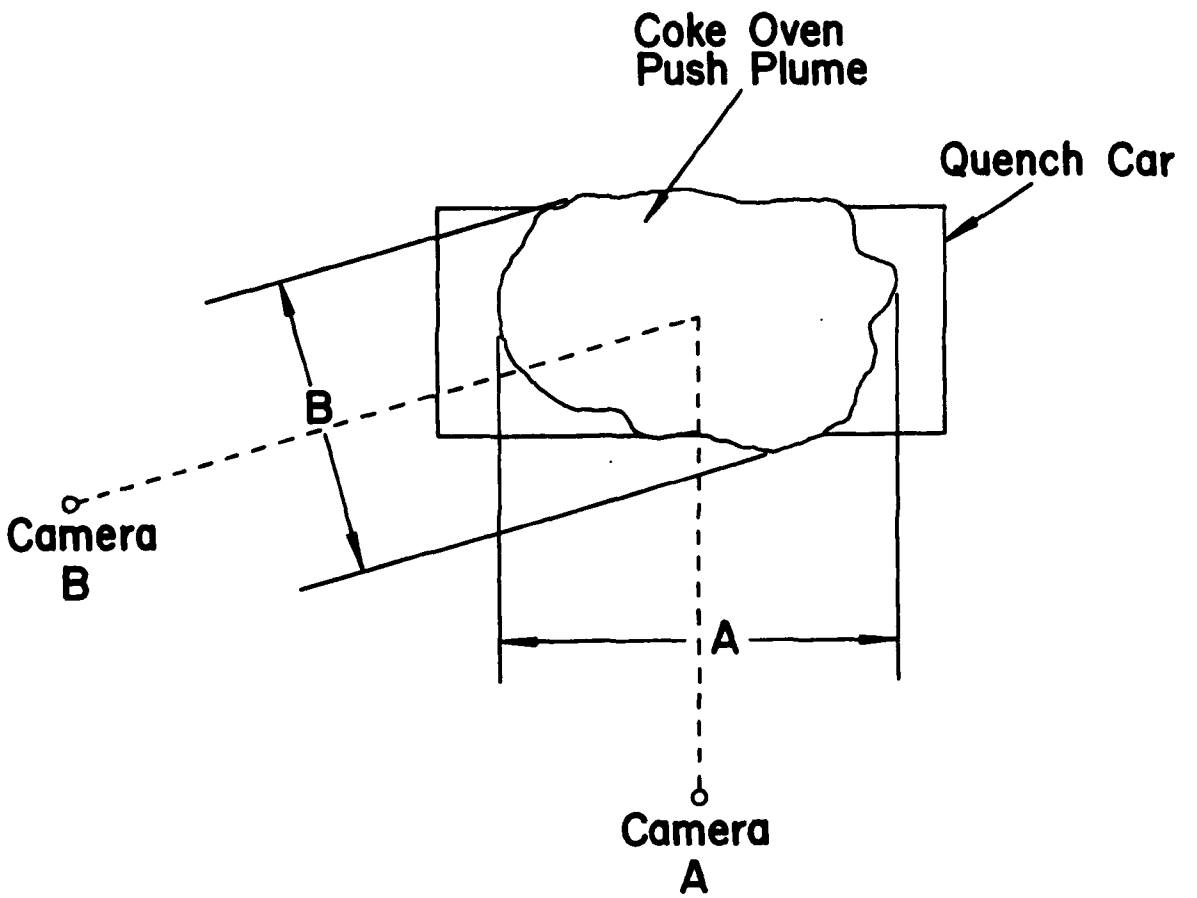

Figure 2 TOP VIEW OF PUSH PLUME MOTION PICTURE CAMERA SETUP

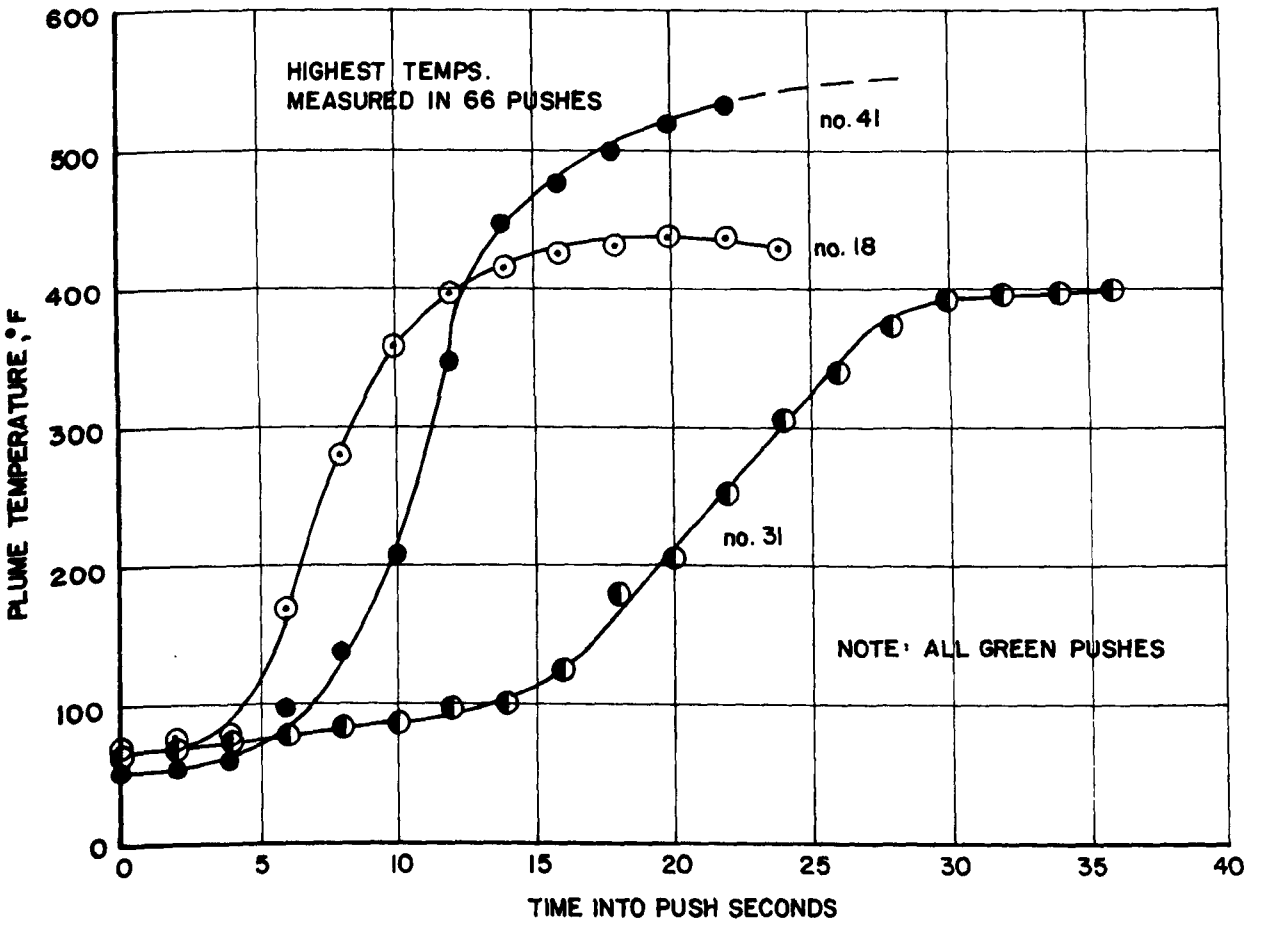

Figure 3 COKE OVEN PUSH PLUME TEMPERATURE - TIME HISTORIES FOR THE 3 HIGHEST TEMPERATURES MEASURED

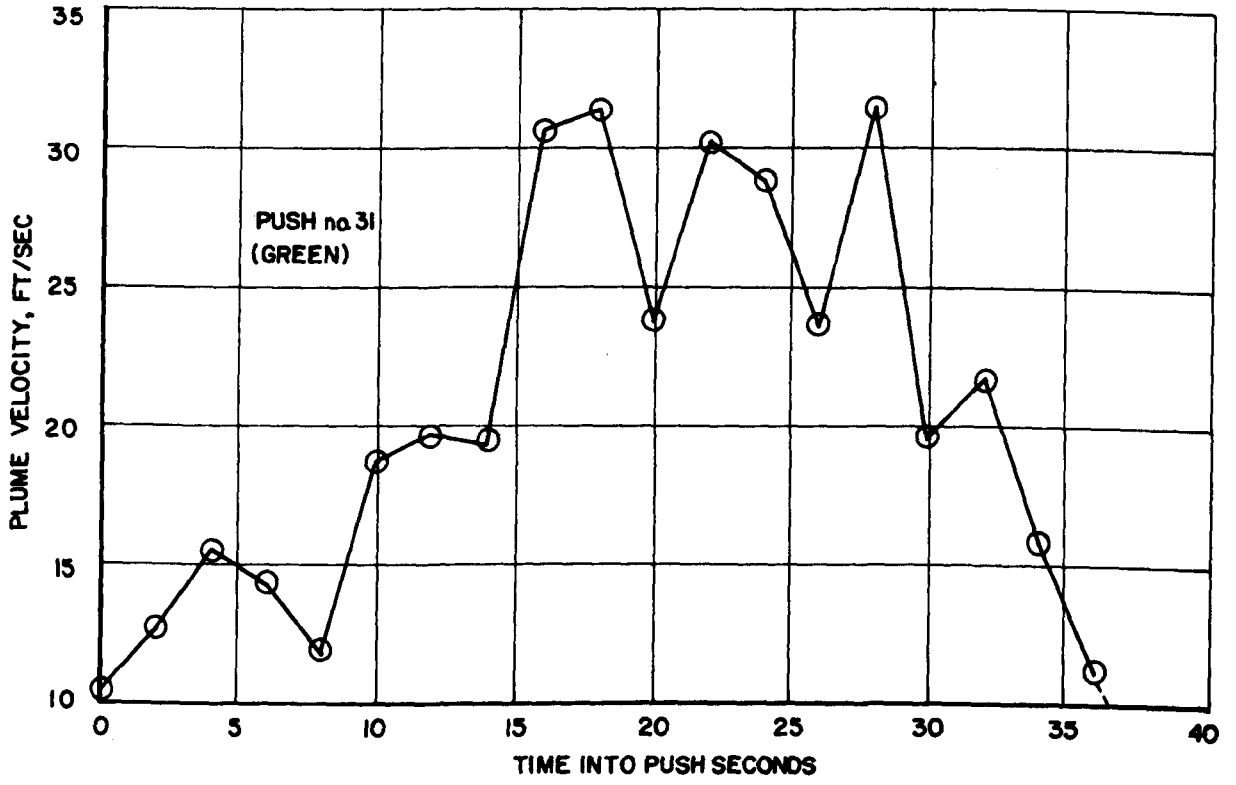

Figure 4 COKE OVEN PUSH PLUME VELOCITY-
TIME HISTORY FOR A TYPICAL GREEN PUSH

ESTIMATING DUST PRODUCTION FROM SURFACE MINING

by

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ABSTRACT

The Federal Land Manager has a responsibility to evaluate the effects of activities he conducts, or permits to be conducted under his administrative jurisdiction, on the air resources of the United States. The operation of surface mining is among these activities. Regulatory classification of surface mines suggests the need for studies of the emission of particulates from them. In particular, two aspects of the emission must receive attention; namely, the chemical/physical characteristics of the particulates, in order to determine what portion of the total particulate burden is "fugitive dust" and what portion is industrial pollution, and secondly, the emissions resulting from specific mining activities such as crushing, screening, dragline overburden removal, stockpiles, etc., must be determined.

This paper describes a preliminary study designed to characterize these emission factors for surface coal mines in and around Craig, Colorado. The study includes consideration of such off-site impacts as deposition downwind of the site, and effects on visibility. The approach includes a general characterization of source configuration and the dispersion pattern, which should lead to an improvement in dispersion modeling for surface mining. Modeling approaches used to calculate downwind concentrations and surface deposition will be reviewed, leading to suggestion of a preferred model. Finally, the main data collection phase of this study will be described. The result will be an improved set of emission factors for surface mining. Future plans to investigate the effects of BACT will also be described.

ESTIMATING DUST PRODUCTION FROM SURFACE MINING

I. INTRODUCTION

Wind blowing over the surface of the earth often generates enough shear stress to mobilize particles in the first few centimeters of the soil. Especially during drought, the amount of material introduced into the atmosphere by wind can be extremely large. The magnitude of the effect depends strongly upon soil type, vegetative cover, and soil moisture content. Natural surfaces, particularly in arid and semiarid regions, develop a "shear-proof" layer that resists wind erosion. Any activity that disturbs the "shear-proof" layer will then cause an increase in blowing dust.

Besides surface mining itself, construction and use of roads, blasting, and overburden removal all increase dust emissions from the surface. How increased mining in the semiarid regions of the Western United States can be conducted with minimal effect on the atmospheric environment is particularly important now that the Clean Air Act Amendments of 1977 require that certain federal lands in the Western United States be managed so that virtually no air quality degradation occurs there.

Recent regulatory proposals from the Environmental Protection Agency (Fed. Reg. 43, 118, 19 June 1978, p. 20395) clarify the application of the concept of PSD (prevention of significant deterioration) to surface mining and discuss the question of fugitive dust. These regulations affirm that surface mining is a stationary source subject to PSD review, but that fugitive dust (defined as particles of native soil uncontaminated by pollutants from industrial activity) will be excluded from such consideration. This exclusion will be reviewed periodically by EPA pending more information, and when new sources are planned, "...the burden of showing to what extent emissions from the proposed source or modification would be made up of fugitive dust rests with the applicant". Fugitive dust is not excluded from estimates of potential emissions (a test to determine if PSD review is needed). Any mine that emits more than 250 tons of particulates per year, including fugitive dust, must apply BACT (Best Available Control Technology).

This regulatory classification underlines the need for studies of particulate emission from surface mines. Two aspects of the emission must receive particular attention, the chemical/physical characteristics of the particulates (to determine what portion of the total particulate burden is "fugitive dust" and what portion is industrial pollution) and emissions resulting from specific mining activities (such as crushing, screening, dragline overburden removal, and stockpiles). The effectiveness of BACT must also be evaluated.

The Bureau of Land Management (BLM) is responsible for evaluating the impact of mining its leased minerals. To evaluate air pollution from surface coal mining in the area around Craig, Colo., the BLM is supporting a cooperative research effort between the Rocky Mountain Forest and Range Experiment Station and Colorado State University.

The major activity in the study will be field measurement to determine type, size, and chemical separation of particulates at various locations downwind from the mine source. Meteorological conditions will also be measured so that modeling can be used to relate the particulate measurements back to emissions and the amount of activity. The objective will be to determine emission factors (i.e. emissions as a function of incremental activities) for surface mining.

To characterize the impact of surface mining on the environment, we will conduct two additional activities. One will establish a deposition monitoring site near Craig to provide information about wet and dry removal of pollutants from the atmosphere. This site will be a component of NC-141, USDA Regional Atmospheric Deposition Project, which will be collecting similar data throughout the United States. Data collected at Craig can be evaluated relative to the entire country.

The second, somewhat speculative activity is related to visibility. Visibility has been a major issue in public concern about air quality for many years. The impact of surface mining on visibility is largely unknown. The assumption that most of the particulates generated from mining are large enough to cause scattering of visible radiation will be tested in a study of background visibility in Northwestern Colorado.

II. CURRENT STATE OF KNOWLEDGE

The latest numbers for emission factors from surface mining, taken from a recent EPA study (EPA-908/1-78-003, Survey of Fugitive Dust From Coal Mines, by PEDCo Environmental, Inc.) are shown in Table 1. While these numbers give some indication of the particulate loading from mining activities, they do not segregate soil from coal particulates, nor are the data sufficient to determine site-specific emissions numbers accurately. Some shortcomings of the numbers in Table 1 are that there is no consideration of such factors as soil surface conditions, or moisture content, and that the numbers are based upon application of a Gaussian model to the entire source area.

Recent work has characterized the background dust from the semiarid Southwestern United States (Gillette et al. 1978). Although the soils are somewhat different, climate conditions in the Craig vicinity are similar, and one would expect roughly similar atmospheric loading.

The results of this work that are most applicable to our study concern particle size distributions. Only small particles (less than 10 μm) are found at altitudes above 1 km, and they are associated with major dust storms. Sampling at ground level (1 m) showed a dust size distribution similar to the aloft aerosol along with a large concentration of particles in the 40-80 μm diameter range. Clearly, the fine aerosols are well-mixed through the lower troposphere, while the large particles remain near the surface. For this reason, the major long range impact of surface mines is the aerosol produced in the size range below 20 μm. This research also suggests that accurate measurement of dust size distribution at one location above the surface may suffice to characterize the total dust produced by the mining activity.

However, the PEDCo report determined particulate loading by measuring concentrations with high volume samplers on or near (1-3 m above) the ground. Such sampling might overestimate the amount of dust exported from the site, unless a method of simulating the dispersion and deposition were incorporated into the analysis.

Modeling was done in the PEDCo study to relate measured concentrations back to emissions from the source. The model used is the standard Gaussian distribution,

$$\chi = \frac{Q}{u} G(x) = \frac{Q}{\pi u \sigma_y \sigma_z} \exp\left[-\frac{1}{2}\left(\frac{y}{\sigma y}\right)^2 - \frac{1}{2}\left(\frac{z}{\sigma z}\right)^2 \right] \quad (1)$$

where χ = concentration, g/m^3

σ_z = vertical standard deviation of plume concentration distribution, m

σ_y = horizontal (perpendicular to plume) standard deviation, m

u = mean wind speed, m/sec

σ_y and σ_z are determined from the equations

$$\sigma_y = c(x + x_0) \quad (1a)$$

$$\sigma_z = a(x = x_0)^b \quad (1b)$$

where a, b, and c are given in Table 2 for different stabilities, and x_0 is determined by observation of the initial distribution of pollutant mass.

Since it is not appropriate to model dust concentrations without simulating deposition, the PEDCo study used a simplified version of the source depletion method originally developed by Van der Hoven (1968). The source depletion model deals with deposition by reducing the effective source strength. Two other modeling approaches, the tilted plume and a surface depletion model, are available.

Tilted Plume

The tilted plume model is a straightforward combination of the Gaussian diffusion formula with a steady particle mass fall velocity. Assuming that the particles in suspension can be described by a discrete size distribution, the fall velocity for particles in the j compartment is

$$\chi = \sum_j \chi_j \quad \text{and} \quad V_d = \sum_j V_{dj}$$

(given for a limited set of Reynolds numbers, $gd_j^2/18\nu$). The trajectory of the particles is tilted at an angle, α, given by

$$\text{Tan}^{-1} \frac{V_d}{u} \quad \text{i.e.} \quad \alpha_j = \text{Tan}^{-1} \frac{V_{dj}}{u}$$

In order to simulate removal from a plume assumed to be released (or have a maximum plume rise) at height h, then simply replacing z by

$$h - x \text{ Sin } \alpha_j \approx h - x \frac{V_{dj}}{u}$$

so the area source expression becomes

$$\chi = \frac{Q}{\pi u \sigma_y \sigma_z} \exp \left[- \frac{1}{2} \left(\frac{y}{\sigma_y}\right)^2 - \frac{1}{2} \left(\frac{h - xV_{dj}}{\sigma_z} \right) \right]^2 \qquad (2)$$

The deposition flux is given by

$$W_j = V_{dj} \chi \qquad (3)$$

This model assumes a completely absorbtive surface with no provisions for particle "bounce" or resuspension.

Source Depletion

The source depletion model simply accounts for a removal by replacing the emission by an effective emission, Q*, where

$$Q^* = \sum_j Q_j^*$$

in equation (1). Then employing conservation of mass, one can relate the change in effective emissions to the deposition flux

$$\frac{dQ_j^*}{dx} = \int W_j dy = - \int_{-\infty}^{\infty} V_{dj} \chi (x,y,o) \, dy \qquad (4)$$

Since this integral has been evaluated for a number of cases, the depletion factor Q* can be calculated by ratio.

Surface Depletion

A more sophisticated and physically realistic model allows the removal of material directly at the encountering surface. Within the Gaussian approximation, this is accomplished by including negative source distributions along with the positive source. Horst (1977) has developed this model by assuming, for the case of an area source, that deposition will occur at a point (ℓ,m,z_d) which causes a reduction in concentration at (x,y,z) downwind, equal to

$$- \left[V_{dj} \chi (\ell,m,z_d) \, d\ell \, dm \right] \frac{G}{u} (x - \ell, y - m, z) \qquad (5)$$

In order to calculate concentration, one sums the source contribution and the total deposition (integration of equation (5) over the total domain of deposition), i.e.

$$\chi(x,y,z) = \frac{Q}{u} G(x,y,h) - \int_{-\infty}^{\infty} \int_{0}^{x} \frac{V_{dj}}{u} \chi(\ell,m,z_d) G(x-\ell,y-m,z) \, d\ell \, dm \quad (6)$$

This has the advantage of allowing specification of the deposition domain, of differential deposition rates, and similarly, of a resuspension coefficient should such a mechanism be needed.

Cole and McVehil (1977) applied these three models to a surface mine for estimating dustfall rates and particulate concentrations. For the material they were concerned about they estimated that the dj (distribution of particle diameters) was approximately 40% with d > 44 μm, and 20% with d < 20 μm. After comparison with experimental results, the most successful model they employed proved to be an integrated version of equation (6). Their results further indicated that some re-entrainment of already deposited dust was necessary to properly predict dustfall rates. In view of this result, our major effort will be oriented toward properly defining the parameters for equation (6), which will provide the most accurate simulation of reality.

III. FIELD PROGRAM

In order to assess the impact of surface mining on the atmospheric environment, we are planning a field exercise at a surface mine in the vicinity of Craig, Colorado (fig. 1). We plan to initiate both short-term and long-term studies. The short-term studies will include meteorological characterizations of the immediate source area and detailed measurements of the mass of particulates suspended in the atmosphere and deposited on the ground surface. Data will also be collected to determine specific mining activities and amounts of material processed per unit time. Meteorological data will be collected on site, using a tethered balloon system and an array of surface stations. Hopefully the study site will be located near a meteorological data tower that has long-term data that will help in planning the timing of our intensive data collection periods, as well as assessing the general applicability of our results.

Short-term studies will also include a particulate emission study at the Craig, Colorado Utah International Coal Mine. A brief description of the field research plan for this study follows.

Delay in starting operation of the Colorado Ute Yampa power plant has resulted in a large stockpile of coal in open fields south of Craig, Colo. Currently, some of this stockpiled coal is being crushed and sold by Colorado Ute. A study of this stockpile area is proposed. Downwind distribution of fugitive dust around the area will be investigated as a function of wind, atmospheric stability, particle size, material composition, and coal handling. The objective is to determine how far downwind significant amounts of coal and overburden remain airborne under different wind and stability conditions.

Meteorological Measurements

Wind and air temperature measurements aloft are available from a tower near he study site operated by Sterns-Rodgers under contract to Colorado Ute. Three-dimensional wind and turbulence data will be taken using Gill u,v,w anemometers mounted on towers at 200 and 600 m downwind from the aforementioned coal stockpile. Also, a tethered balloon sounding system will be available to provide additional information (wind speed and direction, temperature and humidity) at any elevation up to 600 m above the surface. As timing and logistics permit, a monostatic acoustic sounder will be deployed to measure inversion depth in the area.

Particulate Measurements

Dustfall measurements will be taken, using Petri dish collectors spaced in the grid pattern illustrated in figure 2, at intervals no larger than 320 m. Close to emission sources, collectors will be located at 160 m intervals. Petri dishes will be placed at approximately the canopy foliage maximum elevation (15 cm). They will be exposed long enough to allow collection of an analyzable sample, but hopefully for a short enough period that stockpile activity (crushing, etc.) remains constant. The Petri dishes will be fitted with an artificial turf material to improve their collection efficiency and reduce loss from particle bounce. In order to provide a cross reference, selected vegetation samples will be taken to help establish the validity of the Petri dish collectors. Total mass, size distributions, and, via microscopic observation, percentage of coal/surface soil, will be determined from the dustfall collectors. Stacked filter samplers (Cahill et al. 1976) will be used to measure mass and size distribution of suspended particulates. These stacked filter samplers are sufficiently portable to be operated at various levels on the meteorological measurement towers. Each tower will also contain a standard hi-vol sampler at its base, as well as a precise particle sampler at one tower. The specific configuration of these ambient particle samplers will be rearranged according to tracer studies indicating the general air motions.

The study described will be conducted for 1 or 2 days estimated to provide maximum emissions (strong winds, dry soils) during fall and/or spring of 1979.

Long-term concerns include studying the impacts on atmospheric deposition and visibility. We will establish, in conjunction with the Bureau of Land Management's Craig District Office, a wet/dry deposition sampler as a part of the NC-141 Atmospheric Deposition Project. The NC-141 project is a national effort to collect background data describing the chemistry of deposition in a biologically relevant format. Concerns associated with deposition center mainly on any changes in the basic atmospheric processes. The atmosphere provides most of the nutrients required for ecosystem survival (Fox 1977). The atmospheric supply function must be analyzed and protected, particularly with regard to revegetation.

Visibility represents a major concern for the federal land manager. The program of visibility research and monitoring is only now being developed.

Protection of visibility from deterioration by human caused sources has been established by Congress as a national goal. The majority of existing information points at sulfates and nitrogen oxides as the primary cause of reduced visibility (Latimer and Samuelson 1978) in point source plumes as well as on a regional scale (Trijonis and Yuan 1978).

Clearly, any suspended particulate matter smaller than 3 μm in diameter will result in a reduction of visual range, so it is significant to determine this size fraction loading to the atmosphere as a result of mine activities. We are planning to establish a long-term monitoring site at Craig (figure 1) where visibility can be recorded along with background aerosol. The requirements for visibility monitoring include three parameters: visual range, contrast, and chromaticity. The site will include a nephelometer which measures light scattering at the measuring point, thus giving a local measure of the visual range. It will also include a telephotometer measuring both contrast and color alteration of distant targets (George and Zeller 1973). Hopefully, this will be a long-term study so that visibility data can be compared before and after the power plant goes on line.

IV. DATA ANALYSIS

Since a vast amount of data will be generated by these studies, it is important to address the problems of analysis. The objective, as stated, for the short-term studies will be the determination of improved emission factors for mining activities. Basically, two types of data will be collected from the short-term field studies--meteorological and particulate. The meteorological data will be used to establish the windspeed and direction, and stability class for each particulate event which is measured. The models will be applied to all the particulate data utilizing a family of particle size as appropriate. By processing all the dustfall and ambient concentration data in this fashion, we should be able to improve the emissions factors in Table 1.

The long-term studies will provide data for input into national programs. Atmospheric deposition data will be analyzed through the NC-141 system. Various tabulations and graphical comparisons of data from all the 40-plus sites distributed nationwide will be provided on a routine basis. These data will allow immediate comparison with other sites.

Visibility data may prove to be the most difficult to analyze. Our hope is to develop a data base for use in evaluation of all aspects of visibility. The data will initially be used to determine any inputs on visibility resulting from mining operations. Impacts due to the growth in town of Craig and due to operation of the power plant also will be investigated.

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LIST OF SYMBOLS

χ = concentration, gm/m^3

σ_z = vertical standard deviation of plume concentration, m

σ_y = horizontal (perpendicular to plume) plume concentration, m

u = mean wind speed, m/sec

x,y,z = orthogonal coordinates along wind direction, perpendicular in the horizontal and vertical, respectively.

ℓ,m = replacement variables for coordinates

a,b,c = coefficients (see Table 2, equation 1)

x_0 = a virtual source determined visually as the size of the initial pollutant mass distribution, m

V_d = effective deposition velocity, m/sec

$\simeq gd^2/18\nu$ for the Stokes range of particles

d = particle diameter, m

g = 9.8 m^2/sec = acceleration of gravity

ν = kinematic viscosity of the air = .15 m^2/sec @ 20°C

h = effective plume height, m

W = deposition flux, gm/m^2-sec

Q = source emission, gm/sec

Table 1.--Emission factors for individual coal mining operations

Operation	Units	Mine				
		A N.W. Colo.	B S.W. Wyo.	C S.E. Mont.	D Cent. N.D.	E N.E. Wyo.
Dragline	lb/yd^3	0.0056	0.053	0.0030[b]	0.021	
Haul roads	lb/veh-mi[a]					
w/watering		6.8	13.6	3.3[b]		4.3
no watering			17.0		11.2	
Shovel/Truck loading	lb/ton					
coal		0.014	0.007	0.002[b]		0.0035
overburden						0.37[b]
Blasting	lb/blast					
coal				25.1	78.1	72.4
overburden		1690[b]		14.2		85.3
Truck dump	lb/ton					
bottom dump		0.014	0.020	0.005	0.027	
end dump						0.007
overburden						0.002[b]
Storage pile	lb/acre-hr	1.6 u, where u is in m/sec				
Drilling	lb/hole					
coal						0.22
overburden				1.5		
Fly-ash dump	lb/hr	3.9				
Train loading	lb/ton			0.0002		
Topsoil removal	lb/yd^3					
scraping					0.35	
dumping					0.03	
Front-end loader	lb/ton				0.12	

[a]Only veh-mi by haul trucks; travel by other vehicles on haul roads (pickup trucks, ANFO trucks) is incorporated into these values.

[b]These values were all noted to be somehow atypical and should not be used without first determining the limitations to their applicability described in this paper.

Table 2.--Coefficients for the application of a line source Gaussian model to near source dispersion. These values are only appropriate within 10 km of the source.

Stability class	a	b	c
A	0.183	0.945	0.280
B	0.147	0.932	0.197
C	0.112	0.915	0.132
D	0.0856	0.870	0.086
E	0.0762	0.837	0.065
F	0.0552	0.816	0.042

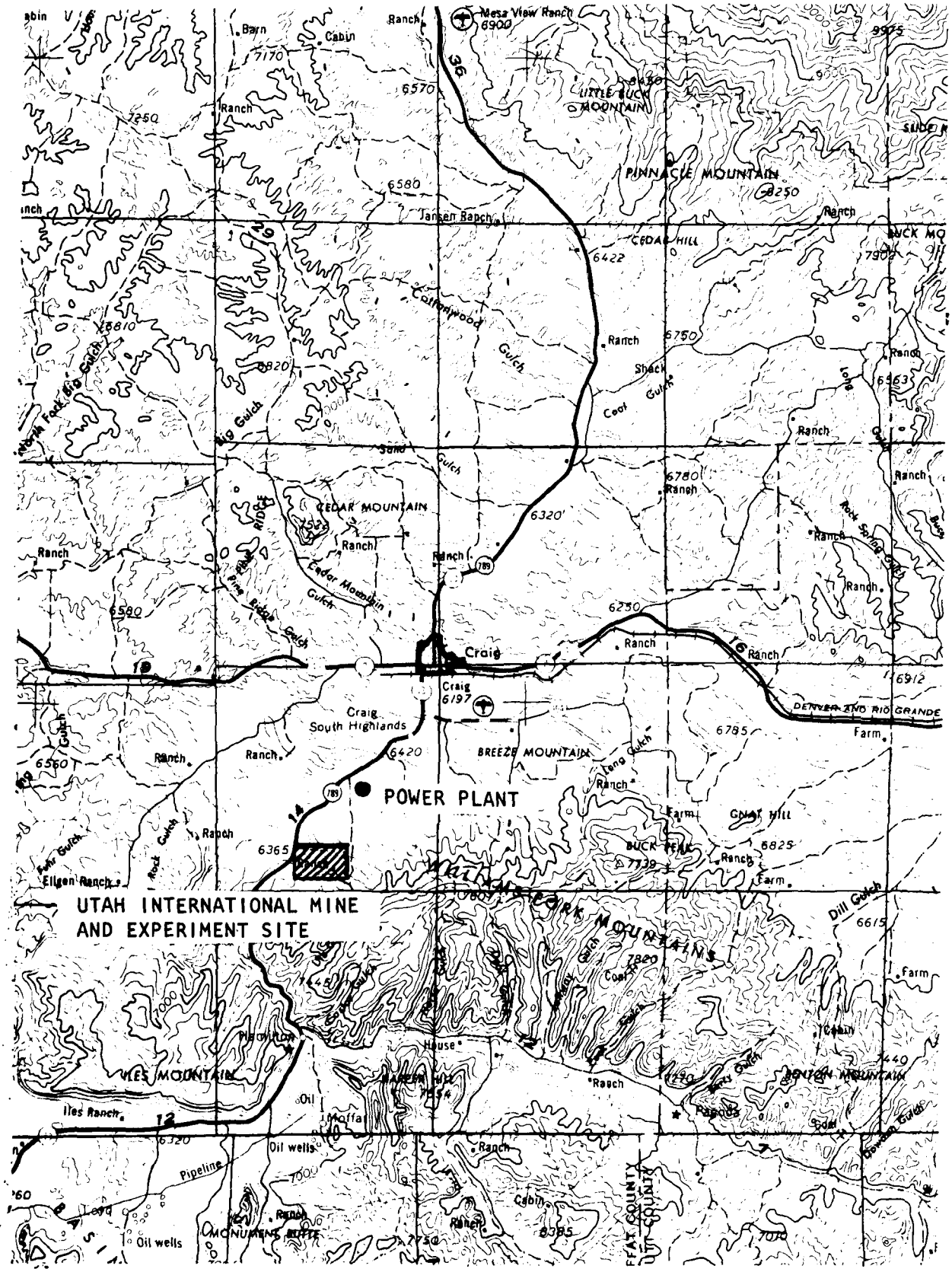

Figure 1.--Map of vicinity of Craig, Colorado. Utah International Mine, Colorado Ute Power Plant, and proposed experimental site are located as shown.

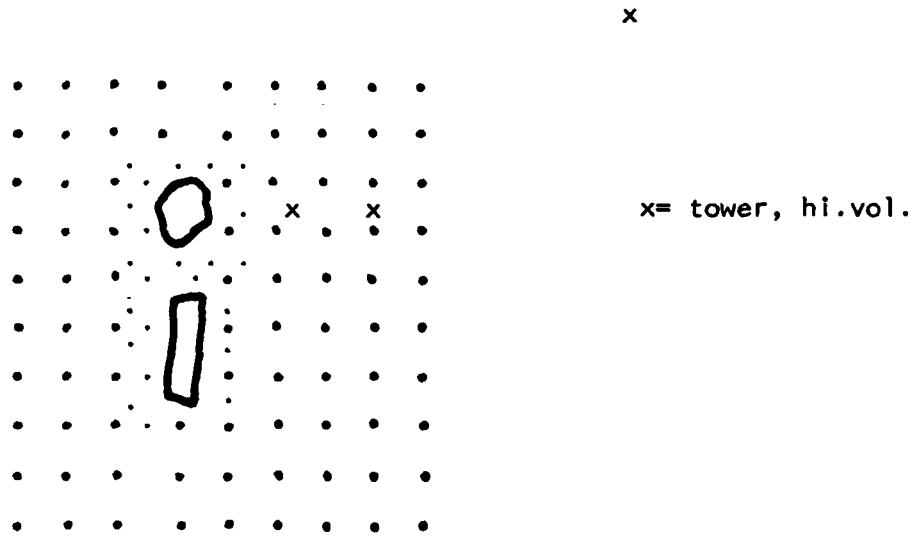

Figure 2.--Approximate location array for samplers around stockpile.

OCTOBER 24, 1978

Tuesday Morning - SESSION III

Session Chairman: Lyle D. Randen
Environmental Engineer
AMAX Coal Company

Tuesday Afternoon - SESSION IV

Session Chairman: James A. Dorsey
Chief, Process Measurements Branch
EPA/IERL-RTP

COMPARISON OF PREDICTED AND OBSERVED EFFECTS OF FUGITIVE DUST FROM COAL OPERATIONS

by

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ABSTRACT

In the vicinity of facilities which prepare, process, or utilize coal, particulate emissions in the air can be measured. This fugitive dust may be detrimental to the environment where the quantity of particulates is great and the concentrations of trace elements in the dust are at levels sufficiently high to be of concern. In the reported study, particulate concentrations were measured experimentally in the vicinity of a mine-mouth electric generating station. Operations at this major power complex included coal mining, handling, transport, and storage as well as construction and normal activities associated with power generation. Concentrations of elemental components of the collected particulates have been used to predict environmental effects; the predictions are based upon currently available values of estimated permissible concentrations. Finally, the predictions of environmental effects are compared with results of a brief field reconnaissance of vegetation growing in the vicinity of the power complex.

INTRODUCTION

Battelle's Columbus Laboratories has contracted with the U.S. Environmental Protection Agency (U.S. EPA) to perform a comprehensive environmental assessment of physical and chemical coal cleaning processes. The broad goal of this program (Contract No. 68-02-2163) is to establish a strong base of engineering, ecological, pollution control, and cost data which can be used to determine which coal cleaning processes are most acceptable from the technological, environmental, and economic viewpoints.

In order to obtain the field data necessary for the overall program, Battelle has undertaken a sampling and analysis program designed to identify the combinations of coal cleaning processes and environmental conditions that are most effective in reducing the total impact of coal use on the environment. This is being accomplished through the characterization of process and effluent streams from a variety of coal cleaning facilities and their associated coal transportation, storage, and refuse disposal areas. As part of this program, fugitive emissions in the vicinity of these operations were measured and evaluated in terms of their potential effects on the surrounding environments.

Specific Objectives

The recent construction of an advanced coal cleaning facility at a power complex near Homer City, Pennsylvania, provided a unique opportunity to obtain environmental data before operation of the facility for potential comparison with similar data to be obtained after operation would begin. Battelle conducted a series of pre-operational, multimedia, grab-sampling campaigns in a study area which included this facility, in order to document the abundance or concentrations of selected key parameters. These data were used to evaluate the air, water, and biological quality in the study area. The pre-operational environmental studies, while not sufficiently long-term to be a true baseline analysis, were conducted prior to operation of the cleaning plant as a reference point for future comparisons.

The objective of this paper is to compare the data obtained with the values listed in the Multimedia Environmental Goals (MEG) documents prepared for the U.S. EPA by Research Triangle Institute (Cleland and Kingsbury, 1977a and b). The MEG methodology was developed to meet the need for a workable system of evaluating and ranking pollutants for the purpose of environmental assessment of energy-related processes.

MEG values have been estimated for 216 pollutants by extrapolating various toxicity data by means of simple models. For most of these pollutants, maximum values have been estimated for each of the three media (air, water, and land). For each of the three media, separate maximum values have been estimated which are considered acceptable in preventing negative effects on (1) human health and (2) entire ecosystems.

The MEG values which are particularly appropriate for comparison with the environmental monitoring data Battelle collected in the area near Homer City are those termed estimated permissible concentrations (EPC's). EPC's are the maximum concentration of a pollutant which presents no hazard to man or biota on a continuous long-term basis. These EPC values are considered acceptable in the ambient air, water, or soil, as appropriate, and do not apply to undiluted effluent streams. The ambient application of EPC's corresponds to the ambient type of sampling conducted by Battelle prior to operation of the Homer City Coal Cleaning Plant. Specifically, particulate ambient concentrations were measured and compared with appropriate EPC's.

A second type of MEG values considered in this paper are minimum acute toxicity effluent (MATE) values. MATE's are concentrations of pollutants in undiluted effluent streams that will not adversely affect those persons or ecological systems exposed for short periods of time. These are appropriate for evaluating the potential hazards of particulates deposited on the ground and on vegetation in the study area.

Description of the Study Area

Nearly all of Battelle's environmental monitoring was conducted within a study area that can be approximately bounded by a circle 4 miles (6.4 km) in diameter. The advanced coal cleaning plant in the center of the study area is about 2 miles (3.2 km) southwest of Homer City, Pennsylvania.

The six major habitat types within the study area are hardwood forest, coniferous forest, cropland, grassland, water bodies, and areas of industrial development. The forested areas are primarily hardwoods, dominated by oak and hickory. Isolated pockets of pine are present as plantations rather than naturally-occurring species. Cropland is extensive in the study area, including contour and strip-cropped fields of corn, wheat, and hay. Grasslands include those areas which are presently grazed and those areas which were previously grazed or farmed and are now in a transition stage toward becoming a forest.

Almost the entire study area is on top of deep mines; nearby are abandoned or active strip mines. Homer City, Pennsylvania, is immediately adjacent to the study area on the Northeast, and Indiana, Pennsylvania, is only 5 miles (8.0 km) north of Homer City. During Battelle's sampling campaigns, both the coal cleaning plant and the refuse disposal area for that facility were under construction in the study area. Finally, the study area includes the Homer City Power Station with its associated coal storage, water treatment, and waste disposal facilities.

The Homer City Station is one part of an integrated power complex which includes two deep coal mines; coal cleaning, storage, and transport facilities; power generation facilities; and waste disposal and treatment facilities (Figure 1). Coal used at the Homer City Station comes from the two dedicated deep mines in the power complex, as well as coal hauled by truck from other mines. Solid refuse from power complex activities is deposited in three different types of disposal areas, including an ash disposal area, mine waste or "boney" piles, and the cleaning plant refuse disposal area.

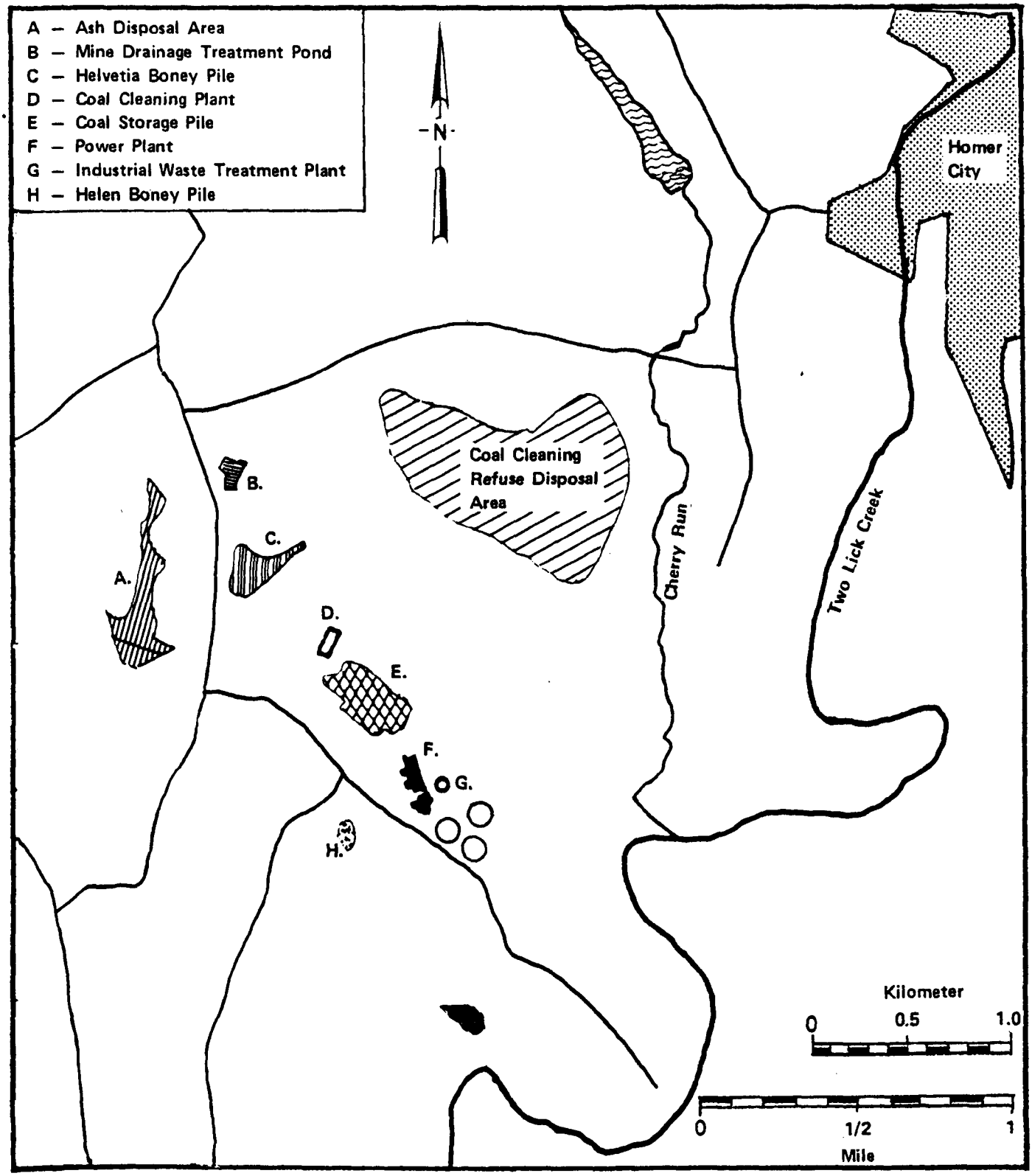

FIGURE 1. MAP OF THE STUDY AREA SHOWING COMPONENTS OF THE POWER COMPLEX

SAMPLING AND ANALYSIS TECHNIQUES

During the period from December 1976 through April 1977, a series of three pre-operational, grab-sampling campaigns were conducted by Battelle in the ambient media of the study area which included the Homer City Power Complex. These environmental monitoring studies involved sampling, laboratory analysis, and/or evaluation of the following components of the environment:

- Fugitive dust
- Water and stream sediments
- Aquatic biota
- Terrestrial biota
- Raw coal and fly ash
- Cleaning plant refuse disposal facility
- Groundwater.

The fugitive dust data of concern in this paper were collected and analyzed for comparison with MEG values. The samples collected during three campaigns were analyzed for both physical and chemical parameters. Information from the survey of terrestrial biota conducted during one campaign was utilized, as later described, in attempts to confirm the results of MEG comparisons.

Fugitive dust monitoring was conducted using high-volume (hi-vol) ambient air samplers during the following three 48-hour sampling periods:

- Campaign I: 8 p.m. December 17 to 8 p.m. December 19, 1976
- Campaign II: 8 p.m. January 5 to 8 p.m. January 7, 1977
- Campaign III: 8 p.m. April 5 to 8 p.m. April 7, 1977.

The first of these three campaigns was conducted over a weekend when both coal transfer and construction activities were low.

A multiple-source fugitive-dust dispersion model was used to select and verify locations for hi-vol samplers (Figure 2). This model takes into account such factors as wind speed, emission rate, particle size, and distance from selected potential dust sources located within the Homer City Power Complex. No dust sources outside of the power complex were incorporated in the model. On the basis of the computer-generated diffusion-modeling results, ten monitoring sites were established at distances of 175 to 2200 M downwind from various local dust sources. One of the ten sites was on private property downwind of the power complex property and one site was on private property upwind of the complex.

Several potential dust sources, both local and regional, were not incorporated into the diffusion model for sampling site selection. Dust generated by vehicular traffic, parking lots, construction activities, several storage silos, and especially the dusty surface of the plant grounds were not included in the model due to their erratic and non-point-source nature. Data for the Homer City power plant stack emissions were not available in time to include in the model. In addition, four other major power stations (Keystone,

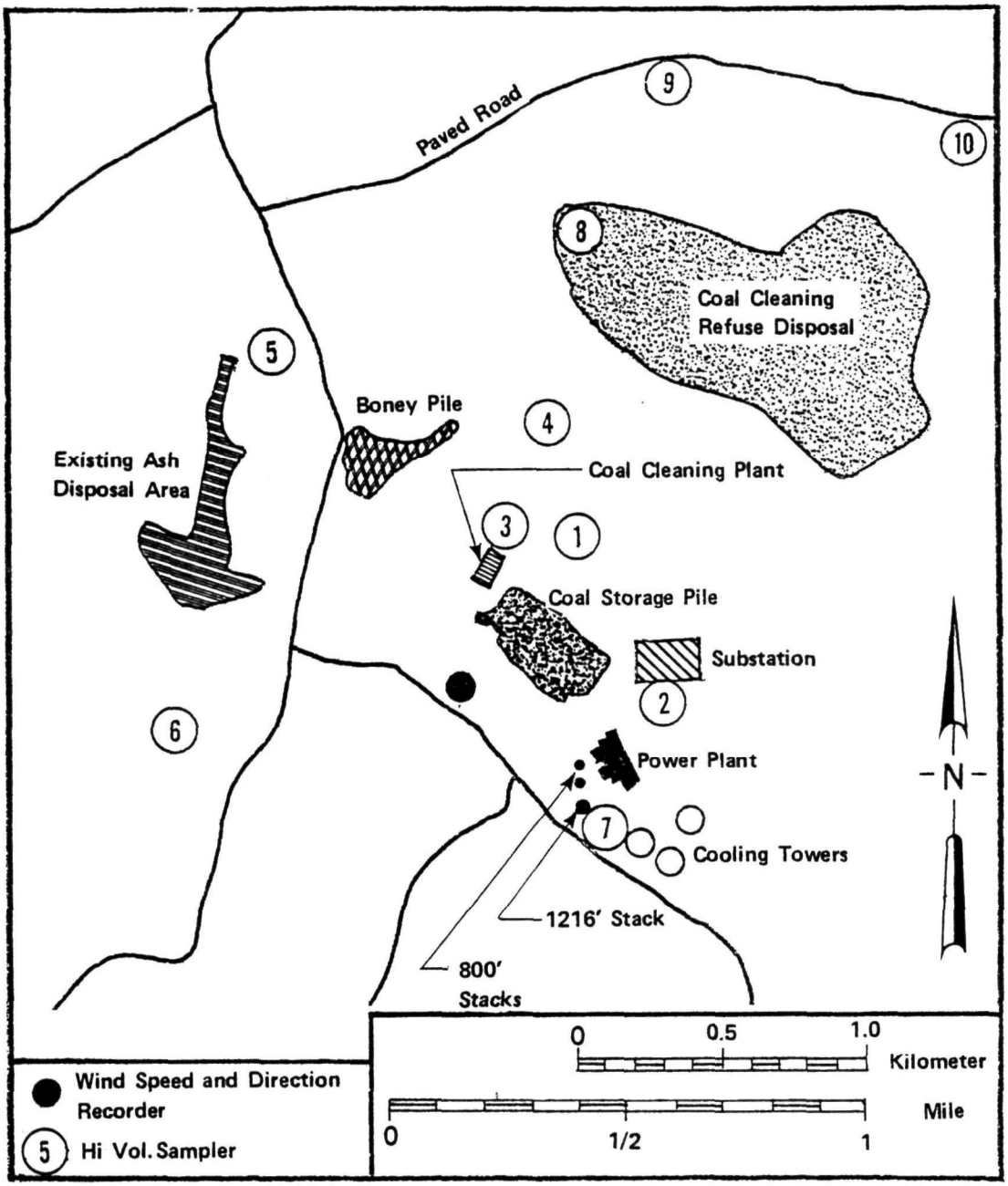

FIGURE 2. LOCATION OF FUGITIVE DUST SOURCES AND MONITORING SITES

Conemaugh, Seward, and Shawville) are located in the same Chestnut Ridge sector of the Allegheny Mountains as Homer City. These utilities are fed from coal mines located either directly under or near the station sites. The model did not include fugitive emission data from any of these facilities.

Potential fugitive dust sources at the Homer City Power Complex were investigated during a pre-sampling site evaluation. Some of the dust sources included an ash disposal area, boney piles at both deep mines, a coal storage pile, road dust, three power plant stacks, and construction-generated dust. The coal cleaning plant with its thermal dryers and the cleaning plant refuse disposal area were under construction during Battelle's sampling campaigns. Since these two areas were considered to be future potential sources of fugitive dust, they were considered in the selection of sampling sites.

In order to identify the type and quantity of pollutants being emitted from fugitive dust sources, a variety of analytical techniques were employed. Particulate mass was determined by weighing the 8 x 10-inch fiberglass filters used in the hi-vol samplers before and after each of the 12- or 24-hour sampling periods. A microscopic analysis was made of particulates to provide a distinction between components such as coal dust, fly ash, pollen, or construction dust. An Andersen sampling head was used on one hi-vol to obtain data on the distribution of particles in five size fractions.

Particulates on the filters from the hi-vol samplers were analyzed for up to 22 elements. The analytical technique used for most elements was atomic absorption; but neutron activation, colorimetry, a specific ion meter, a total organic carbon analyzer, an LDC mercury monitor, and potentiometric titration were also used. Since large amounts of four of these 22 elements (Na, K, Ca, and Mg) were found in the blank filters, the values for these four elements were not reported. Four of the remaining 18 elements (Sb, Ti, V, Se) were analyzed only in the second or third campaign. In general, the filter exhibiting the highest percentage of coal or ash from each site was used for analysis. Data from 15 of the elements analyzed are compared in this paper.

COMPARISON OF ANALYTICAL DATA WITH MEG VALUES

Analytical data for fugitive dust, fly ash, and raw coal sampled in the study area have been converted to the units used in the multimedia environmental goals (MEG) study (Cleland and Kingsbury, 1977a and b). These data are compared with the estimated permissible concentrations (EPC's) and/or the minimum acute toxicity effluent (MATE) values defined in the Introduction to this paper.

Average concentrations of 15 elements analyzed in the fugitive dust from the study area are compared with the EPC's for air in Table 1. Since most of the fugitive dust appeared to emanate from the coal storage pile and decline in concentration within 200-300 m downwind (Figure 3), the data have been averaged for the sampling sites located between 150-175 m and 400-1,800 m downwind from the coal pile. The fugitive dust concentrations for the upwind "control" sampling location are also provided. These field data are followed by the appropriate maximum EPC's for air recommended for each element to prevent negative effects to humans or the surrounding environment during continuous long-term (chronic) exposure. A difficulty in making comparisons between observed and recommended levels of the 15 elements shown in Table 1 is that three EPC's for human health and 10 EPC's for the environment are not available.

Average concentrations for three of the elements (As, Cr, and Pb) analyzed in fugitive dust exceeded the EPC's for human health. These values have been underlined in Table 1. It is noteworthy that two of these elements (As and Cr) had concentrations above the health-based EPC even at the upwind "control" location.

Maximum and minimum concentrations of 15 elements analyzed in fugitive dust are compared with the appropriate EPC's for soil in Table 2. Again, the data are grouped to include sampling sites less than 200 m (i.e., 150-175 m) and greater than 200 m (i.e., 400-1,800 m) downwind of the coal pile. Concentrations of the same elements in the raw coal are also shown. EPC's for protection of human health and the environment are given for 12 elements; no EPC values for iron, chlorine, and fluorine have been determined.

The majority of the elements analyzed showed maximum and frequently minimum concentrations in the fugitive dust that were far greater than the EPC levels suggested for the soil. Ten elements exceeded the EPC's for human health and 11 elements exceeded the EPC's for the environment. Both the maximum and minimum concentrations of 8 elements (As, Cd, Cr, Cu, Pb, Mn, Ni, and Se) in the fugitive dust exceeded the EPC's for both human health and the environment.

Obviously, the concentrations of toxic trace elements in fugitive coal dust that has settled to the ground does not mean that these same concentrations occur in the soil. However, studies involving soil contamination by other types of particulate deposition have shown that toxic trace elements

TABLE 1. FUGITIVE DUST COMPARISONS ($\mu g/m^3$): EPC VALUES FOR AIR VERSUS HOMER CITY DATA

	Trace Element Concentrations, $\mu g/m^3$														
	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Ti	Zn	Cl	F	V	Se
Distance from Coal Pile	Average Concentration in Fugitive Dust During 3 Campaigns at Homer City (24-hr Sampling Periods)[a]														
Downwind 150-175 m[b]	0.014	0.008	.026	0.292	3.45	0.586	0.076	0.00056	0.015	0.44	0.35[i]	1.97[i]	5.47	ND[j]	0.0049
Downwind 400-1,800 m[c]	0.010	0.014	.015	0.119	1.87	0.334	0.093	0.00009	0.013[i]	0.32[i]	0.22	0.82	2.03	ND	0.0026[i]
Upwind Control[d]	0.009	0.005	.014	0.223	1.65	0.258	0.041	0.00003	0.009	0.17[i]	0.13	1.05	1.40	0.02[i]	0.0030[i]
EPC Category	Estimated Permissible Concentrations (EPC's)[e], $\mu g/m^3$														
Health	0.005	0.12[f]	0.002[f]	0.5	--[h]	0.36	12	16[f]	0.04[f]	14	9.5	--	--	1.2	0.5
Ecology	--	0.04[g]	--	--	--	1[g]	--	0.01[g]	--	--	--	--	--	0.1	0.03[g]

(a) All data were collected between December 1976 and April 1977.
(b) Average for sampling sites 1 and 3; downwind of coal pile.
(c) Average for sampling sites 4, 8, and 9; downwind of coal pile.
(d) Sampling site 6; upwind of coal pile about 1600 m and off of the power station property.
(e) From Cleland and Kingsbury (1977a and b).
(f) Based on a Toxic Limit Value (TLV) which recognizes the element's carcinogenic potential
(g) Based on teratogenic potential.
(h) Not available.
(i) Concentrations were not available for some sampling sites during all three campaigns.
(j) ND = not detectable.

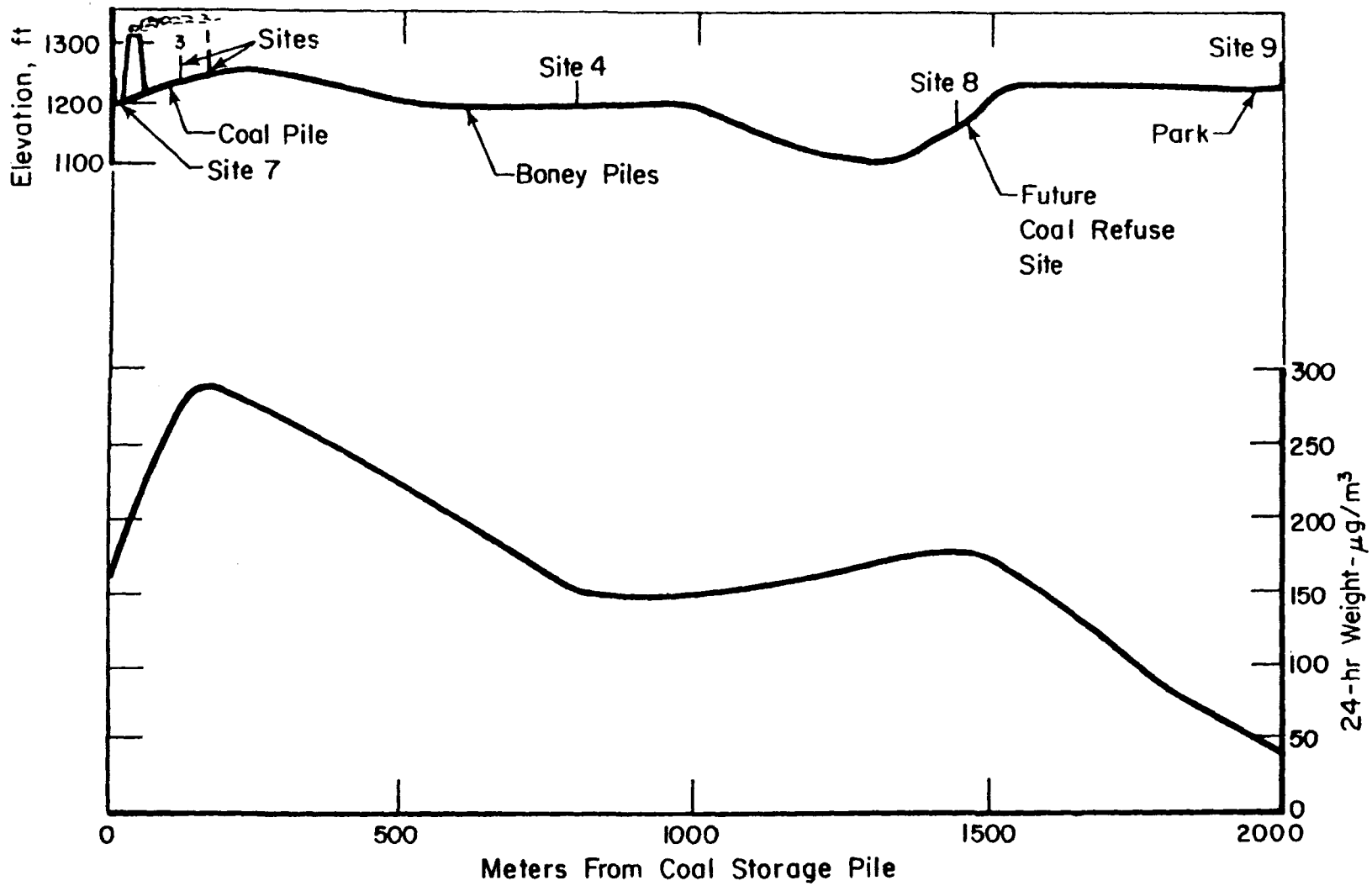

FIGURE 3. FUGITIVE DUST CONCENTRATIONS COMPARED TO A TRANSECT OF THE AREA'S TOPOGRAPHICAL RELIEF

TABLE 2. FUGITIVE DUST COMPARISONS (μg/g): EPC VALUES FOR SOIL[a] VERSUS HOMER CITY DATA

	Trace Element Concentration, μg/g														
	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Ti	Zn	Cl	F	V	Se
	Concentrations in Particulate at Sampling Sites within 200 m of Homer City Coal Pile (Sites 1 and 3)[a]														
Maximum	154	264	471	3,678	28,736	17,241	632	3	264	8,676	3,563	39,081	55,000	ND	33
Minimum	11	18	18	336	6,223	501	65	0.2	23	626	ND[f]	4,043	ND	ND	4
	Concentrations in Particulate at Sampling Sites Between 200 and 2,000 m of Homer City Coal Pile (Sites, 4, 8, and 9)[a]														
Maximum	238	619	667	6,061	57,576	12,857	5,603	2	545	3,007	5,152	27,278	45,600	ND	122
Minimum	34	ND	46	220	11,477	566	6	ND	ND	1,356	943	5,806	1,818	ND	1
	Estimated Permissible Concentrations (EPC's) for Soil[b]														
Health	10	0.06[c]	0.01[c]	200	--[e]	70[c]	10	50[c]	0.1[c]	17	1,000	--[e]	--[e]	1.4	2
Ecology	2	0.01[d]	10	20	--[e]	3[d]	4	3[d]	0.4	800	4	--[e]	--[e]	15	0.1[d]
	Raw Coal Concentrations Determined by Individual Analysis of Three Homer City Coal Sources[g]														
Maximum	48	0.26	35	31	48,750	17.3	74	1.1	16.8	1,329	66	0.26	108	65	ND
Minimum	22	<0.1	30	20	18,000	12	35	0.34	12.8	1,125	46	0.23	91	55	ND

(a) Data from three sampling campaigns conducted by Battelle in the study area.
(b) From Cleland and Kingsbury (1977a and b); all values were multiplied by 100 based on personal communication with Kingsbury (August, 1978).
(c) Based on carcinogenic potential.
(d) Based on teratogenic potential.
(e) Value not available.
(f) ND = not detectable.
(g) Coal sources include: Helen Mining Company and Helvetia Coal Company (from Upper Freeport Seam); and Trucked-in Coal (from Lower Kittanning Seam).

in these particulates can cause ecosystem disruption resulting in the loss of essential nutrients and can also result in increased concentration of these toxic elements in both plants and animals. These types of effects have been demonstrated for lead smelter emissions (Jackson and Watson, 1977; Kerin, 1975) and for fly ash emissions from coal-fired power plants (Furr, et al., 1977). Dvorak, et al. (1978), have speculated that long-term exposure to uncombusted coal dust may cause changes in vegetation community structure similar to those caused by particulates from coal combustion.

Mechanisms for the movement of toxic trace elements from particulate emissions deposited on the ground to the root zone of the soil are complex (Vaughan, et al., 1975; Dvorak, et al., 1978). A partial list of the factors which influence leaching of trace elements from deposited particulates into the soil solution include (1) the size and type of particulates, (2) the amount and acidity of precipitation, (3) the concentrations and physico-chemical properties of the trace elements, (4) the texture, organic content, pH, and other characteristics of the soil, (5) the solubility of elements into the soil solution, and (6) the temperature of the air and soil.

The fugitive dust quantity and composition found during monitoring has probably been accumulating on the ground in a reasonably similar fashion since the power plant (including the coal storage pile) began operation in 1969. Thus, mobile elements in the settled dust may have leached into the soil. The quantity of toxic trace elements available to vegetation, however, needs to be determined by chemical analysis of the soil. In spite of any leaching of trace elements that may have increased soil concentration, the vegetation for some distance from the coal pile has not yet shown any adverse effects that were readily apparent during Battelle's field reconnaissance. An analysis of soil biota and plant diversity, however, was not conducted.

Another basis for comparison is also possible; MATE values for components in solid wastes have also been developed. Inasmuch as the deposited fugitive dusts are tantamount to being a solid waste and these deposits may contact or be absorbed or consumed by plants and animals, comparisons with MATE values for solid wastes would appear to be valid. Such a comparison has been made in Table 3. The table's structure is similar to that of Tables 1 and 2.

In Table 3, the appropriate MATE values are judged to be the ones related to ecology limits. In general, these have lower values than those for health; exceptions are mercury (Hg), chlorine (Cl), and fluorine (F), the latter two for which there are no ecology values available. Twelve of the fifteen MATE values for health are exceeded by the maximum values for both the close-in (<200 m) and the more remote (>200 m) sampling sites. Eleven of the ecology values are exceeded. Comparisons of solid waste MATE values with the elemental concentrations in the raw coals are also provided in Table 3. Elemental concentrations in the raw coal exceed many of the same elemental EPC values exceeded by elements in fugitive dust. However, the levels of toxic elements in the raw coal are generally lower than the levels in the fugitive dust.

TABLE 3. FUGITIVE DUST COMPARISONS (μg/g): MATE VALUES FOR SOLID WASTE[a] VERSUS HOMER CITY DATA

	Trace Element Concentration, μg/g														
	As	Cd	Cr	Cu	Fe	Pb	Mn	Hg	Ni	Ti	Zn	Cl	F	V	Se
	Concentrations in Particulate at Sampling Sites within 200 m of Homer City Coal Pile (Sites 1 and 3)[a]														
Maximum	154	264	471	3,678	28,736	17,241	632	3	264	8,676	3,563	39,081	55,000	ND	33
Minimum	11	18	18	336	6,223	501	65	0.2	23	626	ND[f]	4,043	ND	ND	4
	Concentrations in Particulate at Sampling Sites Between 200 and 2,000 m of Homer City Coal Pile (Sites, 4, 8, and 9)[a]														
Maximum	238	619	667	6,061	57,576	12,857	5,603	2	545	3,007	5,152	27,278	45,600	ND	122
Minimum	34	ND	46	220	11,477	566	6	ND	ND	1,356	943	5,806	1,818	ND	1
	Minimum Acute Toxicity Effluent (MATE's) for Solid Waste[b]														
Health	50	10	50	1,000	300[c]	50	50	2	45	18,000	5,000	260,000[d]	7,500[h]	500	10
Ecology	10	0.2	50	10	50[c]	10	20	50	2	160	20	--[e]	--[e]	30	5
	Raw Coal Concentrations Determined by Individual Analysis of Three Homer City Coal Sources[g]														
Maximum	48	0.26	35	31	48,750	17.3	74	1.1	16.8	1,329	66	0.26	108	65	ND
Minimum	22	<0.1	30	20	18,000	12	35	0.34	12.8	1,125	46	0.23	91	55	ND

(a) Data from three sampling campaigns conducted by Battelle in the study area.
(b) From Cleland and Kingsbury (1977a and b); all values were multiplied by 100 based on personal communication with Kingsbury (August, 1978).
(c) MATE values listed are for ferrous (Fe^{+2}) or ferric (Fe^{+3}).
(d) MATE value listed is for chloride ion (Cl^{-}).
(e) Value not available.
(f) ND = not detectable.
(g) Coal sources include: Helen Mining Company and Helvetia Coal Company (from Upper Freeport Seam); and Trucked-in Coal (from Lower Kittanning Seam).
(h) MATE value listed is for fluoride ion (F^{-}).

CONCLUSIONS AND RECOMMENDATIONS

Elemental concentrations in fugitive dust measured in the study area exceeded both EPC and MATE values for air, soil quality, and solid wastes. For example, three out of 15 elements analyzed in fugitive dust had concentrations above the health-based EPC's for air quality. Comparisons with ecology-based EPC's for air quality, however, were very difficult due to the absence of ten EPC values.

Although no soil concentrations were determined, comparisons of elemental concentrations in fugitive dust were made with ecology-based EPC's for soil due to the potential problem of toxic elements leaching into the soil from deposited fugitive dust. Eleven of the 15 elements studied had concentrations in the fugitive dust that were above the ecology-based EPC's for soil. Twelve solid waste MATE's for health and eleven for ecology were exceeded also. In spite of the dust (particularly coal dust) present on the ground for some distance around the coal pile, however, the vegetation has not yet begun to show any obvious adverse effects.

Although applicability of EPC and MATE values are currently somewhat limited because of known deficiencies now being corrected, the trend in these observations appears clear. Many trace element concentrations in the study area are higher than desirable. The conclusion is not, however, that immediate corrective action is needed. There is no obvious damage to the vegetation even though the present conditions have evidently persisted for some time.

Several recommendations can be made based on this study. First, more field experiments are needed to validate these results. The amount of available data is small and more extensive sampling and elemental analysis, particularly of soil and plant and animal tissues, are needed. These steps are necessary to determine the fate of the trace metals in the fugitive dust.

Additional research also needs to be conducted on EPC and MATE values to evaluate and rank pollutants for the purpose of environmental assessment. Much of this work was recommended in the initial MEG document (Cleland and Kingsbury, 1977a) and is now or will soon be in progress. For example, MEG's need to be related to the specific compounds or ionic forms of an element that are most toxic rather than having a single value represent all compounds and ions which have a common "parent" element. Synergistic and antagonistic effects need to be considered, because these effects may drastically change the hazard ranking of a pollutant in a specific situation. MEG's are also needed for many of the master parameters, such as the "totals" identified by Cleland and Kingsbury (1977a: 115) (e.g., total particulates).

In another vein, the comparison of trace element concentrations in fugitive dust and MEG values points out the need for laboratory and field research, particularly in relation to fugitive dust which consists predominantly of coal particles. First, the rates at which toxic elements leach from coal dust into a variety of soil types need to be explored. Second, the concentrations of toxic elements present in the soil around a large, open coal pile need to be determined where this pile has been in existence for a long period of time. Third, laboratory bioassay and long-term field studies need to be conducted on the effects of coal dust on plants and animals.

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INDUSTRIAL NON-POINT SOURCES:

ASSESSMENT OF SURFACE RUNOFF

FROM THE IRON AND STEEL INDUSTRY

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ABSTRACT

TRC - THE RESEARCH CORPORATION of New England was retained in April 1976 by the Environmental Protection Agency to assess whether surface runoff from iron and steel mills is an environmental problem. The program included:

- Identifying and characterizing sources of surface runoff unique to iron and steel mills.
- Assessing the specific problems associated with surface runoff and evaluating the contribution made by individual sources to the total problem.
- Identifying industry control systems which could be used to treat contaminated stormwater.

TRC researched the limited existing data available, conducted plant tours, and designed and performed a field survey at two fully integrated mills on tidal rivers. Data collected at the two sites indicate that the coal and coke storage piles and the coal and coke handling areas have the highest potential for contaminating stormwater. The data also indicate that TSS runoff concentrations are typical of urban runoff concentrations while TDS values are approximately double the typical urban runoff concentrations.

Stormwater controls which presently exist within the steel industry are limited. The only system specifically designed for stormwater control exists at Armco's Houston Works where coal piles have been diked as a control measure for both fugitive air emissions and stormwater runoff. Some mills collect stormwater runoff with process wastewater for subsequent treatment at a terminal plant. Those methods which are applicable to the industry include rainfall detention ponding rings for flat roofs, swirl degritters, and retention basins or sedimentation ponds.

TRC concluded that, with the exception of runoff from coal and coke storage areas, stormwater runoff is not a problem when compared to point source control.

1.0 INTRODUCTION

Since industries and municipalities are on the way to meeting the point source standards of the 1977 interim goal of PL 92-500 (Federal Water Pollution Control Act Amendments of 1972), the effect of non-point source pollution on water quality is gaining more attention. The National Commission on Water Quality reported in 1976 that "non-point pollutant sources are significant to the Commission's study because they may in some instances overwhelm and negate the reductions achieved through point source effluent limitations."[1] Based on these findings the Commission recommended to Congress that "control or treatment measures shall be applied to agricultural and non-point discharges when these measures are cost-effective and will significantly help in achieving water quality standards."[2]

Non-point sources are diffuse in nature, usually intermittent, site specific, not easily monitored at their exact source, related to uncontrollable meteorological events (precipitation, snow melt, drought), and not usually repetitive in nature from event to event. The primary transport mechanism for non-point sources is water runoff from meteorological events. The three basic modes of runoff transport are overland (surface) flow, interflow (also called interstitial flow), i.e., flow through the ground between the surface and groundwater levels, and groundwater flow. Surface runoff will usually contain the highest quantity of contaminants and is the most rapid method of transport of pollutants from non-point sources.

Because of the great quantities of water and raw material used in making iron and steel, mills are usually located near waterways. Contaminated stormwater runoff from these mills could rapidly reach these waterways; thus the potential of causing a detrimental environmental impact is often present.

In April 1976 the Metallurgical Processes Branch of the Industrial Environmental Research Laboratory (IERL) of the Environmental Protection Agency (EPA) at Research Triangle Park, North Carolina, retained TRC - THE RESEARCH CORPORATION of New England to perform an assessment of surface runoff from iron and steel mills as a task under an existing contract (68-02-2133). The principal objective of this program was to provide EPA with an evaluation which it can use in determining if stormwater runoff from iron and steel mills is an environmental problem and should be included in the Agency's long-term planning as an area of concern.

The program had the following sub-objectives:

1. To identify sources of surface runoff unique to iron and steel mills and to characterize runoff streams in terms of quantity and composition.

2. To assess the specific problems of surface runoff at iron and steel mills and evaluate the contribution made by these individual sources to the overall problem.

3. To identify control systems used by the industry or by other industries which are or could be used to treat contaminated storm-water.

This paper presents a general overview of the program with emphasis on the field sampling portion of the program which was conducted at two iron and steel mills.

2.0 EXISTING DATA AND IDENTIFICATION OF SURFACE RUNOFF SOURCES

Before this program, little work had been performed on surveying stormwater and identifying potential sources of stormwater contamination in the steel industry. Previously, the most comprehensive studies had been undertaken by Armco Steel Corporation's Houston Works in Houston, Texas,[3] and Kaiser Steel's Fontana, California plant.[4]

In the Armco study, the mill was divided into drainage basins, each characterized according to size, activity, and land cover (i.e., buildings, paved area, railroad track, undeveloped land, stockpiles, and ponds). Each basin was sampled for several storms. Parameters measured included total suspended solids (TSS), oil and grease, biochemical oxygen demand (BOD_5), total organic compound (TOC), and chemical oxygen demand (COD).

Armco found that stormwater quantity and quality varied appreciably with drainage basin characteristics and location. This limited the validity of any correlation of parameter concentrations between basins. Furthermore, the quality of stormwater runoff was found to vary directly with storm duration and intensity and also with the number of antecedent dry days prior to storms. As antecedent dry days increase, so does the potential particulate matter to be scoured. Other significant results of the Armco study were the absence of a "first flush" effect, and the absence of significant quantities of organic matter. The "first flush" effect is a condition where the matter accumulated in a basin since the last runoff event is scoured from the area at the start of the next storm event. In almost all cases a flow dependent effect was observed; i.e., peak parameter concentrations occurred at peak runoff flows.

The Kaiser program involved sampling during the rainy season (February and March) in 1975. Runoff from twelve storm events was sampled for chloride, conductivity, and oil and grease. The oil and grease results from the Kaiser program were much higher than those obtained at Armco.

Since the Armco and Kaiser studies were the only data existing on stormwater runoff from steel mills, several plants were toured as part of this program in an effort to combine a number of factors which affect site specific runoff, such as terrain, climate, mill locations and operations, into an overall industry-wide assessment of the most probable sources of stormwater contamination. The following companies were contacted and/or visited:

United States Steel
National Steel
Armco Steel
Republic Steel
Youngstown Sheet and Tube
Inland Steel
Kaiser Steel
CF&I Steel
Alan Wood Steel

Runoff from the activities and operations of steel mills was segmented into the following groups:

- Runoff from storage and disposal piles (coal, coke, slag, iron).
- Runoff from adjacent urban areas into the mill.
- Runoff from slag handling and processing facilities.
- Runoff of accumulated materials from roof and ground areas from several mill operations (blast furnace, sinter plant, BOF shop, open hearth, coke and by-product plant, coal and coke handling, and finishing areas).

Because runoff is site specific, it was impossible to compare the contaminated stormwater potential of an area in one particular mill to the same area in another mill. Climate, terrain, operations, maintenance, and the location of processes relative to each other are unique to each mill. Therefore, a rating system was devised which ranked the relative potential of each activity or operation at an individual plant. Based on the assessments of TRC personnel, the ratings were entirely subjective, except where physical data were available (e.g. Armco's Houston Works). The following activities or operations were rated as having the greatest potential for contaminating stormwater:

- Coal storage piles
- Coke storage piles
- Slag dumps
- Iron ore and pellet storage piles
- Coal and coke handling

3.0 FIELD PROGRAM

A field survey was performed at two different sites in the spring of 1977. Both sites were fully integrated mills on tidal rivers. The general characteristics of each site are described in Table 1.

Tables 2 and 3 describe the individual drainage basins sampled at Sites 1 and 2 respectively. At Site 1, the drainage basins had been defined in previous stormwater runoff programs conducted by plant personnel. Because there are no well defined drainage basins within Site 2, the delineation of drainage areas was based upon the storm sewer plans for the plant. The storm sewer system for Site 2 originates at the roof drains from most of the plant operations, includes the road and railroad line runoffs, and terminates in either a canal leading into the tidal river or into the river directly.

The general test plan for the survey was designed to determine:

1. Background conditions at each sampling location prior to a storm event, i.e., dry weather flow conditions.

2. Volume of stormwater runoff and pollutant concentrations in the runoff as a function of time for the storm event.

The following additional data were gathered:

1. Rainfall accumulation as a function of time for the storm event.

2. Dustfall accumulation between storms.

No attempt was made to assess the effects of the stormwater runoff on the receiving water.

The parameters measured in this survey were:

Runoff Flow	Rainfall
Total Iron	Total Suspended Solids (TSS)
Dissolved Iron	Total Dissolved Solids (TDS)
Phenols	Cyanide
Ammonia	Sulfates

Oil and grease and organic parameters such as BOD_5, COD and TOC were not measured because previous work performed by TRC showed that these parameters would not be of sufficient magnitude to be of concern.

TABLE 1

GENERAL SITE CHARACTERISTICS

	Site 1	Site 2
Age of Plant	37 Years	25 Years
Developed Acreage	575	3900
Terrain	Flat, Semi-Permeable	Flat, Permeable
Runoff Receiving Body	Tidal River	Tidal River
Plant Operations	Coke Plant, Sinter Plant, Blast Furnaces, Electric Furnaces, Finishing Operations	Coke Plant, Sinter Plant, Blast Furnaces, Open Hearth Furnaces, Electric Furnaces, Finishing Operations
Period of Sampling	3/77 to 4/77	5/77 to 6/77
Number of Sampling Points	5	13
Permanent Flow Devices	Yes	No

TABLE 2

DESCRIPTION OF INDIVIDUAL DRAINAGE BASINS SAMPLED
SITE 1

Basin	Activities and Operations	Acreage
005	Wide Flange Mill; Shipping Office; Roundhouse (car, truck, and railroad car repair facility); western halves of the No. 1 Electric Furnace Shop, No. 2 Plate Mill, Plate Shipping Building, Heavy Plate Shear Building, and Plate Heat Treat Building.	142.8
006	Direct Reduction Plant.	4.6
009	West end of the Mold Foundry; area between the Coke Plant proper and the east end of the Stock House; Coke Transfer.	6.6
010	Coal transfer; main coal conveyor belt from the dock area to the coal storage area; Coal Shaker Building; numerous coal transfer points located in immediate vicinity of the west end of the Coke Plant area	2.6.
011	Mold Preparation Shop; eastern part of the No. 2 Electric Furnace Shop; eastern half of the Coke Ovens; Coke Oven By-products area; coal pile storage area; eastern half of the Mold Foundry; employee parking area.	60.5

TABLE 3

DESCRIPTION OF INDIVIDUAL DRAINAGE BASINS SAMPLED
SITE 2

Basin	Activities and Operations	Acreage
002	Diesel Repair Shop.	5.5
003	Slag Filled Borrow Area; Railroad tracks.	4.8
004	Mold Preparation Shop; northeast side of Open Hearth Furnaces; Railroad tracks.	18.6
005	South end of Open Hearth Shop; Railroad tracks; Slag Dump Area; Mold Preparation.	4.0
006	Hot mills; Slab cooling area; Slab mill; Billet mill.	12.0
007	Hot strip mills.	5.2
008	Blast furnace; Sinter Plant; Employee Parking; Ore Conveyors.	9.1
009	Sinter Plant; Ore Conveyors; Roadways.	11.4
010	One half of Open Hearth Plant; Coke Plant; Coke yards; Numerous Railroad lines; Coke By-Products Complex.	144.6
011	Coal storage.	9.9
012	Southern end of Coke Ovens - surface runoff.	1.3
013	Southern end of Coke Ovens - surface runoff.	1.5
014	Ladle Repair Shop; Railroad track area.	0.6

Previous work by Armco revealed very high concentrations of COD and TOC which were concluded to be a result of inert coal and coke fines and not reactive organics.

The sampling sites were located in the areas of material storage and disposal and coal and coke handling. No slag dump runoff data were obtained as neither site had a representative slag dump. In addition, due to tidal backflow problems, no iron ore or pellet storage pile runoff data were obtained at Site 1. The sample collection method, flow monitoring technique, sampling schedule, and parameters to be analyzed at each of the sampling sites within Site 1 and 2 are summarized in Tables 4 and 5 respectively.

3.1 Field Survey Results

3.1.1 Site 1 Results

Table 6 summarizes the storm event data for Site 1. Of the five storm events sampled at Site 1, only the storm of March 31 approximated the high intensity, short duration rainfall typical of this semitropical area. From historical observations of previous storm events at Site 1, it was expected that the total rainfall at various locations around the plant would differ over the course of a storm. This uneven distribution of rainfall was never observed during the field program. During the sampling program, the rainfall was typically a steady drizzle with occasional heavy downpours uniformly distributed over the entire plant. In all five events, rain wedge totals closely corresponded to the recording rain gages.

Table 7 summarizes the flow data from Site 1. Time-weighted average flow data plus the range of flow for both dry and wet weather sampling are listed. The dry weather flows at outfall 010 were not measurable; the water levels over the weir were essentially zero except for a small trickle which volumetrically was negligible. Wet flow data were limited at outfall 010 due to occasional tidal backflows. At outfalls 005 and 011 wet flows were significantly higher than dry flows. Outfall 009 showed the effects of tidal backflow from the river and neither samples nor flow measurements could be obtained at the weir during any of the storm events.

Tables 8 through 12 all refer to the pollutant data measured at Site 1. The range (Table 8), the mean (Table 9) and the average mass loadings (Tables 10 through 12) of pollutants show the differences between dry and wet weather conditions. Average mass loadings of pollutants for dry weather conditions were calculated by multiplying the mean concentrations value measured during each storm by the time-weighted average flows from Table 7. Average mass loadings for wet weather conditions were calculated by multiplying the time weighted average concentrations by the time-weighted average flows, both determined from the concentration and flow curves for each rainfall event. The time weighted average wet weather flows pertain to the time over which

TABLE 4

SUMMARY OF SAMPLING SITES AND SAMPLING PROGRAM
SITE 1

OUTFALL	SAMPLE COLLECTION METHOD	FLOW METHOD	SAMPLING SCHEDULE FOR STORM EVENTS	PARAMETERS TO BE ANALYZED
005	ISCO Sampler with weir	ISCO Flow Meter and Printer	Every storm event	TSS, TDS
009	ISCO Sampler with weir	ISCO Flow Meter and Printer	Every storm event	TSS, TDS, Total Fe, Dissolved Fe, Phenols, Cyanide, Ammonia
010	ISCO Sampler with weir	ISCO Flow Meter and Printer	Every storm event	TSS, TDS, Total Fe, Dissolved Fe, Phenols, Cyanide, Ammonia, Sulfates
011	ISCO Sampler with weir	ISCO Flow Meter and Printer	Every storm event	TSS, TDS, Total Fe, Dissolved Fe, Phenols, Cyanide, Ammonia

TABLE 5

SUMMARY OF SAMPLING SITES AND SAMPLING PROGRAM
SITE 2

OUTFALL	SAMPLE COLLECTION METHOD	FLOW METHOD	SAMPLING SCHEDULE FOR STORM EVENTS	PARAMETERS TO BE ANALYZED
002	Grab	Bucket and stop-watch	Every storm event (when possible)	TSS, TDS
003	Grab	Bucket and stop-watch	Every storm event (when possible)— low priority	TSS, TDS
004	ISCO Sampler with weir	ISCO flow meter and printer	Sample 2 of sites 004, 006, & 007 for each storm	TSS, TDS Total Fe Dissolved Fe
005	Grab	Bucket and stop-watch	Every storm event (when possible)— low priority	TSS, TDS
006	ISCO Sampler with weir	ISCO flow meter and printer	Same as 004	TSS, TDS
007	ISCO Sampler with weir	ISCO flow meter and printer	Same as 004	TSS, TDS
008	Grab	None	Every storm event (when possible)	TSS, TDS Total Fe Dissolved Fe
009	Grab	None	Every storm event (when possible)	TSS, TDS Total Fe Dissolved Fe Metals
010A 010B	ISCO Sampler	Gurley meter	Every storm event	TSS, TDS Total Fe Dissolved Fe Phenols, Ammonia, Cyanide
011	Grab	None	Every storm event	TSS, TDS, Sulfate, Phenols, Ammonia, Total Fe, Dissolved Fe, Metals
012	Grab	None	Sample 2 of 012, 013, & 014 for each storm	TSS, TDS, Phenols, Sulfates, Ammonia, Total Fe, Dissolved Fe, Metals, Cyanide
013	Grab	None	Same as 012	TSS, TDS, Phenols, Sulfates, Ammonia, Total Fe, Dissolved Fe, Metals, Cyanide
014	Grab	None	Same as 012	TSS, TDS, Total Fe Dissolved Fe
015	Grab	None	Every storm event	TSS, TDS, Phenols, Sulfates, Ammonia, Total Fe, Dissolved Fe, Cyanide

Table 6

STORM EVENT DATA
SITE 1
MARCH - APRIL, 1977

Date	Storm Beginning	Storm Ending	Total Rainfall		Average Rainfall Intensity		Maximum Rainfall Intensity	
			cm	(inches)	cm/hr	(in/hr)	cm/hr	(in/hr)
3/24/77	0500	2130	0.84	(0.33)	0.05	(0.02)	0.13	(.05)
3/27-3/28/77	2000 (3/27)	0200 (3/28)	1.42	(0.56)	0.23	(0.09)	0.61	(0.24)
3/31/77	1410	1430	0.20	(0.08)	0.61	(0.24)	1.07	(0.42)
4/4/77	0200	0500	0.36	(0.14)	0.13	(0.05)	0.41	(0.16)
4/16/77	0430	2000	0.71	(0.28)	0.05	(0.02)	0.56	(0.22)

Table 7

DRY vs WET FLOWS[a][d]
SITE 1
MARCH - APRIL, 1977

OUTFALL	005				010		011			
DATE	DRY		WET		WET[b]		DRY		WET	
	Avg. Flow lpm(gpm)	Range lpm(gpm)	Avg. Flow lpm(gpm)	Range lpm(gpm)	Avg. Flow lpm(gpm)	Range lpm(gpm)	Avg. Flow lpm(gpm)	Range lpm(gpm)	Avg. Flow lpm(gpm)	Range lpm(gpm)
3/24			1056 (279)	227-2233 (60-590)	12.5 (3.3)	0 - 29.1 (0 - 7.7)			189 (50)	45 - 534 (12-141)
3/27 - 28			6083 (1607)	454-15026 (120-3970)	16.0 (4.2)	0 - 67.0 (0 - 17.7)			708 (187)	38 - 2203 (10 - 582)
3/29	473 (125)	435-568 (115-150)					38 (10)	27-53 (7-14)		
3/31			401 (106)	227-984 (60-260)	ND[c]	ND[c]			405 (107)	95-939 (25-248)
4/4			2112 (558)	568-4542 (150-1200)	ND[c]	ND[c]			170 (45)	15-367 (4-97)
4/5	227 (60)	227 (60)					3.4 (0.9)	0-13.2 (0-3.5)		
4/16			3456 (913)	228-15900 (60-4200)	13.3 (3.5)	0 - 49.02 (0 - 13.0)			583 (154)	83-1374 (22-363)
4/18	216 (57)	76-254 (20-67)					87 (23)	76-106 (20-28)		

(a) No flow data were taken at Outfalls 006 and 009, nor at the coal pile drainage ditch.
(b) There was no measurable dry flow at 010 during the program.
(c) ND - No flow data were obtained.
(d) Flow values are time weighted averages for the entire event.

TABLE 8

RANGE OF POLLUTANT CONCENTRATIONS AT THE
SAMPLING LOCATIONS AT SITE 1
MARCH - APRIL, 1977

Pollutant	Range of Pollutant Concentration, mg/ℓ					
	Outfall 005		Outfall 010		Outfall 011	
	Dry	Wet	Dry	Wet	Dry	Wet
Total Suspended Solids	4-31	11-113	4-649	10-1272	7-42	9-151
Total Dissolved Solids	327-463	238-964	2007-5438	661-4993	668-1049	427-1196
Total Iron			1.1-8.3	1.2-3.6	1.1-2.7	0.96-5.8
Dissolved Iron			0.1-0.6	0.1-0.2	0.10[a]	0.10-0.30
Phenols			16-34	0.02-1.1	0.02-0.68	0.01-0.52
Cyanide (Total)			n.d.-0.99[b]	n.d.[b]	n.d.[b]	n.d.-0.01[b]
Ammonia			54-96	3.6-73	0.57-26	0.65-28
Sulfate			400-1580	180-490		

[a] Only one value obtained.

[b] n.d. - not detectable - detectable limit = 0.001 mg/ℓ.

TABLE 9

MEAN POLLUTANT CONCENTRATIONS, IN mg/ℓ AT SITE 1
MARCH - APRIL, 1977

Outfall	005		010		011	
Pollutant	Dry	Wet	Dry	Wet	Dry	Wet
TSS	15	45	84	184	18	35
TDS	396	541	3078	2158	868	919
Total Iron			3.3	2.4	1.9	2.6
Dissolved Iron			0.4	0.1	0.1	0.2
Phenols			25	0.37	0.13	0.086
Cyanide (Total)			0.5	- (a)	- (a)	0.002
Ammonia			73	43	9.1	3.4
Sulfate			718	312		

(a) Several non-detectable values were also obtained.

TABLE 10

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MARCH - APRIL, 1977
OUTFALL 005 - SITE 1

Date	3/24 (Wet)			3/27-28 (Wet)			3/29 (Dry)			3/31 (Wet)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	39	1200 (317)	2.82 (6.2)	48	3845 (1016)	11.1 (24.4)	16	473 (125)	0.45 (0.99)	38	401 (106)	0.91 (2.0)
Total Dissolved Solids	938	1200 (317)	67.5 (149)	332	3847 (1016)	76.6 (168.5)	353	473 (125)	10.0 (22.0)	581	401 (106)	14.0 (30.8)

Date	4/4 (Wet)			4/5 (Dry)			4/16 (Wet)			4/18 (Dry)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	37	2434 (643)	5.46 (12.0)	17	227 (60)	0.23 (0.51)	75	4205 (1111)	19.0 (41.8)	14	227 (60)	0.19 (0.42)
Total Dissolved Solids	669	2434 (643)	97.7 (214.9)	443	227 (60)	6.0 (13.2)	371	4205 (1111)	93.6 (206.0)	409	227 (60)	5.6 (12.3)

TABLE 11

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MARCH - APRIL, 1977
OUTFALL 010 - SITE 1

Date	3/24 (Wet)			3/27-28 (Wet)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	76	12.7 (3.36)	0.06 (0.13)	1717	34.4 (9.1)	3.54 (7.80)
Total Dissolved Solids	2170	12.7 (3.36)	1.65 (3.64)	150	34.4 (9.1)	0.31 (0.68)
Total Iron	2.61	12.7 (3.36)	0.002 (0.004)			
Dissolved Iron				0.119	34.4 (9.1)	0.0002 (0.0005)
Phenol	0.46	12.7 (3.36)	0.0004 (0.0009)	0.559	34.4 (9.1)	0.001 (0.003)
Ammonia	50	4.43 (1.17)	0.013 (0.03)	41.0	34.4 (9.1)	0.08 (0.19)
Sulfate	285	14.7 (3.88)	0.25 (0.55)	224	34.4 (9.1)	0.46 (1.02)

TABLE 12

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MARCH - APRIL 1977
OUTFALL 011 - SITE 1

Date	3/24 (Wet)			3/27-28 (Wet)			3/29 (Dry)			4/5 (Dry)			4/16 (Wet)			4/18 (Dry)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	11	217 (57.29)	0.14 0.32	97	1764 (466)	10.3 (22.6)	14	38 (10)	0.03 (0.07)	14	3.4 (0.9)	0.003 (0.007)	51	568 (150)	1.74 (3.83)	30	87 (23)	0.16 (0.34)
Total Dissolved Solids	1343	217 (57.29)	17.5 (38.5)	624	1764 (466)	66.0 (145.3)	683	38 (10)	1.56 (3.43)	1021	3.4 (0.9)	0.21 (0.46)	1062	568 (150)	36.2 (79.6)	785	87 (23)	4.1 (9.0)
Total Iron	0.98*	282 (74.4)	0.02 (0.04)	5.8**	742 (196)	0.26 (0.57)	1.3	38 (10)	0.003 (0.007)	2.6	3.4 (0.9)	0.001 (0.002)	3.63*	935 (247)	0.2 (0.45)	1.9	87 (23)	0.01 (0.02)
Dissolved Iron	0.1**	275 72.8	0.002 (0.004)	0.1**	696 (184)	0.004 (0.009)	0.1**	38 (10)	0.0002 (0.0005)	0.1	3.4 (0.9)	2.0×10^{-5} (4.4×10^{-5})	0.2*	935 (247)	0.01 (0.02)	0.1	87 (23)	0.001 (0.002)
Phenol	0.117	262 (69.22)	0.002 (0.004)	0.038	1669 (444)	0.004 (0.009)	0.055	38 (10)	0.0001 (0.0003)	0.26	3.4 (0.9)	5.3×10^{-5} (1.2×10^{-5})	0.08*	477 (126)	0.002 (0.004)	0.025	87 (23)	0.0001 (0.0003)
Ammonia	1.49	252 (66.63)	0.023 (0.05)	13.04	742 (196)	0.58 (1.28)	22	38 (10)	0.05 (0.11)	4.9	3.4 (0.9)	0.001 (0.002)	0.912	568 (150)	0.03 (0.07)	0.97	87 (23)	0.005 (0.011)

*straight average
**one value only

each parameter was sampled and may vary for the different parameters within each storm event. In some instances, due to lack of data, straight average concentrations (or in some cases, one data point) were used to calculate wet weather average mass loadings. When no flow data were measured, mass loadings were not calculated.

At all outfalls the mean dissolved solids were higher than the suspended solids. At outfalls 005, 010, and 011, where automatic sampling was performed, the dissolved solids were consistently higher than the suspended solids, often by more than an order of magnitude. The reaction of dissolved solids varied with each outfall and each storm event. However, after plotting all the dissolved solids data and comparing these curves to the rainfall intensity and flow curves, no conclusion can be made concerning the reaction of dissolved solids to a storm event.

The reaction of total suspended solids to a storm event also varied with each outfall and event. In a few cases, suspended solids correspond directly to rainfall intensity and flow, but in most instances, there was a time lag between the rainfall intensity peaks and suspended solids concentration peaks.

The pollutant data from outfall 010 do show some interesting results. As indicated in Tables 8 and 9, the dry weather concentrations of total dissolved solids, total iron, dissolved iron, phenols, cyanide, ammonia, and sulfates were greater than the wet weather concentrations, indicating that the stormwater runoff at outfall 010 diluted these pollutants.

This same dilution effect was observed for phenol and ammonia concentrations at outfall 011, although the levels were much lower than those measured at outfall 010. The mass loading data indicate that the dry weather loading is at least one order of magnitude less than the wet weather loading of phenols. In most cases the same is true for the ammonia loadings.

From the limited data taken at outfalls 010 and 011, the concentrations of phenols exhibited a consistent pattern. Increases and decreases in measured phenols loading in these drainage basins correspond directly to increases and decreases in rainfall intensity with very little time lag.

In most cases, ammonia showed a general trend of decreasing concentration over the period of the storm, indicating that the stormwater acted to dilute the ammonia over the course of the storm rather than cause a "first flush" effect. In no case was the "first flush" effect observed.

3.1.2 Site 2 Results

Only two storm events occurred during the field program (May-June, 1977) which were of sufficient magnitude to produce surface runoff. Data from these events are summarized in Table 13.

TABLE 13

STORM EVENT DATA
SITE 2
MAY - JUNE, 1977

Date	Storm Beginning	Storm Ending	Total Rainfall		Average Rainfall Intensity		Maximum Rainfall Intensity During Storm	
			cm	(inches)	cm/hr	(in/hr)	cm/hr	(in/hr)
6/9-6/10/77	0500 (6/9)	1500 (6/10)	4.45	(1.75)	0.13	(0.05)	1.42	(0.56)
6/20/77	0900	2030	2.59	(1.02)	0.23	(0.09)	_[a]	_[a]

[a] No rainfall intensity data were collected on June 20 due to equipment failure and manpower constraints.

The first storm event started as a steady downpour which then tapered off to a drizzle with occasional heavy showers. Surface runoff was evident at all of the sampling locations. The rainfall intensity curve for this storm event is shown in Figure 1 along with the flow at outfall 007.

The second storm event was short in comparison to the first, but again resulted in a considerable amount of surface runoff at all of the sampling sites. This storm was also a heavy downpour. Due to manpower constraints and equipment failure, very little data except total rainfall and storm duration was gathered.

There were also several other small storm events which resulted in 1.3 cm of rain or less. Because most of the plant area is semi-permeable and level, surface runoff was not detected during any of these storms.

Table 14 shows the average flows and ranges of flow for several of the outfalls during dry and wet weather. Complete information exists only for outfalls 004, 006, and 007.

The flow data for storm events at outfalls 004, 006, and 007 show some interesting trends. The flow peaks at outfalls 006 and 007 corresponded very closely to rainfall intensity peaks with almost no time lag. Figure 1 also shows the hydrograph of June 9-10 for outfall 007. At outfall 004 the time lag between rainfall intensity peaks and flow peaks ranged from 0.5 to 3.5 hours. The difference was probably due to the type of drainage basin associated with each outfall. Outfalls 006 and 007 receive stormwater either directly from roof drains or from paved areas. The basin which drains to outfall 004 is a mostly unpaved (permeable) area, causing the time lag between rainfall peaks and runoff peaks.

Tables 15 and 16 show the range of concentrations and the mean concentrations of the pollutants analyzed at each of the outfalls for both dry and wet weather. Tables 17 through 19 indicate the average mass loadings of the pollutants analyzed at outfalls 004, 006, and 007 for both dry and wet weather conditions. Mass loadings were calculated in the same manner as at Site 1.

Total dissolved solids concentrations were much higher than total suspended solids concentrations at all of the outfalls during both dry and wet weather conditions with two exceptions, those being the wet weather concentrations at outfalls 006 and 011. A consistent pattern was established for total suspended solids. A direct relationship exists between TSS concentration and rainfall intensity corresponded directly to an increase in TSS concentration with no time lag. This is shown in Figure 2.

Several interesting trends occurred with total and dissolved iron. Six out of nine outfalls showed that an increase in rainfall intensity also corresponded directly to an increase in total iron concentration with no time lag. This was not true of dissolved iron, since five out of nine outfalls showed dissolved iron to vary inversely with total iron. As total iron concentration decreased, dissolved iron concentration increased and vice versa.

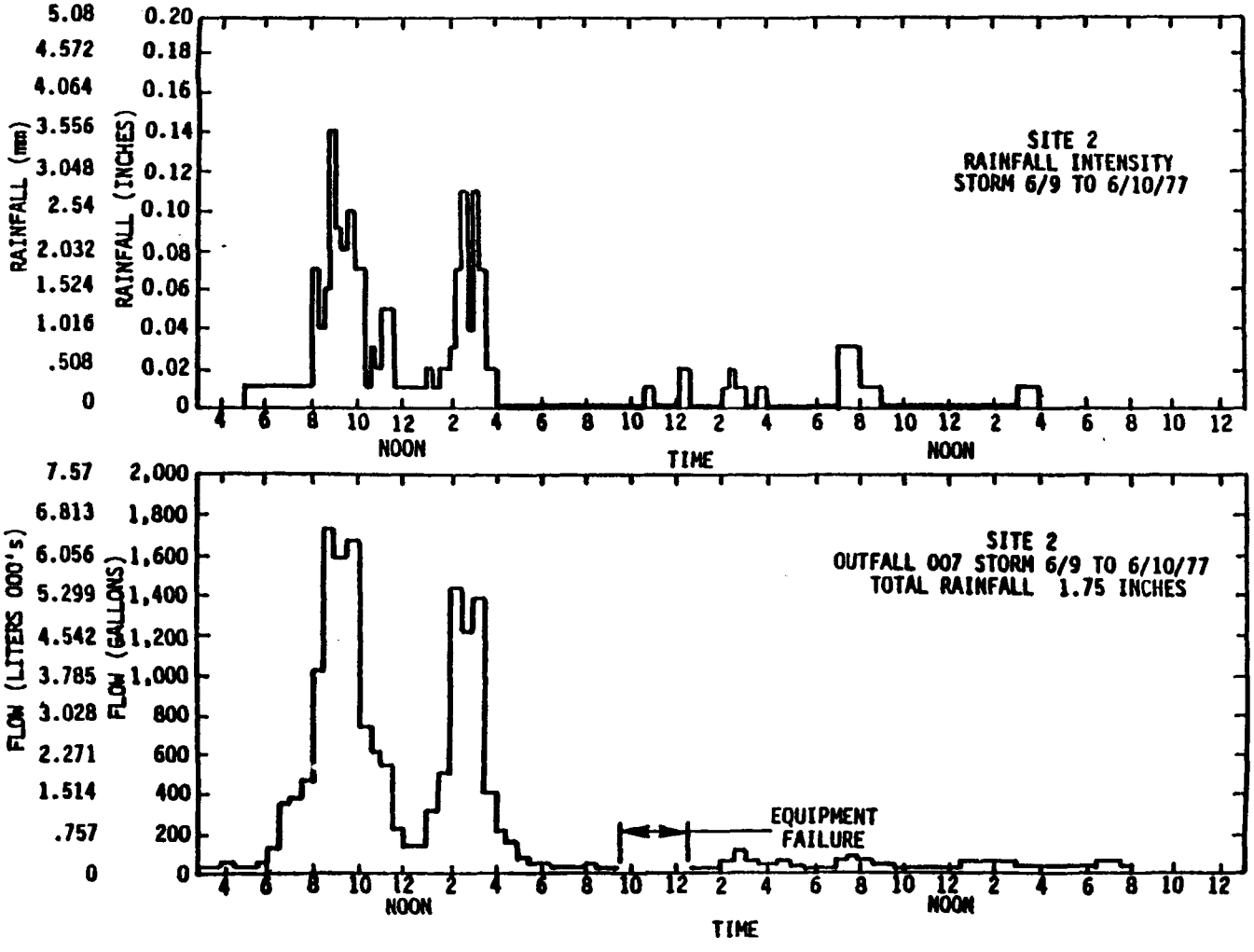

Figure 1: Rainfall and Flow in Basin 007 Versus Time, Site 2, 6/9 to 6/10/77. Storm

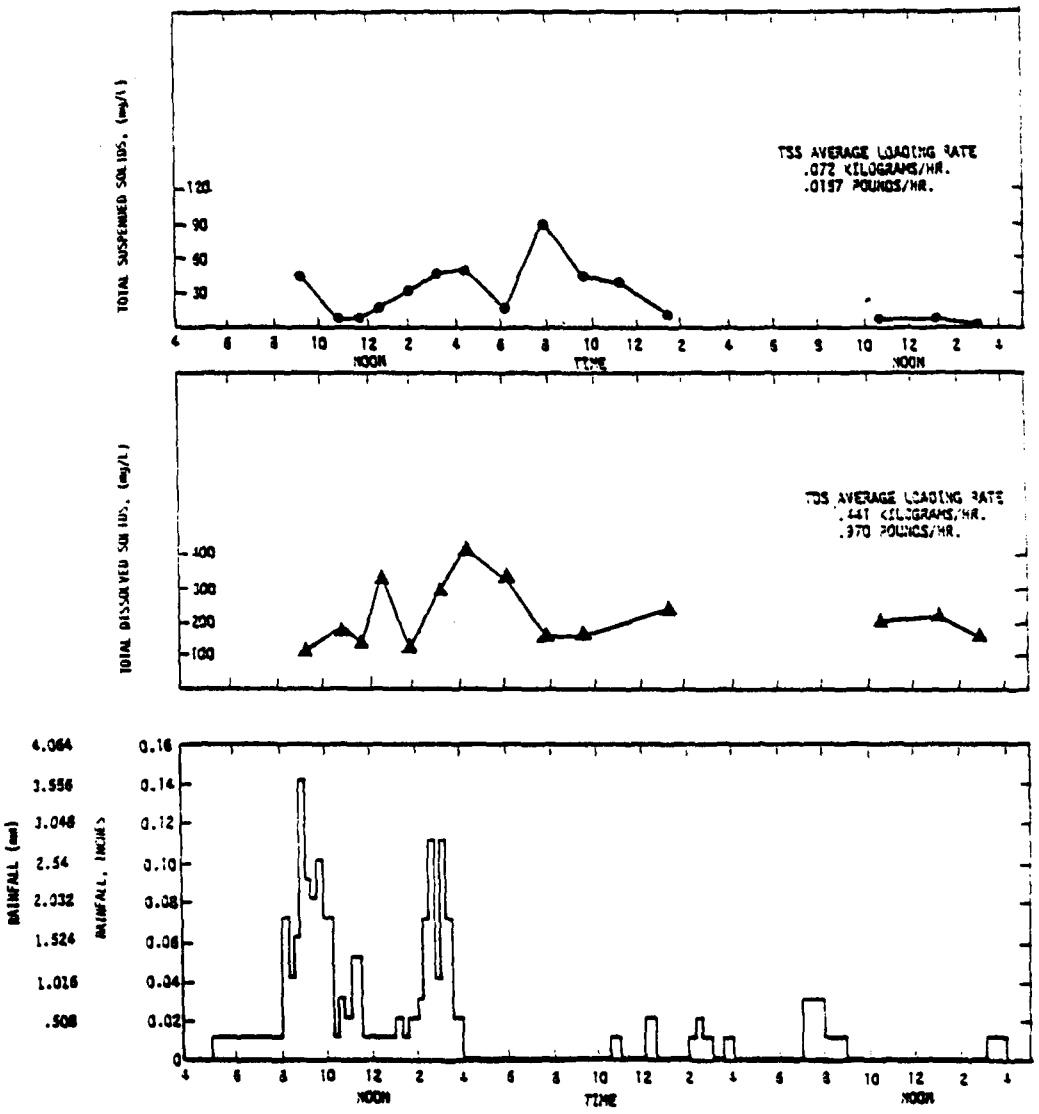

Figure 2: Outfall 007 - Site 2
TSS and TDS Concentrations Versus Time
Compared to Rainfall Intensity

TABLE 14

DRY VS. WET FLOWS[a]
SITE 2
MAY - JUNE, 1977

Outfall	Date	Sampling Condition	Average Flow lpm (gpm)	Range, lpm (gpm)
002[b]	5/10	Dry	-	-
	5/18	Dry	53 (14)	23-91 (6-24)
	6/9-10	Wet	-	-
	6/20	Wet	-	-
004[c]	5/10	Dry	163 (43)	132-310 (35-82)
	5/18	Dry	413 (109)	223-727 (59-192)
	6/9-10	Wet	549 (145)	163-988 (43-261)
	6/20	Wet	382 (101)	189-795 (50-210)
006[c]	5/10	Dry	1120 (296)	655-3410 (173-900)
	5/18	Dry	2150 (568)	1540-3293 (407-870)
	6/9-10	Wet	2498 (660)	730-4290 (193-1133)
	6/20	Wet	3066 (810)	1692-9463 (447-2500)
007[c]	5/10	Dry	4.5 (1.2)	4.0-4.9 (1.0-1.3)
	5/18	Dry	2.9 (0.8)	2.5-4.0 (0.7-1.0)
	6/9-10	Wet	45 (12)	1.1-216 (0.3-57)
	6/20	Wet	291 (77)	45-5776 (12-1526)
009[b]	5/10	Dry	-	-
	5/18	Dry	5344 (1412)	5223-5465 (1380-1444)
	6/9-10	Wet	-	-
	6/20	Wet	-	-
010A[b]	5/10	Dry	-	-
	5/18	Dry	1.08×10^5 (28570)	1.03×10^5-1.12×10^5 (27280-29580)
	6/9-10	Wet	-	-
	6/20	Wet	-	-
010B[b]	5/10	Dry	-	-
	5/18	Dry	5.14×10^4 (13590)	3.3×10^4-6.6×10^4 (8640-17480)
	6/9-10	Wet	-	-
	6/20	Wet	-	-

[a] Flow data were not collected at outfalls 003, 005, 008, 011, 012, 013, 014, and 015.

[b] Straight averages.

[c] Time-weighted averages.

TABLE 15

RANGE OF POLLUTANT CONCENTRATIONS AT THE
SAMPLING LOCATIONS AT SITE 2 IN mg/l
MAY - JUNE, 1977

POLLUTANT	Outfall 002 Dry	002 Wet	003 Dry	003 Wet	004 Dry	004 Wet	006 Dry	006 Wet	007 Dry	007 Wet	008 Dry	008 Wet	009 Dry	009 Wet
Total Suspended Solids	11-29	9-176		11-132	2-47	3-39	20-416	8-2537	1-60	3-119	22-56	19-89	4-58	55-109
Total Dissolved Solids	91-2019	112-284		93-148	113-205	160-359	102-159	145-490	54-245	107-418	112-172	224-265	116-138	151-251
Total Iron					0.20-1.2	0.18-2.2					1.0-2.2	2.8-7.5	0.78-1.4	3.0-5.2
Dissolved Iron[d]					n.d.-0.2	0.1-0.4					0.1-0.3	0.1-0.7	0.1-0.3	0.2-0.4

POLLUTANT	Outfall 010A Dry	010A Wet	010B Dry	010B Wet	011[a] Dry	011[a] Wet	012[a] Dry	012[a] Wet	013[a] Dry	013[a] Wet	014[a] Dry	014[a] Wet
Total Suspended Solids	5-37	3-48	13-28	12-702		56-2684		29-563		12-1380		24-121
Total Dissolved Solids	76-125	131-253	89-133	137-239		299-681		222-546		353-1690		232-524
Total Iron	0.57-1.3	0.93-5.9	1.5[b]	0.74-225		11-25		1.5-26		0.82-28		0.95-14
Dissolved Iron[d]	0.1	0.1-0.4	0.2[b]	n.d.-1.1		0.2		0.1-0.6		0.9-1.1		0.2-1.2
Phenol[e]	n.d.-0.01	n.d.-0.02	n.d.-0.01	n.d.-0.06		n.d.-0.02		0.01-0.19		0.01-0.04		
Cyanide(Total)[f]	0.01	0.01	n.d.-0.01	n.d.-0.02		(c) -		0.09-0.1		0.38-0.72		
Ammonia	3.8-8.6	0.1-0.7	7.1[b]	0.07-4.1		0.23-0.43		0.41-1.6		18-39		
Sulfate						195-270		52-128		36-190		

(a) No dry weather samples collected.

(b) Only one sample analyzed.

(c) Cyanide was not analyzed at this outfall.

(d) n.d.-not detectable. Detectable limit for dissolved iron is 0.02 mg/l.

(e) n.d.-not detectable. Detectable limit for phenol is 0.001 mg/l.

(f) n.d.-not detectable. Detectable limit for total cyanide is 0.001 mg/l.

TABLE 16

MEAN POLLUTANT CONCENTRATIONS IN mg/l AT SITE 2
MAY - JUNE, 1977

		Pollutant							
Outfall	Sampling Condition	TSS	TDS	Total Iron	Dissolved Iron	Phenol	Total Cyanide	Ammonia	Sulfate
002	Dry	20	749						
	Wet	47	178						
003[a]	Dry								
	Wet	21	110						
004	Dry	13	157	0.61	0.08				
	Wet	11	216	0.51	0.14				
006	Dry	96	130						
	Wet	298	223						
007	Dry	15	118						
	Wet	35	227						
008	Dry	44	149	1.6	0.21				
	Wet	45	241	5.2	0.2				
009	Dry	32	124	1.1	0.2				
	Wet	73	201	4.0	0.2				
010A	Dry	18	104	0.85	0.1	0.01	0.01	18.45	
	Wet	23	183	1.75	0.2	0.004	0.01	0.26	
010B	Dry	19	102	(b)	(b)	0.005	0.005	(b)	
	Wet	60	183	18	0.2	0.01	0.011	0.6	
011[a]	Dry								
	Wet	853	471	18	0.18	0.01		0.33	232
012[a]	Dry								
	Wet	257	360	11.4	0.2	0.04	0.2	1.0	78
013[a]	Dry								
	Wet	392	959	12.6	1.0	0.03	0.55	29.3	129
014[a]	Dry								
	Wet	64	416	5.8	0.5				

(a) No dry weather samples collected.

(b) Only one sample analyzed.

TABLE 17

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MAY-JUNE, 1977
OUTFALL 004 - SITE 2

Date	5/10 (Dry)			5/18 (Dry)			6/9-10 (Wet)			6/20 (Wet)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	9	163 (43)	0.09 (0.2)	15	413 (109)	0.37 (0.81)	10	662 (175)	0.4 (0.88)	9	401 (106)	0.22 (0.48)
Total Dissolved Solids	155	163 (43)	1.5 (3.3)	160	413 (109)	4.0 (8.8)	250	662 (175)	9.9 (21.8)	203	401 (106)	4.9 (10.8)
Total Iron	0.2 [a]	163 (43)	0.002 (0.004)	0.68	413 (109)	0.02 (0.04)	0.49	662 (175)	0.02 (0.04)	0.36	401 (106)	0.01 (0.02)
Dissolved Iron	n.d. [b]	163 (43)	-	0.06	413 (109)	0.001 (0.002)	0.11 (0.002)	693 (183)	0.005 (0.011)	0.04	424 (112)	0.001 (0.002)

[a] One value only.

[b] n.d.-not detectable. Detectable limit is 0.02 mg/l.

TABLE 18

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MAY-JUNE, 1977
OUTFALL 006 - SITE 2

Date	5/10 (Dry)			5/18 (Dry)			6/9-10 (Wet)			6/20 (Wet)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	41	1120 (296)	2.8 (6.2)	130	2150 (568)	16.8 (37)	495	1851 (489)	55 (121)	32	2824 (746)	5.4 (11.9)
Total Dissolved Solids	112	1120 (296)	7.5 (16.5)	148	2150 (568)	19.1 (42)	244	1851 (489)	27.1 (59.6)	186	2824 (746)	31.6 (69.3)

TABLE 19

AVERAGE MASS LOADINGS OF POLLUTANTS
DRY VS. WET WEATHER
MAY-JUNE, 1977
OUTFALL 007 - SITE 2

Date	5/10 (Dry)			5/18 (Dry)			6/9-10 (Wet)			6/20 (Wet)		
Parameter	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)	Avg. Conc., mg/l	Avg. Flow, lpm (gpm)	Avg. Mass Loading, kg/hr (lb/hr)
Total Suspended Solids	15	4.5 (1.2)	0.004 (0.009)	24	2.9 (0.8)	0.004 (0.009)	36	33.3 (8.8)	0.07 (0.15)	22	165.4 (43.7)	0.22 (0.48)
Total Dissolved Solids	149	4.5 (1.2)	0.04 (0.09)	84	2.9 (0.8)	0.01 (0.02)	222	33.3 (8.8)	0.44 (0.97)	244	165.4 (43.7)	2.42 (5.32)

Although there were only limited data for phenols, a pattern was observed similar to that at Site 1. Phenol concentration peaks were found to correspond to rainfall intensity peaks.

There appears to be no relationship between cyanide or sulfate concentration and rainfall intensity, although limited data prevent drawing any definite conclusions. No consistent pattern exists.

As at Site 1, ammonia concentrations tended to decrease over the period of the storm. Ammonia concentration peaked around the time of the first rainfall intensity peak and then slowly decreased throughout the remainder of the storm event. Apparently, the stormwater dilutes the ammonia rather than causing a "first flush" effect. In no case was the "first flush" effect observed.

4.0 SUMMARY OF FIELD RESULTS

Based on the data collected at the two sites, the coal and coke storage piles, and the coal and coke handling areas have the highest potential for contaminating stormwater. Table 20 is a summary of average concentrations of the various pollutants in these areas for the two mills sampled.

In order to determine the potential gross impact of stormwater runoff from the mills sampled, the stormwater runoff mass loadings were compared to the point source mass loadings which would exist under proposed BAT control.[5] Since BAT is EPA's next step in the control process (July, 1984), this comparison appears to be valid.

Table 21 compares selected annual and hourly runoff mass loadings to point source loadings based on proposed Best Available Technology (BAT) Effluent Guidelines for TSS. This table shows that TSS runoff loadings are generally higher than point source loadings. In addition to TSS, the field data indicate that runoff from coal piles could produce substantial mass loadings of ammonia, phenols and total iron.

In most cases at both sites, the parameter concentrations were rainfall intensity dependent (i.e., the concentration increased with increased rainfall intensity and vice versa). In some cases, the size and characteristics of the drainage basin had an effect on the time lag between rain intensity and runoff flow, and the time lag between runoff flow and parameter concentrations. Finally, the runoff data did not show a "first flush" effect.

TABLE 20

HIGHLIGHTS OF RESULTS
OF FIELD SAMPLING PROGRAMS

SITES 1 AND 2

MARCH-JUNE, 1977

Pollutant	Site No.	Potential Problem Areas	Average Wet Concentrations, mg/l
TSS	2	Coal Stor.	853
	1	Coke Stor.	505
	2		392[a]
	1	Coke & Coal Handling	184
TDS	2	Coal Stor.	471
	1	Coke Stor.	745
	2		939[a]
	1	Coke & Coal Handling	2158
TOTAL IRON	2	Coal Stor.	18
	1	Coke Stor.	32.3
	2		12.6[a]
	1	Coke & Coal Handling	2.4
DISSOLVED IRON	2	Coal Stor.	0.2
	1	Coke Stor.	0.09
	2		1.01[a]
	1	Coke & Coal Handling	0.12

Pollutant	Site No.	Potential Problem Areas	Average Wet Concentrations, mg/l
PHENOL	2	Coal Stor.	0.01
	1	Coke Stor.	0.06
	2		0.03[a]
	1	Coke & Coal Handling	0.37
AMMONIA	2	Coal Stor.	0.33
	1	Coke Stor.	2.1
	2		29.3[a]
	1	Coke & Coal Handling	43
CYANIDE	2	Coal Stor.	n.d.[b]
	1	Coke Stor.	0.01
	2		0.55[a]
	1	Coke & Coal Handling	n.d.[b]
SULFATE	2	Coal Stor.	232
	1	Coke Stor.	n.a[c]
	2		129[a]
	1	Coke & Coal Handling	312

[a] There were two sampling points near the coke storage area at Site 2. The average concentrations for only one (outfall 013) are shown.

[b] n.d. - none detected.

[c] n.a. - not analyzed.

TABLE 21

COMPARISON OF AVERAGE ANNUAL AND HOURLY POINT SOURCE LOADINGS WITH AVERAGE ANNUAL AND HOURLY RUNOFF LOADINGS OF TSS FOR SELECTED DRAINAGE BASINS

SITES 1 AND 2

March-June, 1977

Site	Outfall	Average Annual Loadings Based on BAT Effluent Guidelines[3,6] Kg/yr(lb/yr)	Average Annual Runoff Loadings Kg/yr(lb/yr)	Average Hourly Loadings Based on Maximum 1 Day BAT Effluent Guidelines[5] Kg/hr(lb/hr)	Rainfall Events Average Mass Loadings of Pollutants in Runoff Kg/hr(lb/hr)				
					3/24/77	3/27-3/28/77	4/16/77	6/9-6/10/77	6/20/77
1	009	—	TSS 3600 (8000)	—	—	—	—	—	—
	010	TSS 1850(4100)	TSS 80 (180)	TSS 0.6 (1.3)	TSS 0.06(0.13)	TSS 3.54 (7.8)		—	—
	011	TSS 1850(4100)	TSS 3315 (7290)	TSS 0.6 (1.3)	TSS 0.14(0.32)	TSS 10.3 (22.6)	TSS 1.74 (3.83)	—	—
	011 (Coal Pile)	—	TSS 4.1×10^5 (9.0×10^5)	—	—	—	—	—	—
2	010	TSS 1.8×10^4 (4.0×10^4)	TSS 1.5×10^6 (3.3×10^6)	TSS 6.0 (13)	—	—	—	TSS 219 (482)	TSS 136 (299)
	011 (Coal Pile)	—	TSS 7760 (1.7×10^4)	—	—	—	—	—	—
	012	—	TSS 310 (680)	—	—	—	—	—	—
	013	—	TSS 550 (1210)	—	—	—	—	—	—

5.0 IRON AND STEEL INDUSTRY CONTROL SYSTEMS

Stormwater controls which presently exist within the steel industry are limited. The only system specifically designed for stormwater control exists at Armco's Houston Works, where coal piles have been diked as a control measure for both fugitive air emissions and stormwater runoff. Runoff collected within the diked area flows by gravity to an earth pond. In nearly two years of operation, losses from evaporation and percolation have prevented any observed overflow from this pond. On dry days, 190,000 liters (50,000 gallons) of water (equivalent to 6 mm of rain) are sprayed on the coal piles to control fugitive dust emissions. This water is supplied from a separate concrete pump basin which receives water from the blowdown of a coke plant cooling tower.

Several mills contacted in this program collect stormwater runoff with process wastewater from certain mill areas and the water is subsequently treated at a terminal plant. This necessitates a system of combined sewers within the plant and in several cases a holding pond is needed prior to treatment to handle high flows from storms.

Many mills store their raw materials (predominantly iron ore) in concrete bunkers and bins. Some of these bunkers have concrete floors and stormwater has to be pumped out periodically. These bunkers were not installed for stormwater control but rather to guard against material loss; however, they can serve a control purpose by containing runoff which can then be pumped to a treatment system.

6.0 CONCLUSIONS AND RECOMMENDATIONS

The results of this program show that there are areas within iron and steel mills which may pose a problem with respect to contaminated stormwater.

From the field results at the two sites sampled we can conclude the following:

1. With the exception of runoff from coal and coke storage areas, the majority of the basins tested in the field survey had pollutant discharges which, on an annual basis, were less than the proposed BAT Effluent Guidelines for the point sources located within the basins. No data were obtained from iron ore and pellet storage piles and active slag dumps.

2. Runoff samples from the coal storage piles indicated TSS concentrations to be typical of urban runoff while TDS values were approximately twice typical urban runoff concentrations.

3. At both plants, runoff from coal and coke handling areas and the coke plant area generated higher hourly mass loadings of total suspended solids than the average hourly loadings for point sources based on maximum 24-hour loadings in the proposed BAT Effluent Guidelines.

4. The coal storage areas sampled in this study had much lower runoff concentrations for TSS, TDS, total iron, and sulfate than those found in runoff from utility coal containing higher percentages of sulfur.

Based on the program we recommend the following:

1. Site specificity should be the most important consideration when evaluating surface runoff problems at individual plants because many factors affect the runoff loadings, including:

    - o Plant activities and operations
    - o Climate
    - o Soil conditions
    - o Size of drainage basins
    - o Location of activities and operations relative to one another
    - o Neighboring industries and urban areas
    - o Proximity of plant to receiving waters
    - o Plant size

If stormwater runoff is found to be a problem at a specific site, more work should be performed to determine the feasibility of cost-effective controls for mill areas identified as having potential stormwater contamination problems.

2. At some plants, it may be beneficial to treat stormwater from certain areas to bring runoff mass loadings down to the same order of magnitude as point sources based on the proposed BAT Effluent Guidelines. The most likely area is coal piles where it may be beneficial to treat runoff for total suspended solids, total dissolved solids, and total iron.

3. Future studies should be directed to quantify and qualify stormwater runoff from the iron ore storage and slag disposal areas.

4. Because many steel plants are built on permeable soils next to waterways, groundwater contamination from storm events is possible. Future programs should investigate potential groundwater contamination from the industry.

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4. Kaiser Steel Corporation, Fontana, California, Annual Summary of Surface Runoff performed for the California Regional Water Quality Control Board, 1975.

5. Code of Federal Regulations, Title 40 - Protection of Environment, Part 420 Iron and Steel Manufacturing Point Source Category, Effluent Guidelines and Standards as of July 1, 1976.

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Differential Tracing of Oily Waste and the Associated Water Mass by Tagging with Rare Earths

by

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Abstract

A method was developed for tagging oily waste with dysprosium and the associated fresh water with samarium. Neutron activation analysis permitted determination of rare earth concentrations as low as 40 ng/L in 15-mL water samples. Shipboard sampling procedures were developed that allowed measurement of the three-dimensional distribution of the spreading wastes and associated water. The method was applied in two field experiments that involved tracing oily wastes and polluted water from the Indiana Harbor Canal (IHC) into Lake Michigan.

For a summer, floating-plume experiment, about 1400 shipboard samples were collected. Employment of the dual-tracer technique led to the following results: (1) after artificial mixing into the water column by a passing ship, the tagged oil did not immediately resurface, and (2) there were no distinguishable differences between the movement of the oil and water over 4 km of travel.

During a winter, sinking-plume experiment, 1200 lake-water samples were collected from a boat and from the raw-water intakes of Chicago's South Water Filtration Plant (SWFP). These data provided positive evidence of the intake of IHC effluent and oily waste at the SWFP. The different tracers for the oily waste and underlying water gave evidence of separate pathways to the SWFP, reflecting differing transport modes for surface and near-bottom waters.

INTRODUCTION

It is important to distinguish between the dynamics of waste-receiving waters and the motions of the associated oily wastes within and upon these waters. Adequate understanding of these separate but complementary motions is essential for the development of predictive models. A method was developed for simultaneously tagging both oily wastes and the underlying water, each with a unique tracer, and for determining their individual motions in fresh water.

Traditionally, two classes of substances have been used in tracer studies to tag water or pollutants in water: fluorescent dyes and radionuclides. The tracing approach in the present study uses Rare Earth Element (REE) tags and neutron-activation analysis (NAA) for tag detection. Among the advantages of this method are the following:

1. No radiation hazard to the environment exists.
2. Detection of REE concentrations as low as 6×10^{-10} g in a 15-mL sample is possible.
3. Tracer loss to suspended matter and sediments appears negligible.
4. Rare earth element (REE) tags can be selected that are non-toxic to the environment.
5. Detection and quantification of several REE tracers simultaneously is simplified.

Two tags were needed to trace simultaneously oily wastes and the waste receiving waters. The rare earth elements dysprosium (Dy) and samarium (Sm) were chosen as the most suitable for the present study because

1. They have high detectability and short half-lives, and
2. Their natural occurrence in the coastal waters of Lake Michigan is at concentrations below the limits of detectability provided by the methods used in the present study. Water sample analysis (forty-eight 15-mL raw-water samples) showed that no natural Dy or Sm could be detected.

Application of the tracing technique by Argonne National Laboratory (ANL) was focused on a series of experiments conducted on the movement of oily wastes in the coastal waters of southwestern Lake Michigan from the Indiana Harbor Canal (IHC)(Fig. 1). Several possible sources of oily pollutants exist in the canal. Effluents from an oil refinery, steel mills, and municipal sewage treatment plants contain such pollutants (Snow, 1974). Small spills from oil transfer facilities and runoff from industrial sites also contribute to the oily-waste loading. For this study no individual source was tagged, but rather a simulated oily waste was prepared and spilled in the canal.

The source of water in the IHC is Lake Michigan. Industry draws water

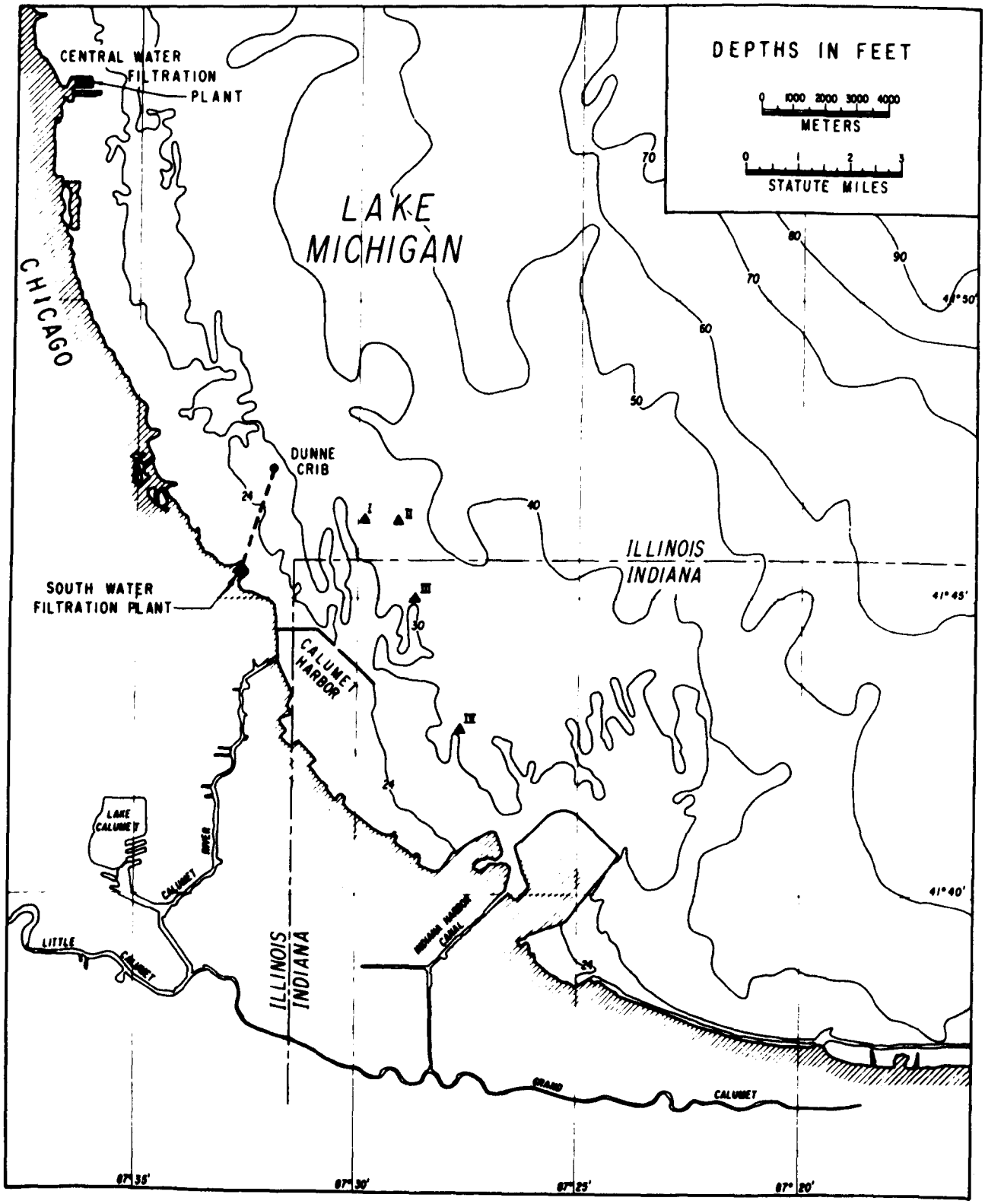

Fig. 1. Location Map for Study Area and Current-Meter Positions (I-IV) in Southwestern Lake Michigan.

from the lake, uses the water for industrial cooling processes, and then returns the water to the canal 5-8 C° warmer than the lake. The canal water moves into the lake as a thermal plume, sinking during much of the time in winter, and forming a surface plume during the remainder of the year. Significant differences in the oil-transport regime are expected during floating- and sinking-plume conditions. The most difficult transport/dispersion regime to observe in the field is that of the sinking plume, because it is difficult to follow and sample a tagged mass of canal water that forms only a thin layer (commonly 1- to 5-m thick) as it spreads over the bottom. Nevertheless, the importance of sinking-plume transport in carrying contaminants from the IHC to municipal water intakes in southwestern Lake Michigan dictated devoting significant efforts and resources to the tagging and tracing of IHC water during sinking-plume conditions.

When the wind is from the southern quadrants, during the winter sinking-plume conditions, the City of Chicago's South Water Filtration Plant (SWFP) experiences periods of high hydrocarbon odors that require using expensive activated charcoal in the treatment process. Prior to this study, it was thought that the odors came from IHC effluent which had migrated northwestward along the lake bottom (Vaughan, 1970) passing over the water intakes of the SWFP; however, it had not been proved. This study provides concrete evidence that IHC effluent enters the raw water intakes of the SWFP.

EXPERIMENTAL PROCEDURES

Tagging Considerations

Dysprosium (Dy) was the agent chosen to tag the simulated oily waste, and samarium (Sm) was selected to tag the IHC water. A third agent, the fluorescent dye rhodamine-WT, was also used to tag the canal water so that, with the use of a pump and fluorometer, the tagged water mass could be followed by the sampling boat.

Waters of the IHC are turbid and highly polluted. Introduction of an REE tag into the IHC water column in the ionic form would hazard its removal from solution by the formation of insoluble compounds by reaction with some of the many species in solution in the IHC or on the bottom of the canal, and adsorption on suspended solids.

Channell (1971) showed that the persistence of lanthanum complexed with EDTA (Ethylenediaminetetraacetic acid) in waters that are in contact with bottom sediment is almost twice that for lanthanum dissolved in acid. In view of Channell's findings, chelation of REE tags was considered necessary. The water tracer, Sm, was complexed with DTPA (Diethylenetriaminepentaacetic acid). DTPA was used because it was cheaper and was easier to complex with Sm than EDTA.

Numerous bench tests (>50) were conducted to determine how well the simulated refinery/steel-mill oily waste retained the Dy tag. The simulated waste was made up of equal parts of 30-W motor oil, #2 diesel fuel, and engine drain oil. The results of the bench testing are discussed in detail in McCown et al. (1978) and McCown et al. (1976). A "loss factor" of 0.4% loss of Dy/mL of tagged oil/mL of receiving water was determined from the bench tests.

Sampling Procedures

Documentation of transport and mixing of a plume requires many data points. This requirement dictates the removal of water samples from several depths and at many locations in the lake. In addition, the position of the boat when samples are being taken must be known to a high degree of accuracy. Therefore, a three-dimensional water-sampling system was designed that included:

1. A Motorola Mini-Ranger microwave ranging system interfaced with an x-y plotter for real-time positioning,
2. A fluorometer, pump, and hoses to follow the dye patch, and
3. A dynamically depressed faired cable with attached small tubes that extend to the depths to be sampled.

When tracking a sinking plume, an additional large tube was placed in the faired cable, which was used to draw bottom water samples. A schematic of the sampling system, as set up for a sinking plume, is shown in Fig. 2.

The intermediate-depth water samples were pumped onboard, through 4.7-mm I.D. nylon tubes, by a Masterflex multichannel tubing pump. The flow rates, through ≈20 m of tube, was 1.67 mL/s. This relatively low flow rate was sufficient because the volume of each sample was only 15 mL. The intermediate-depth tubes were all the same length, so that sample-removal delay times were equal.

The bottom sample collected for the sinking-plume study was drawn through a 9.6-mm I.D. nylon tube, which was connected to the faired cable at a point 1 m above the depressor fin (Fig. 2). The end of the bottom-sampler tube was connected to a 7.3 kg (16 lb) shot that trails the depressor fin by ≈12 m so that the shot drags the bottom. The bottom sampler had a relatively high flow rate of 12.5 mL/s and was driven by a positive-displacement pump. Water from the bottom was fed directly to the fluorometer. A Valved-tee at the fluorometer inlet directed a small portion (3.33 mL/s) of water to the sampling manifold for subsequent NAA at Argonne. All the samples for NAA were drawn in new, 15-mL lab-grade polyethylene Polyvials.

In addition, a surface-skimming water sampler was built; however, no acceptable means could be determined for calibrating it. Samples drawn by the surface skimmer were collected in the field, but the results of these samples are presented in only one instance (see discussion on Fig. 7 in section on Sinking-Plume Results) as an indication of tracer presence or absence.

During the sinking-plume study water samples were also drawn from the raw-water streams of the shore and crib intakes at the SWFP.

NAA Procedures

The collected water samples were analyzed at ANL's CP-5 reactor. Complete details of the analytical procedures and an error analysis are discussed in McCown et al. (1978). The significant features of the NAA procedures, developed at ANL, are the following:

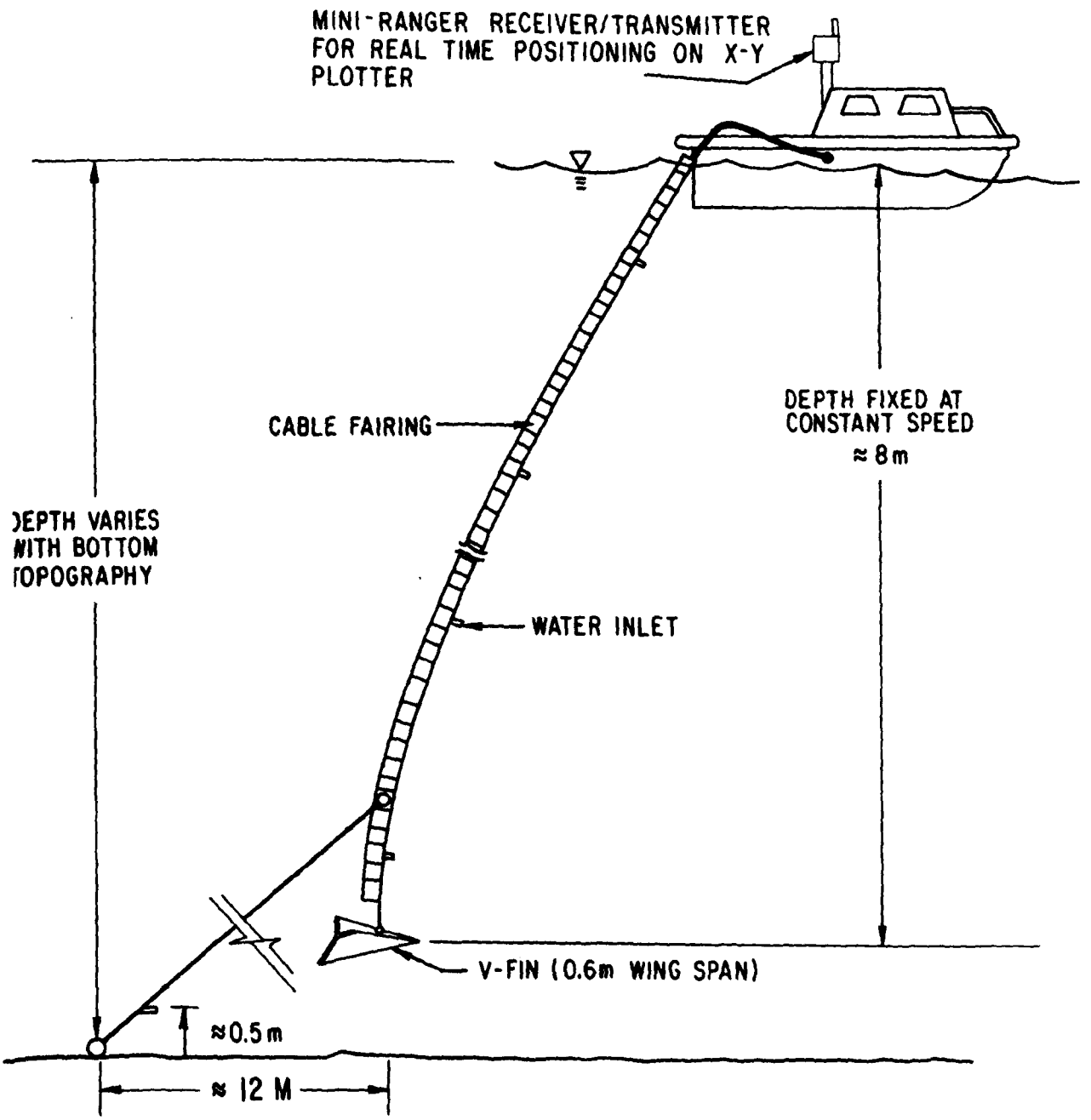

Fig. 2. Schematic of the Three-Dimensional Underway Water-Sampling System as Set Up for a Sinking-Plume.

1. No pre-irradiation sample preparation is necessary except evaporation of the liquid phase and cleaning of the outside of the Polyvial,
2. The samples are irradiated in the same Polyvials in which they were collected,
3. Seventeen samples per hour can be analyzed by the method,
4. The per sample cost of analysis, including personnel, is $5.00, and
5. Minimum detectability of 6×10^{-10} g for Dy and 2×10^{-8} g for Sm is possible with samples of only 15 mL.

Current Measurements

For the sinking-plume experiment, lake-current data were gathered at the four stations (I-IV) shown on Fig. 1. The moorings were placed in about 10 m of water, and a Bendix Q-15R current-meter sensor was positioned about 1.5 m above the bottom at each station; at Station I, a second meter was positioned about 5 m above the bottom in order to obtain information on the vertical structure of the currents. Detailed information on current-meter mooring design, recording methods, and processing is contained in Harrison, et al. (1977).

In-situ current-meters were not placed for the floating-plume experiment because of the relatively short duration and limited horizontal extent of the experiment. ANL current-meters were moored offshore of the Calumet Area for a different study so that general nearshore current information was available.

RESULTS AND DISCUSSION

Floating-Plume Results

On September 26, 1976, the IHC water was tagged *in-situ* with 4.5 kg (10 lb) of samarium that ha' been complexed with DTPA, and with 3 kg of rhodamine WT dye. The samarium/DPTA/Rh-WT tag was dispersed into the top meter of IHC water by pumping the tag through a pipe with many small holes drilled along one side. The tagging process required 10 min and occurred in the center of the canal at a point about 500 m from the lakeward entrance (Fig. 3).

The Dy-tagged, simulated oily waste was poured on the surface of the IHC effluent at the same time and in the same location as the in-situ water tag (Sm). The oily-waste and tracer consisted of 57 L of the waste mixed with 0.45 kg (1 lb) of Dy. The Dy had been dissolved in a 50% acetic acid solution.

Shortly after tagging and before sampling had commenced, an empty ore carrier passed directly through the center of the dye patch. Winds on September 26, were light from the south. There were no significant waves, and the lake current was <0.05 m/s flowing to the northwest. The plume that resulted from the release of the tracers and subsequent mixing by a passing ore carrier was mapped four times in the next 10 hr, and over 1300 15-mL water samples were collected. Sampling depths were 0.5, 1.0, 1.5, 2.5, and 3.5 meters. About 800 of the collected water samples were analyzed by neutron activation. The lowest Dy concentrations that were contoured at the 0.5 m level (80 ng/L = 200 counts) for each of the four plume mappings are plotted in Fig. 3.

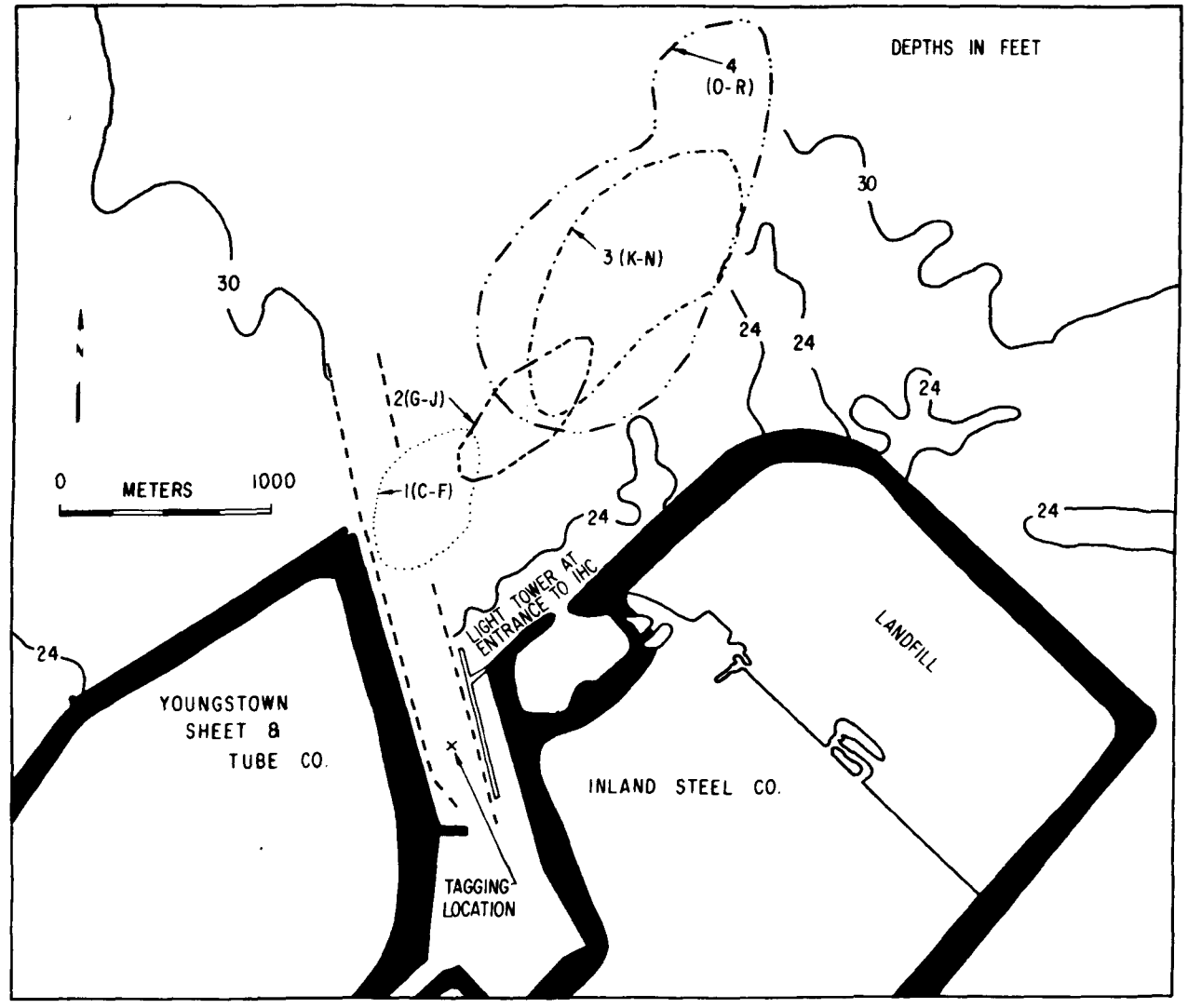

Fig. 3. REE Tagging Location and Contours of Lowest Detectable Amount of Dy that was Contoured for Plume Mappings 1-4, Floating-Plume.

Table 1 lists the elapsed time after tagging, the percent of the original Dy that was accounted for in each plume, the average concentration integrated over the depth, and the average dilution ratios relative to plume 2.

Table 1. Floating Plumes 1-4 Average Concentration Parameters

Plume	Elapsed Time After Tagging(s)	% Dy Accounted For	Average Concentration Over the Depth (Kg/m^3)	Dilution Compared to 2
1	4.8×10^3	131[a]	4.8×10^{-7}	-
2	1.1×10^4	83	4.6×10^{-7}	1:1
3	2×10^4	77	2.2×10^{-7}	2:1
4	3×10^4	67	4.9×10^{-8}	10:1

[a]High percentage due to non-synopticity of measurement. (See text for explanation.)

Diffusion coefficients were calculated using elapsed time and observed concentration variances for the last two cloud mappings (3[K-N] and [O-R]) at the 0.5 m level. These two mappings were in Lake Michigan proper and thus would be subject to lake-type diffusion. The calculated horizontal diffusion coefficients are shown in Table 2 with appropriate values reported by Murthy (1976) for Lake Ontario. The values of these diffusion coefficients indicates that our data are similar to data from the Great Lakes found using dye as the tracer.

Table 2. Calculated Horizontal Diffusion Coefficients (m^2/s)

	Dy/Oil		Sm/Water		Murthy's Values[a]	
Time	K_y[b]	K_x[c]	K_y	K_x	K_y	K_x
2.5×10^4 sec	2.2	8.4	0.2	6.9	1.5	10.5

[a]Lake Ontario (dye)

[b]Longitudinal

[c]Lateral

The percent of the Dy/oil and Sm found above 1.25 m (based on the total amounts accounted for), is plotted as a function of time after tracer release in Fig. 4. Each separate plume mapping is indicated by time in seconds on the bottom horizontal axis. The transition from the area protected by the Inland Steel Landfill to the nearshore zone of Lake Michigan is indicated by the vertical dashed line.

Floating-Plume Discussion

The Dy loss factor (0.4% Dy lost/mL oil/mL water) that was determined above may be used to estimate if significant amounts of Dy dissociated from the oil during the floating-plume study. The Dy is detected in plume 3 over a volume of 1.6×10^9 L (400m x 1000m x 4m). That volume would indicate only a 4% loss of Dy from the oily waste. Therefore, the loss of Dy from the oily waste into the water through plume 3 is not thought to be significant.

In plume 4, Dy can be detected over a volume of 6.2×10^9 L (600m x 1000m x 4m) for a maximum Dy loss from oil of 15%. It is doubtful if even a 15% loss would significantly affect the trends exhibited by the results.

Note that the Dy tracer loss from the oily waste discussed above is not necessarily related to the "percent Dy accounted for" in Table 1. If some of the Dy were lost from the oily waste, it could remain in nearby waters and thus be sampled and detected.

The "percent Dy accounted for" was determined by integrating the concentrations at sampling points over the three-dimensional structure of the measurable plume and comparing that weight with the original amount of Dy in the oil spilled. Deviations from 100% are probably due to the spacing of the grid and the diffusion of Dy to undetectable concentrations at the plume edges. Plume 1, which shows 131% of the original Dy accounted for appears strange, but the seeming disparity is due to the non-synopticity of measurement of that plume. Plume 1 was sampled in a region where the flow of the canal was relatively fast and the direction of movement was changing from directly out of the canal to a more lakeward heading. Consequently, measurements of plume 1 were the least synoptic of the four measured plumes.

Figure 4 shows that the amount of Dy and Sm above 1.25 m changes with time and horizontal position. Both Sm and Dy migrate toward the surface until the plume passes into the nearshore zone of Lake Michigan. After passing into the nearshore zone, the plume mixes downward.

The initial movement of both tracers seems to have occurred because warm canal water flows out and rises over the inflowing, colder Lake Michigan water. However, when the IHC plume enters Lake Michigan proper, it is then subjected to large-scale, lake-type diffusion and it is more dense than initially due to cooling. These two factors cause the downward migration of the plume when it enters the lake proper.

After the oily wastes were mixed into the water column, their movement did not differ significantly from the underlying Sm-tagged waters. As indicated above, mixing coefficients for the Dy/oil and Sm/water are similar.

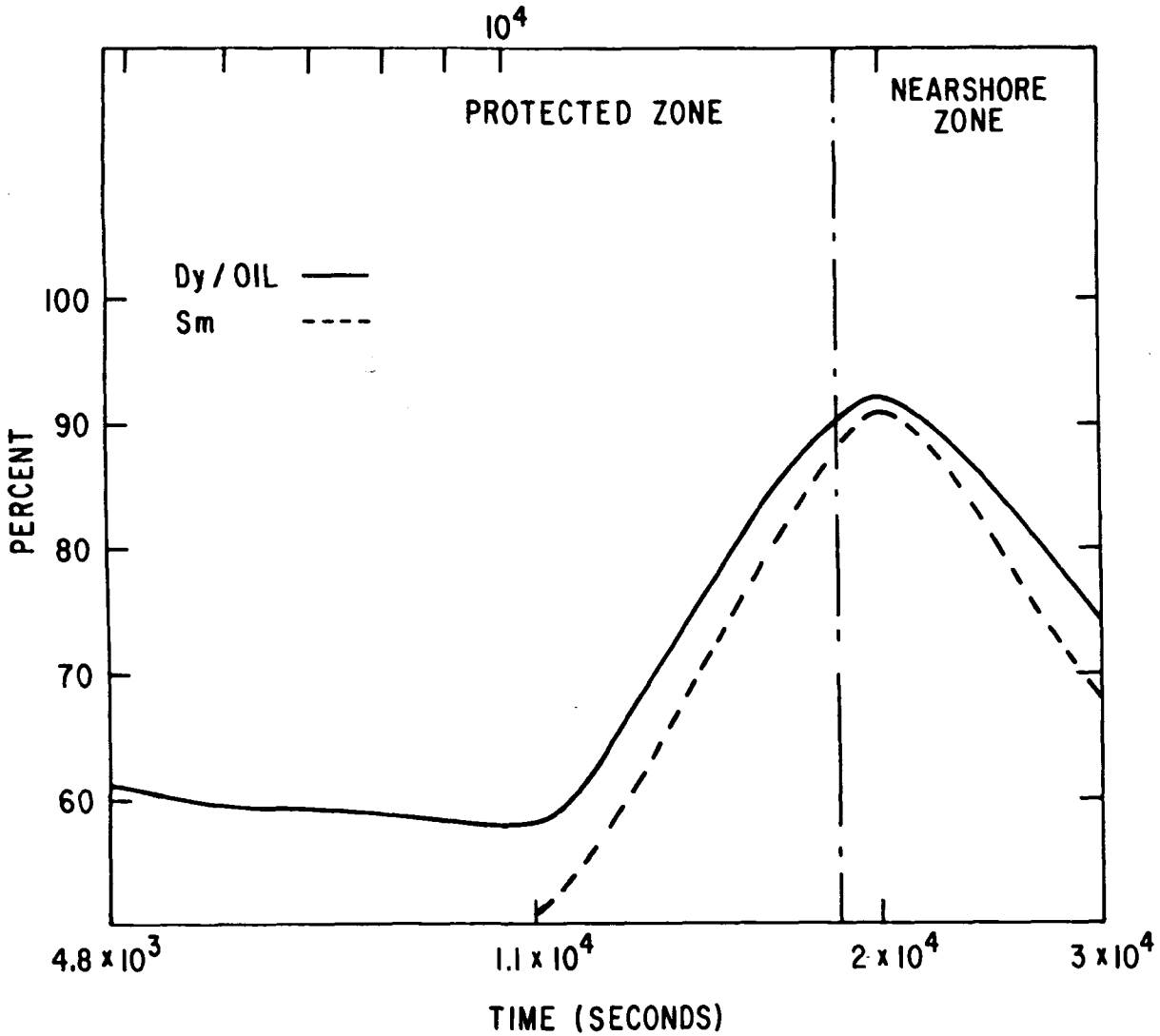

Fig. 4. Percent of the Total Dy/Oil and Sm Accounted for by Measurement Found above 1.25 m vs Time after Tracer Release.

Moreover, Fig. 4 indicates no significant differences in the gross vertical movement of the Dy/oil and Sm/water.

The above results indicate that the oily waste remained suspended in the water column and did not float back to the surface. The tendency for oils to remain in suspension and not float back to the surface has also been noticed and documented by McAuliffe (1977), Shaw (1977), Brown and Huffman (1976), Brown and Searl (1976), Peake and Hodgson (1966, 1967), and Gordon et al. (1973). Their results may be summarized as follows:

1. Under agitation, oils break up and disperse into the underlying waters where most remain suspended and move with the water, and
2. Colloidal particles (10^{-9} to 10^{-6} m) remain in suspension for extended periods.

A distinct difference exists between the small, limited spill (57 L) artificially mixed into the water column discussed in this study and a major spill involving millions of liters. Extrapolations from the results above to the case of a major spill may not be valid, and different relative motions between the oil and underlying water might be expected for a major spill. This would be particularly true during the initial phases of spreading of a major spill where gravity, viscosity, and surface tension are dominant. However, when that type of slick becomes thin and oil mixes into the water column, behavior similar to that reported in this study might be found.

Sinking-Plume Results

On March 2, 1977, IHC water was tagged *in-situ* with 13.6 kg (30 lb) of samarium that had been complexed with DTPA, and with 7.5 kg of rhodamine WT dye. The samarium/DPTA/Rh-WT tag was dispersed in the same manner as for the floating plume. The tagging process required 10 min and occurred at the "REE tagging location" (Fig. 5), which was approximately 900 m upflow from the point where the canal water was sinking. The zone of convergence of canal and lake water was easily determined with a temperature probe and is indicated in Fig. 5 as "surface temperature convergence." As the survey vessel NEPTUNE proceeded lakeward, the surface water temperature dropped from 7.5 to 1.8°C within a few meters. Canalward of the sinking zone, the water temperature was vertically isothermal. Just lakeward of the sinking zone, however, the temperature profile showed warm (≈4.0°C) water near the bottom, overlain by colder (≈1.8°C) water.

The Dy-tagged simulated oily waste was poured on the surface of the IHC effluent at the same time and in the same location that the in-situ water tag (Sm) was injected. The oily waste and tracer consisted of 170 L of the waste mixed with 1.4 kg (3 lb) of Dy dissolved in a 50% acetic acid solution.

A progressive vector diagram of local winds and bottom currents is shown in Fig. 6 for the period March 1-4 for the winds and March 1-5 for the bottom currents. The current meters were deployed for this study for the period January 5 to March 26, 1977. Complete details of the current structure during the period are published in Harrison et al. (1977). In summary, major velocity components are well-correlated within the study area for the period of measurement and suggest the existence of a rather uniform flow field.

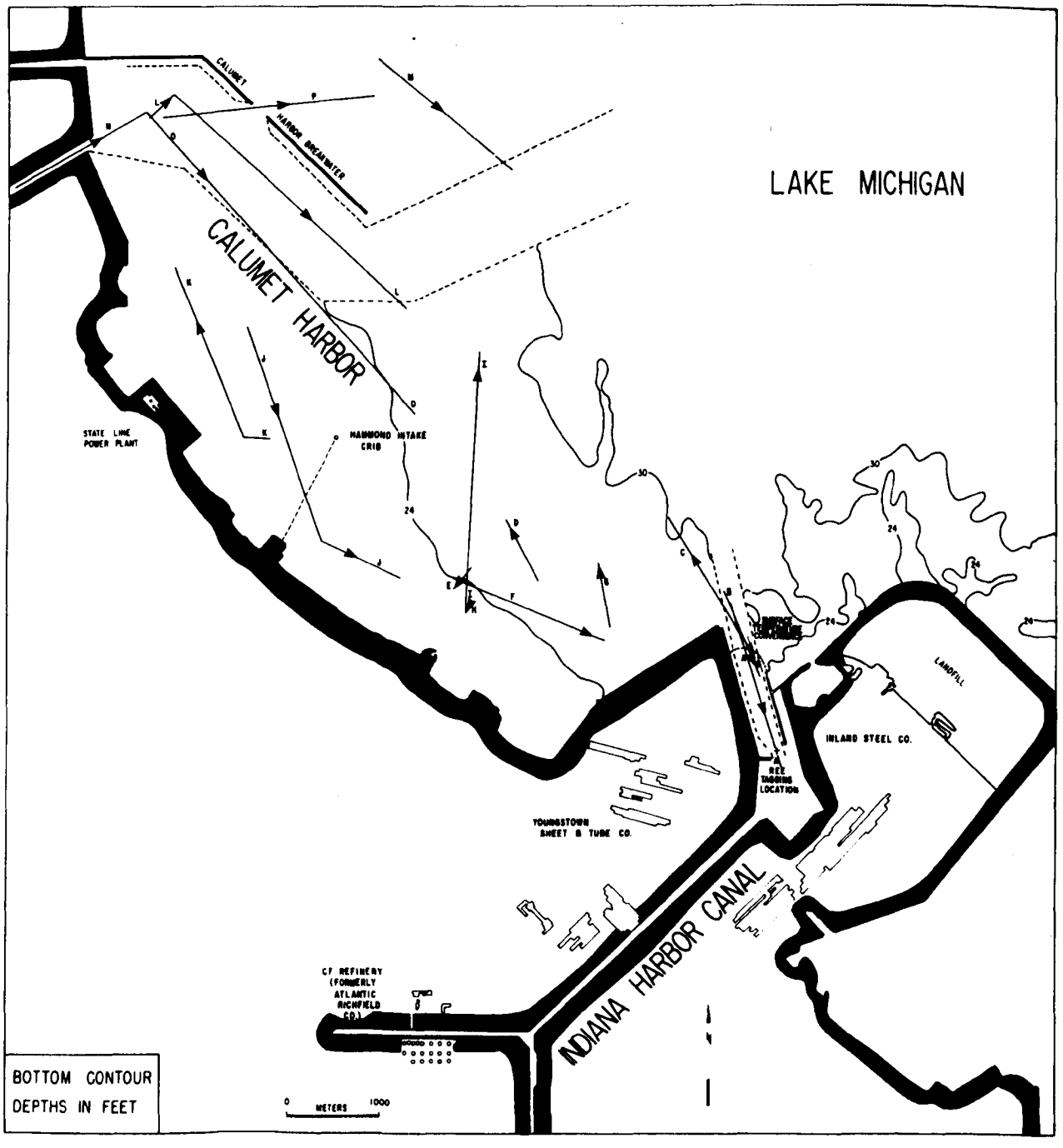

Fig. 5. REE Tagging Location, Temperature-Convergence Zone, and Positions of Sampling Transects, Sinking-Plume.

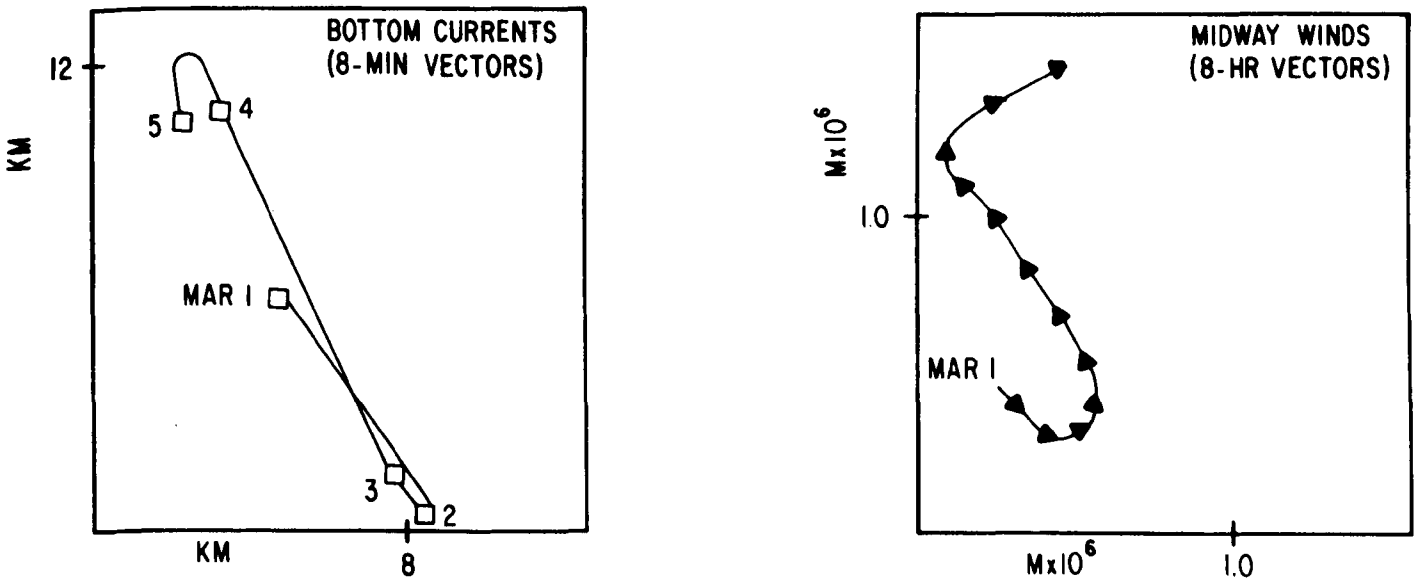

Fig. 6. Progressive-Vector Diagrams for March 1-4, 1977, Midway Airport Winds, and March 1-5, 1977, Lake Michigan Bottom Currents at Station III (Fig. 1).

About 1050 water samples were collected in Polyvials aboard the NEPTUNE while tracking the dye that was used to tag the IHC effluent. Of the 1050 samples approximately 200 were collected in Transects A, B, and C (Fig. 5) in the canal entrance and near entrance areas. All 200 were analyzed by NAA for their Dy and Sm concentration. Of the remaining 800 samples collected in the Calumet Harbor area, 350 were analyzed (that is every other or every third sample along a given transect was analyzed unless a high value was observed, in which case samples on either side were analyzed) and only ten of those indicated the presence of Sm (canal water); however, many of the samples from the Calumet Harbor indicated the presence of Dy (simulated oily waste).

All sampling transect locations are plotted on Fig. 5. Each transect has an arrow indicating the direction of boat travel, and the ends of each transect correspond to the first or last sampling station. The distance between sampling stations was about 100 m.

Figure 7 is a plot of the vertical Dy concentration encountered in Transect A that passed directly through the convergence zone. The sampling stations are numbered in ascending order corresponding to the direction of boat travel. Contour values for Dy in the figure are nanograms per liter

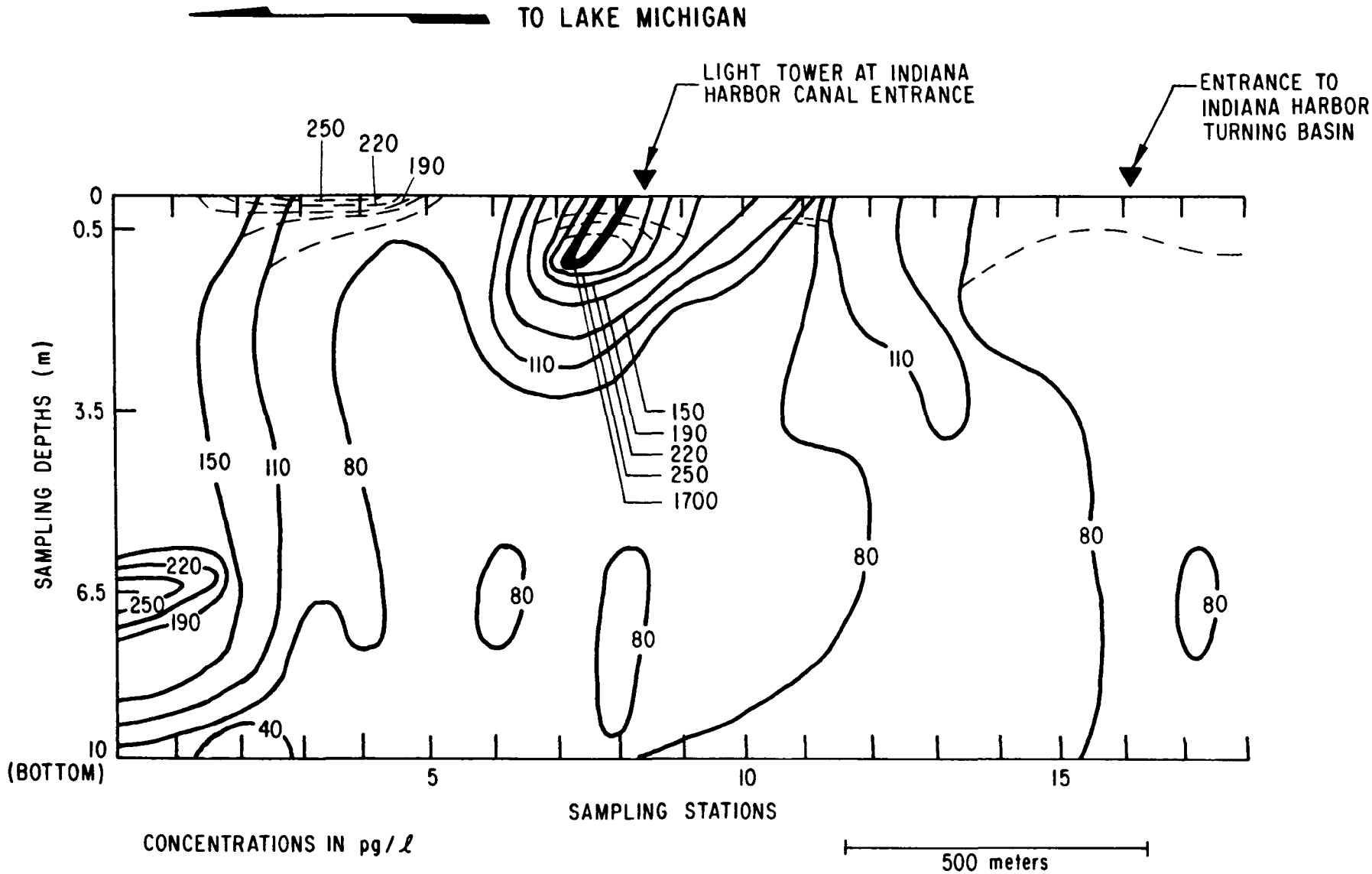

Fig. 7. Transect A: Dy Concentrations. (All 90 samples collected were analyzed. The dashed contours indicate how the figure would look if concentration values for the surface samples could have been considered.)

(ng/L); that is, 10^{-9} g/L. A region of sinking Dy/oily waste is clearly shown between stations 1 and 5. The dashed contours indicate how the data would have been contoured if data from the surface sampler were considered and showed that some of the Dy/oil crossed the sinking zone on the surface.

Plots of all other transects and further explanation of the data are in McCown et al. (1978). In summary, all of the transects in the Calumet Harbor area indicated fairly uniform vertical mixing of the Dy tracer. In a number of instances, a singular point occurred having a very high Dy concentration, probably due to a large oil particle in the sample.

About 240 water samples were drawn from the raw-water streams for the shore and crib intakes at the SWFP. These 15-mL samples were drawn every 10 min for almost all the period between 2200 hr on March 3 to 1240 hr on March 4, 1977. From 1240 hr to 1600 hr March 4, 1977, samples were drawn every 20 min. Water-intake samples were drawn in the same type of new 15-mL Polyvials that were used for the shipboard samples.

All 240 of the samples drawn at the SWFP were analyzed by NAA and those that contained detectable Sm and/or Dy are shown on Fig. 8, a plot of relative amounts of tracer as a function of time for each raw-water intake. Relative amount is computed as the amount of a given REE found in a Polyvial sample divided by the quantity of that REE that was released in the IHC effluent times 100.

Sinking-Plume Discussion

Results of the winter plume-tracking field study show that the effluent from the IHC enters the City of Chicago's water-purification system at the SWFP's Dunne Crib and shore intakes. (Any contaminants entering the Chicago water system are removed at the filtration plant by activated charcoal treatment, but the removal process is expensive.) A detailed picture of the effluent's path to the crib or shore intake cannot be established from the tracer data because no measurements of the plume were made in the vicinity of the intakes. However, two paths may exist for transport of IHC effluent to the SWFP's water intakes. Each path is governed by the nature of entry of IHC effluent into the lake, incompletely mixed near-surface water taking one path and well-mixed, 4°C bottom water taking the other.

During the development of a sinking plume, all of the IHC water does not sink to the bottom when it mixes with Lake Michigan waters at the subduction zone. If all of it sank to the bottom, the near-surface temperatures immediately lakeward of the convergence zone would be the same as ambient Lake Michigan water. But temperature profiles immediately lakeward of the convergence zone show the near-surface water to be 1-1.5°C, not near 0°C as is typical for ambient Lake Michigan water when ice is on the lake. Therefore, some portion of IHC water (the portion that does not mix completely) stays near the surface and moves toward the shore due to the action of winds blowing from the southerly quadrants. This was the portion of the IHC effluent that was tracked by the NEPTUNE, and it typically follows a path along the shoreline.

The other path, followed by the portion of IHC water that sinks to the

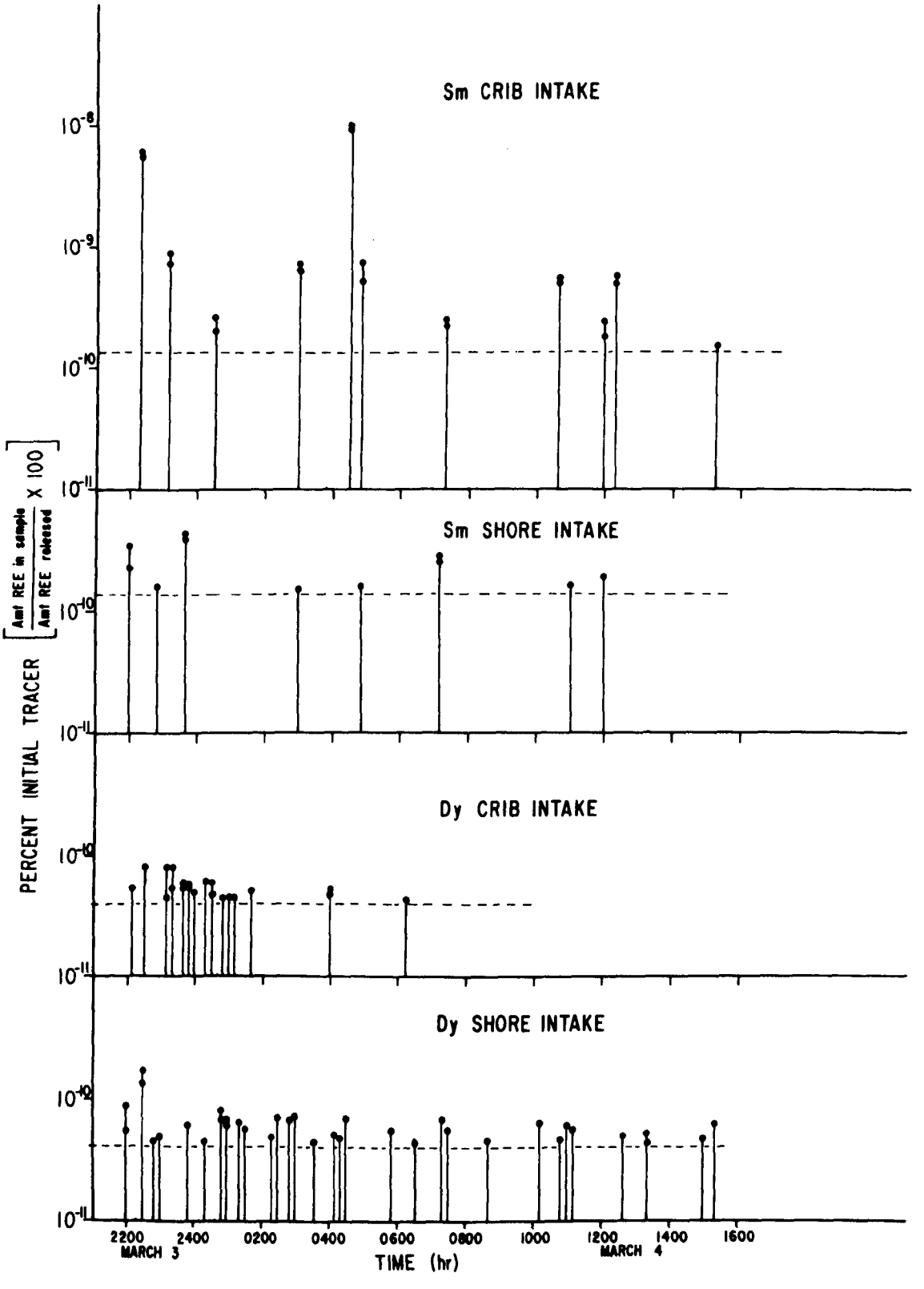

Fig. 8. Relative Amount of Sm or Dy in the Samples Collected at the SWFP as a Function of Time. (Dashed line indicates the minimum detectable amount; two dots at a sample indicate the sample was irradiated twice and shows the value of each irradiation.)

bottom, roughly parallels the 9.1-m (30-ft) depth contour (Fig. 1). This isobath coincides with the lakeward terminus of the IHC entrance channel. Such bottom water would move directly into the lake and then northwestward along the coast, missing the area enclosed behind the Calumet Harbor Breakwater. This sinking-plume portion of the IHC effluent would follow the second path to travel directly to the SWFP. Topographically, the depths of the crib intake and of the IHC entrance channel are about the same, 9.1 m (30 ft). Effluent flowing out of the IHC along the bottom would tend to remain near the 9.1-m (30-ft) contour as it moved northwestward parallel to shore, and this path would carry it directly to the crib intake. Further evidence of the existence of a direct lakeward path is found in the high concentrations of Sm detected in the crib samples and the fact that these samples were drawn at the same time that the NEPTUNE was drawing water samples in Calumet Harbor, water samples that contained Dy (oily waste) but virtually no Sm (canal water).

Figure 8 shows the relative Sm amounts in the raw-water samples collected at the SWFP. Seven of the eleven crib samples contained higher concentrations of Sm than any of the eight shore samples. Two of the crib samples (March 3 at 2220 hr and March 4 at 0430 hr) contained very high concentrations of Sm. In contrast, only fourteen of the crib samples contained Dy and all but two of these were in a three-hour-and-ten-minute span (2210-0120). Thirty-one of the shore samples contained Dy, however, and these were almost equally spaced throughout the entire 17-hour sampling interval at the SWFP.

These results indicate that significantly more Sm (the water tracer) went to the crib intake than to the shore intake. Also, significantly more Dy (the oil tracer) went to the shore intake than to the crib intake. This appears logical because a large portion of the oily waste (Dy) would tend to remain on the surface and thus would be blown toward the shore by the SE winds that occurred during the experiment. Therefore, the oily waste would have had a higher probability of being drawn into the shore intake. The water (Sm tracer), however, would be more likely to sink at the subduction zone and be carried along the 9.1-m (30-ft) isobath with the prevailing northwesterly current mentioned above; further evidence of this partitioning of the oily waste and IHC water is found in the fact that so little Sm (the water tag) was seen in the Calumet Harbor area.

A simple turbulent diffusion model was used to estimate the mixing of the water tag (Sm) during the experiment. The limited Sm data at the shore and crib intakes and the possible partitioning of the tag described above, however, do not permit validation of even the simple model proposed. Nevertheless, a general comparison between model predictions and measured dilution ratios at the crib intake may prove instructive.

Table 3 presents dilution ratios for the Sm samples collected during the experiment at the SWFP's crib and shore intakes. The dilution ratio is defined as the ratio of the initial concentration in the water at the tagging location to the concentration in the collected samples. Initially 13.62 kg of Sm were mixed in the IHC waters over a patch about 24 m x 2 m in area and 1 m in depth, yielding an initial concentration of 0.284 g/L.

The mixing of the tagged portion of the IHC effluent was modeled in two steps. The dilution ratio due to mixing as the patch moved from the tagging location out onto the lake was modeled first. As the patch moved to the

Table 3. Sm Concentrations and Dilution Ratios for Water Samples Collected at the SWFP

Date	Time, CDT	Concentration,[a] µg/L	% Error	Dilution Ratio, $\frac{C_I}{C_F}$[c]
	Crib-Intake Raw-Water Samples			
3 Mar.	2220	61.0[b]	2.0	4.7×10^3
"	2310	7.7[b]	8.3	3.7×10^4
4 Mar.	0030	2.8[b]	23	1.0×10^5
"	0300	6.7[b]	9.6	4.2×10^4
"	0430	102.1[b]	1.4	2.8×10^3
"	0450	6.1[b]	10	4.7×10^4
"	0720	2.4[b]	24	1.2×10^5
"	1040	5.0[b]	13	5.7×10^4
"	1200	2.1[b]	27	1.4×10^5
"	1220	5.1[b]	12	5.6×10^4
"	1520	1.6	34	1.8×10^5
	Shore-Intake Raw-Water Samples			
3 Mar.	2200	2.9[b]	22	9.8×10^4
"	2250	1.6	42	1.8×10^5
"	2340	4.0[b]	18	7.1×10^4
4 Mar.	0300	1.6	46	1.8×10^5
"	0450	1.6	41	1.8×10^5
"	0710	2.7[b]	25	1.1×10^5
"	1100	1.8	34	1.6×10^5
"	1200	1.9	32	1.5×10^5

[a] Minimum detectable amount equals 1.4 µg/L.

[b] Indicates average of two irradiations.

[c] C_I, initial concentration, equals 0.284 g/L. C_F is final concentration.

vicinity of the SWFP's shore and crib intakes, the dilution ratio due to mixing in the lake was combined with the previous ratio and in addition was calculated with a model employing 4/3's-law mixing and another employing linear mixing. The two models produced minimum dilution or center of patch dilution ratios of 6.1×10^3 and 3.0×10^3 respectively in the vicinity of the water intakes. The exact location of the patch relative to the intakes is unknown; however, dilution ratios calculated for the center of the patch are of about the order of magnitude (Table 3) of those determined from sampling at the crib and shore intakes. Details of the models and the coefficients used are discussed in McCown et al. (1978).

SUMMARY

A novel method was developed for simultaneous tagging of both oily wastes and the underlying water, each with a unique tracer, and for determining their individual motions in fresh coastal waters.

The overwater sampling aspect of the experiment required the development of an underway three-dimensional water sampling system for the collection of numerous 15-mL water samples. An NAA technique permitted the analysis of about 17 samples per hour with no pre-irradiation sample preparation other than cleaning of the outside of the Polyvial. The samples were collected and subsequently irradiated in the same Polyvial. Tracer detection limits were 6×10^{-10} for Dy and 2×10^{-8} g for Sm in the 15-mL samples. The method was applied to trace simulated oily waste and the underlying, polluted water from the IHC into Lake Michigan during both the floating- and sinking-plume conditions.

The results of the floating-plume experiment indicated that:

1. When the tagged oil was subjected to severe downward mixing by a passing ore carrier, it did not resurface immediately but remained mixed in the water column,
2. Lake diffusion coefficients calculated from the Dy and Sm data were similar to diffusion coefficients determined by others in the Great Lakes using other tracer techniques, and
3. After the oil was mixed into the water column by the ore carrier, there were no distinguishable differences between the movement and diffusion of the oil/Dy and the movement of the water/Sm.

The results of the sinking-plume experiment indicated that:

1. IHC effluent is definitely transported to the raw-water intakes of Chicago's SWFP under certain lake and meteorological conditions,
2. A partitioning of the oily wastes and underlying water (resulting in separate pathways to the SWFP's raw-water intakes) was made apparent by the employment of the dual tracer system, and
3. Simple model estimates of IHC plume dilution at the SWFP were supported by the experimental measurements.

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TP 8162

ESTIMATION OF FUGITIVE HYDROCARBON EMISSIONS FROM AN OIL REFINERY BY INVERSE MODELING

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ABSTRACT

In order to evaluate an ambient air measurement approach for the definition of fugitive emissions, the hydrocarbon emissions from an oil refinery were estimated through inverse diffusion modeling. The AVQUAL model was utilized to predict the hydrocarbon source terms from the measured ambient hydrocarbon data. The methodology employed in this study is unique in the following ways: (1) several source areas were handled simultaneously; (2) a tracer gas was used to define the meteorology on a real-time basis; and, (3) to minimize experimental errors an over-specified problem was solved to determine the hydrocarbon emissions.

Five different types of hydrocarbon sources for the refinery were assumed. The χ/Q values resulting from these sources were computed for all fifteen downwind receptor locations. Subsequently, a linear programming approach with a non-negative Q constraint was used to solve for the emission rate, Q. Results showed qualitative agreement with direct source testing data.

1. INTRODUCTION

Many industrial, commercial, and domestic activities emit gaseous hydrocarbons and other organic compounds into the atmosphere. These sources include all kinds of fuel burning, solvent usage, and waste disposal operations, as well as the more obvious chemical processing and petroleum refining and marketing. In the South Coast Air Basin (SCAB) of California, petroleum refineries contribute a significant portion of the stationary emissions of hydrocarbons.

Refinery hydrocarbon emissions are primarily of the fugitive type. Leakage from valves, fittings, flanges, pumps, and storage tanks, etc., comprises a major portion of the emission, although some stack emissions result from the operation of process heaters, boilers, incinerators, and flaring. It is impractical, if not impossible, to test all the fugitive emission sources in a refinery to arrive at the emission factors. One approach to inventory the emissions is by means of direct source testing, with emission factors derived by statistically combining the results of selective test sources. Another approach is by ambient testing.

An ambient testing approach requires a workable diffusion model and measurement of ambient hydrocarbon levels in the vicinity of a refinery. When the atmospheric links between emission strength and ambient concentrations are established, the hydrocarbon emission factors can be estimated through inverse diffusion modeling. Theoretically, the ambient testing approach accounts for every emission source if all emissions are well-mixed, and thus provides a more accurate inventory in a rather simple way. This paper describes the methodology used in testing a refinery in the SCAB, for the purpose of evaluating a specific ambient testing approach. Recommendations are also presented for improving the technique.

2. TEST DESCRIPTION

An ideal test site should be relatively compact, and reasonably distant from other hydrocarbon emission sources such as gas and oil wells and other industries so that interference would be insignificant. Also, a heavy traffic flow area is not desirable. Furthermore, the test refinery should be representative of the refining operations in the SCAB.

The most favorable meteorological condition for testing would include a stable atmosphere, and light and uniform wind fields. This kind of condition usually prevails in the early morning hours when drainage flow exists. Conducting testing in the early morning hours has the added advantage of minimum interference from traffic emissions. The test site further requires enough downwind working space for the sampling.

Extensive site survey and literature research of the past meteorological data led to the selection of a refinery located in Paramount, California. Although no direct control over the test conditions (i.e., the meteorology itself) could be exercised, it was possible to select those days for testing which exhibited the most favorable conditions in the refinery area. This was done through the services of a forecaster who could predict when those days would most likely occur.

2.1 Test Equipment and Approach

To establish links between emissions and ambient concentrations, a tracer gas was released at the refinery. By knowing the tracer gas releasing rate and the receptor point concentrations, the source-receptor relationships could be identified through diffusion modeling. Sulfur hexafluoride (SF_6) was used as the tracer. This gas is ideal because it is inert, has very low background level (<6 ppt), and can be detected down to ppt concentration.

Both SF_6 and hydrocarbons were measured simultaneously so that the relationship established for a given period of time by the SF_6 was clearly applicable to the hydrocarbon case for the same time span. Real time monitoring would have been desirable but not practical, owing to the number of receptors required (15 downwind plus 1 to 2 upwind receptors). As an alternative, one-hour integrated samples were taken and immediately analyzed by using a flame ionization detection (FID) hydrocarbon analyzer and an electron capture detection SF_6 analyzer. This was possible by moving the AeroVironment mobile laboratory (Airlab) to the test site during the entire test period. The possibilities of sample alteration were thus minimized. In addition, meteorological parameters needed for diffusion modeling were monitored by using instruments installed on the Airlab. A mechanical weather station, secured on the top of one of the refinery storage tanks, served as a backup.

Some integrated hydrocarbon samples were collected and sent to an analytical laboratory for analysis by a gas chromatography-mass

spectrometry (GC-MS). This way the hydrocarbon concentrations determined by different analytical means, namely FID and GC-MS, could be correlated, and hydrocarbon speciation data obtained.

o Ambient Air Sampling

Two types of ambient air sampling systems were employed: one for FID analyses, and one for the GC-MS analyses. To collect samples for the FID analyses, ambient air was drawn from 6 feet above ground through a Tygon tubing and a prefilter by a Spectrex Model AS-100 gas sampling pump, and delivered into a 5-liter Tedlar bag. The delivery rate was pre-set so that at the end of the one-hour sampling period the Tedlar bag was inflated enough to provide the analysis needs.

For the GC-MS analyses two samplers were used. One of the samplers was the same Tedlar bag sampler described above. The other was an adsorption sampler, in which an adsorption tube filled with activated charcoal was used to concentrate the ambient hydrocarbons. Ambient air was drawn from a sample point 6 feet above ground through a Tygon tubing, the adsorption tube, a rotameter, and a filter by a Nepture Dyna-Pump Model 2. After sampling, the exposed adsorption tube was immediately capped and later sent away to the laboratory as a set with the Tedlar bag sampled during the same time span. These bag samples were analyzed for low molecular weight hydrocarbons (C_6 and below). Adsorption tube samples were analyzed for higher hydrocarbons (C_6 and above). The overlapping of C_4 - C_6 from both samplers served as a cross-check.

o SF_6 Releasing and Analyzing

The SF_6 tracer gas was passed through a copper coil, a liquid trap, a mass flowmeter, and a rotameter, and released into the atmosphere 28 feet above ground. Release flowrates were measured by use of a Hastings Linear Mass Flowmeter and continuously recorded by a strip chart recorder.

Analysis of SF_6 content in the ambient air sample was accomplished by use of an AID Model 511 Portable Gas Chromatograph with electron capture detector. This instrument, produced by Analytical Instrument Development Inc., Avondale, Pennsylvania, was capable of detecting down to the part per trillion (ppt) level. Instrument calibration was performed before and after the instrument was used on each test day.

o Hydrocarbon Analyzing

A Beckman Model 6800 Air Quality Chromatograph was employed at the study site to analyze the ambient air sample hydrocarbon contents. This instrument operates on the FID principle, and was capable of measuring total hydrocarbon (THC), methane (CH_4), and carbon monoxide (CO). The measurements of CO content gave information pertaining to traffic interference.

Instrument calibration was carried out before and after the instrument use for each test day. The hydrocarbon calibration gas was methane. Thus, THC data reported were expressed as CH_4.

FID instrument response is related not only to the concentration of the hydrocarbon being measured, but also to the "effective carbon number" of the hydrocarbon compound. The effective hydrocarbon number varies depending on the number of carbon atoms in the molecule and on the type of compound. Thus, an FID instrument calibrated with CH_4 generally would would respond to the heavier hydrocarbons, and the data collected could be misleading. In order to compensate for the deficiency, some ambient air samples from the vicinity of the refinery were collected for the GC-MS analyses. These samples were taken side-by-side with those analyzed by the on-site FID instrument. This approach constituted the "field calibration" for the FID results. The GC-MS analytical results also could reveal hydrocarbon speciation information.

o Meteorological

An anemometer installed 8 meters above in the Airlab was used for measuring wind speed and wind direction. Sigma meters were used to measure σ_w and σ_θ, which represent the r.m.s. values of vertical wind speed and horizontal wind direction variation, respectively. The outputs were monitored by using strip chart recorders. As a back-up, a mechanical weather station was secured on the top of one of the storage tanks for continuous monitoring of wind speed, wind direction and temperature.

2.2 Test Procedure

A typical test began shortly after midnight when drainage flow prevailed, and the atmospheric conditions were stable. Tracer gas was released at a predetermined location within the refinery, 28 feet above ground at a rate of approximately 2.5 lb/hr. Enough time was allowed (at least one hour) so that the ambient SF_6 concentrations would reach steady state before the sampling was initiated.

The wind direction chart was first examined (wind data were recorded continuously throughout the testing), and the prevailing wind direction determined. Helium-filled balloons were released to make certain that the wind direction was also uniform at higher elevations. Strategic receptor locations for both upwind and downwind sides were then assigned and field technicians deployed.

Samplers were turned on at approximately the same time ($\pm$5 minutes) by field technicians, and hourly samples were collected thereafter until sunrise. Ambient samples were collected up to 10 a.m. Hourly integrated samples collected were brought to the Airlab immediately for analysis for $THC/CH_4/CO$. The remainder of the samples were then analyzed for SF_6 concentrations in a nearby upwind motel room; this was a precaution taken to prevent the SF_6 analyzer from being contaminated. Samples for GC-MS analysis were taken at three locations.

Testing was carried out on four days in November - December 1976. Figure 1 illustrates the typical receptor locations, the SF_6 release point, and the Airlab location for a given test day. The sampler locations for GC-MS analysis are also indicated.

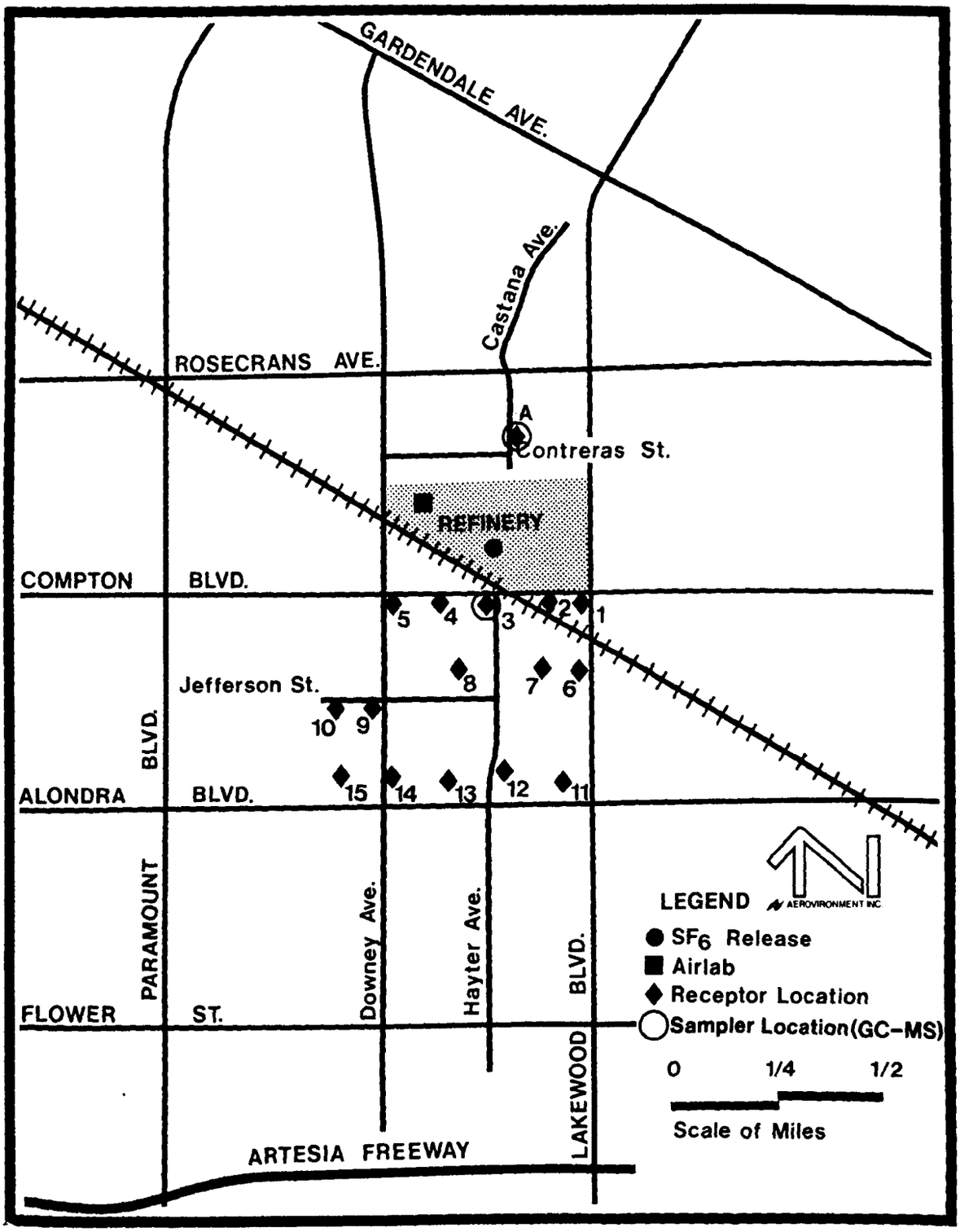

FIGURE 1. Schematic representation of test site for 30 November 1976.

3. DIFFUSION MODEL

The diffusion model used for this study was the AVQUAL model. AVQUAL is a microscale diffusion model developed from Taylor's turbulent diffusion theory (Taylor 1921), which explicitly incorporates ground roughness and heat flux. It is simple and accurate. The presence of inversions is accounted for. Furthermore, this model has the capability of simulating unsteady effects, and wind shifts can readily be incorporated. A detailed description of this model has been documented by Lissaman (1973).

In the AVQUAL model, the ratio of ground level ambient concentration of a given receptor point to the source strength is a function of meteorological conditions. Parameters involved are wind speed, wind direction, and vertical and horizontal dispersion speeds etc. This relationship is expressed as follows:

$$\frac{\chi}{Q} = \frac{\exp\left[-\frac{1}{2}\left(\frac{u}{a} - \frac{x}{aT}\right)^2\right]\exp\left[-\frac{y^2 + z'^2}{2a^2T^2}\right]}{\pi\sqrt{2\pi}\, a_v r^2} +$$

$$\frac{xu}{2\pi a_v a r^3}\exp\left[-\frac{1}{2}\left(\frac{u}{a}\right)^2\left(\frac{y^2 + z'^2}{r^2}\right)\right]\left[1 + \text{erf}^*\frac{1}{\sqrt{2}}\left[\frac{xu}{ar} - \frac{r}{aT}\right]\right]$$

where χ = ambient receptor point concentration at time T
Q = source strength
u = mean wind speed for time span T
x = receptor downwind distance from the source
y = receptor crosswind distance from the source
z = source height
r^2 = $x^2 + y^2 + z'^2$
z' = z/λ
λ = ratio of vertical dispersion speed to horizontal dispersion speed; i.e., a_v/a

As is evident, x and y are dependent on the mean wind direction for time span T.

For a given test run, χ, Q, u, x, y and z are all measurable parameters. T is equivalent to the time span over which the integrated samples are collected. Furthermore, λ and a_v are calculable by equating a_v to σ_w and λ to a_v/a, where $a = u \cdot \sigma_\theta$. In the model validation, Q, u, x, y, z, λ, and a_v are fed into AVQUAL to calculate χ. The theoretical χ values thus calculated are compared with the measured χ values. Significant discrepancies observed between them would indicate that the measured meteorological data are insufficient to validate the model. In this case, it is necessary to carry out a model calibration.

4. TEST RESULTS AND MODEL VALIDATION/CALIBRATION RESULTS

4.1 Test Results

Testing was conducted on 23, 24 and 30 November, and 1 December 1976. Except for the 24 November testing, the meteorological conditions during the test hours were favorable. The atmosphere was calm with a stability class of E or F (Pasquill, 1962). Wind was generally directly from the north with a wind speed between 3-7 mph, a typical drainage flow condition in that area. Occasionally, though, the wind would shift greatly and the wind shift persisted for prolonged periods of time. These were the runs which were difficult as far as model validation was concerned.

During the 24 November testing the atmospheric conditions were too calm. There was virtually no wind most of the time. The refinery plume at that time would disperse more or less evenly toward all directions. Consequently, all of the receptors including the "upwind" one recorded about the same concentrations; THC was around 10 ppm, CH_4 around 5 ppm, and SF_6 around 20 ppt

Table 1 shows the results of successful test hours. A receptor location designated with "A" indicates an upwind site, while that designated with 1 through 15 was downwind site. There were two upwind sites for Run 15 as shown in Table 1.

It is obvious that the contribution from the refinery plume to a specific receptor point would be the difference in concentration between that receptor point and the upwind point. This is true for all species measured, namely SF_6, THC, CH_4, and CO, provided that the incoming wind is uniform and that no interference is present. These net results will be used for the diffusion model validation/calibration, and for the hydrocarbon emission predictions.

Some of the samplers collected air samples for both FID and GC-MS analyses as mentioned previously. A relationship was established for the non-methane hydrocarbons from the Douglas refinery air samples:

$$(NMHC)_{GC\text{-}MS} = 1.36\ (NMHC)_{FID}$$

The correlation factor of 1.36 was reasonable considering that the FID instrument was calibrated by using CH_4 standards. This correlation factor was eventually applied to the final emission prediction data.

4.2 Model Validation/Calibration Results

In the model validation of AVQUAL by use of SF_6, Q, u, x, y, z, χ, and a_y were fed into AVQUAL to calculate χ as mentioned in Section 3. The theoretical χ values thus calculated were compared with the measured χ values. Significant discrepancies were observed between them, indicating the measured meteorological data were insufficient to validate the model.

TABLE 1. Summary of test results.

Run No: 1
Date: 11/23/76
Time: 0330-0430
Wind Speed: 1.6 m/sec
Wind Direction: 340 deg
σ_w: 0.07 m/sec
σ_θ: 7.2 deg
Temp: 11°C
Stability Class: E
Q_{SF_6}: 288,960 µg/sec

Receptor	SF_6 (ppt)	THC (ppm)	CH_4 (ppm)	CO (ppm)
A	3.0	5.4	2.5	3.8
1	6.4	6.6	2.5	4.0
2	3.0	6.7	2.5	3.8
3	1525.0	7.3	2.5	3.7
4	116.0	7.9	2.5	3.7
5	9.6	6.0	2.5	4.0
6	-	6.0	2.5	3.5
7	6.0	5.6	2.5	3.7
8	-	6.3	2.5	3.6
9	150.0	5.8	2.5	3.8
10	420.0	5.4	2.5	3.9
11	3.0	6.9	2.5	4.0
12	3.0	6.0	2.5	3.8
13	-	7.1	2.5	3.9
14	13.0	6.1	2.5	3.8
15	21.0	7.6	2.5	3.8

TABLE 1. (Continued)

Run No: 9
Date 11/30/76
Time: 0330-0430
Wind Speed: 2.5 m/sec
Wind Direction: 335 deg
σ_w: 0.02 m/sec
σ_θ: 4 deg
Temp: 9°C
Stability Class: F
Q_{SF_6}: 299,604 µg/sec

Receptor	SF_6 (ppt)	THC (ppm)	CH_4 (ppm)	CO (ppm)
A	19.5	5.1	2.6	2.5
1	26.0	6.0	2.6	2.8
2	22.5	6.1	2.5	2.2
3	4300.0	9.4	2.6	2.3
4	13.5	9.2	3.2	2.5
5	16.5	5.5	3.0	3.3
6	-	6.0	2.6	2.3
7	20.5	5.5	2.5	2.1
8	460.0	7.0	3.0	2.8
9	-	5.2	3.0	3.1
10	17.5	5.9	3.1	5.2
11	37.0	5.4	2.8	3.2
12	740.0	6.0	2.6	2.5
13	1010.0	6.5	2.8	2.5
14	16.5	5.3	2.9	2.9
15	17.5	6.0	3.0	2.6

TABLE 1. (Continued)

Run No: 10
Date: 11/30/76
Time: 0430-0530
Wind Speed: 2.0 m/sec
Wind Direction: 330 deg
σ_w: 0.01 m/sec
σ_θ : 5 deg
Temp: 8°C
Stability Class: F
Q_{SF_6} : 299,604 µg/sec

Receptor	SF_6 (ppt)	THC (ppm)	CH_4 (ppm)	CO (ppm)
A	13.5	4.7	2.7	2.3
1	16.0	5.3	2.5	2.2
2	17.5	5.7	2.4	1.5
3	4800.0	10.6	2.4	1.5
4	13.5	5.5	3.1	2.0
5	13.0	4.6	2.7	2.9
6	13.0	6.0	2.7	1.8
7	24.0	5.3	2.4	1.6
8	-	-	-	-
9	13.0	4.6	2.8	2.7
10	11.8	5.0	2.8	2.1
11	130.0	5.7	2.6	2.8
12	500.0	6.3	2.7	2.3
13	22.5	5.1	2.7	2.1
14	11.5	4.6	2.8	2.7
15	13.5	4.9	2.9	2.2

TABLE 1. (Continued)

Run No: 11
Date: 11/30/76
Time: 0530-0630
Wind Speed: 2.2 m/sec
Wind Direction: 340 deg
σ_w: 0.01 m/sec
σ_θ: 4 deg
Temp: 8°C
Stability Class: F
Q_{SF_6}: 299,604 µg/sec

Receptor	SF_6 (ppt)	THC (ppm)	CH_4 (ppm)	CO (ppm)
A	5.6	4.7	2.6	4.1
1	9.8	6.4	2.4	5.0
2	5.6	5.5	2.3	1.9
3	4700.0	7.6	2.4	2.4
4	10.5	10.0	3.1	2.7
5	6.3	4.8	2.6	6.6
6	7.7	5.6	2.7	4.5
7	7.0	4.9	2.5	2.3
8	455.0	7.9	2.4	1.1
9	6.3	4.7	2.6	4.8
10	9.0	4.5	2.6	2.0
11	62.0	5.3	2.6	2.7
12	1200.0	6.1	2.4	1.8
13	425.0	6.1	2.6	2.5
14	-	-	-	-
15	9.4	4.6	2.6	3.1

TABLE 1. (Continued)

Run: 15
Date: 12/1/76
Time: 0800-0900
Wind Speed: 1.6 m/sec
Wind Direction: 330 deg
σ_w: 0.09 m/sec
σ_θ : 7 deg
Temp: 13°C
Stabillity Class: F
Q_{SF_6}: 286,220 µg/sec

Receptor	SF_6 (ppt)	THC (ppm)	CH_4 (ppm)	CO (ppm)
A1	76.0	5.2	2.2	5.0
A2	82.0	6.0	2.3	5.5
1	-	7.3	2.3	4.1
2	200.0	7.8	2.4	5.2
3	1100.0	6.7	2.4	7.4
4	34.5	5.4	2.2	4.9
5	96.0	6.4	2.3	5.5
6	150.0	6.5	2.2	4.7
7	1400.0	7.2	2.3	5.3
8	71.0	6.0	2.4	6.0
9	450.0	6.4	2.3	6.1
10	480.0	6.7	2.3	4.4
11	-	OS	2.3	5.0
12	-	6.9	2.3	5.3
13	480.0	6.0	2.3	5.1
14	500.0	5.2	2.3	4.1
15	498.0	6.8	2.3	5.9

This was most likely caused by the nonrepresentativeness of the Airlab meteorological measurements. Specifically, the meteorological conditions at the SF_6 release location were different from those observed at the Airlab. This is understandable because the refinery facilities, such as storage tanks, cooling towers, and fractionation towers etc., could cause slight wind direction changes and more horizontal mixing. Perhaps more important is the fact that the refinery operation's heat output would create significantly more vertical mixing than what was observed at the Airlab. As a result, development of diffusion factors empirically by model calibration was necessary.

In the model calibration, mean wind direction, χ, and a_v were determined by trial and error to minimize the difference between predicted and measured values of SF_6. The other parameters such as Q, u, and z were influenced by the refinery operations and thus the measured data were used. Table 2 shows the comparison of the measured and calibrated meteorological data for the test runs where satisfactory agreement between predicted and measured SF_6 data could be obtained. Figure 2 illustrates the typical predicted SF_6 isopleths and SF_6 concentrations at each receptor point for each calibrated run. Also given is the measured SF_6 concentration at each receptor point for comparison purposes.

The calibrated mean wind direction was consistently greater than the measured value by 15 to 25 degrees, as shown in Table 2. The calibrated values of both a_v and χ were approximately two orders of magnitude larger than the measured values. Since χ is the ratio of a_v to a, it follows that only a_v, the vertical dispersion speed, was significantly impacted by the refinery operations; whereas a, the horizontal dispersion speed, was in the same order of magnitude as was observed in the Airlab. Thus, it is possible that had the meteorological instruments been installed near the SF_6 release point within the refinery complex the AVQUAL model would have validated satisfactorily using the measured meteorological data.

The model calibration factors listed in Table 2 were used directly for the hydrocarbon emission predictions in the following section.

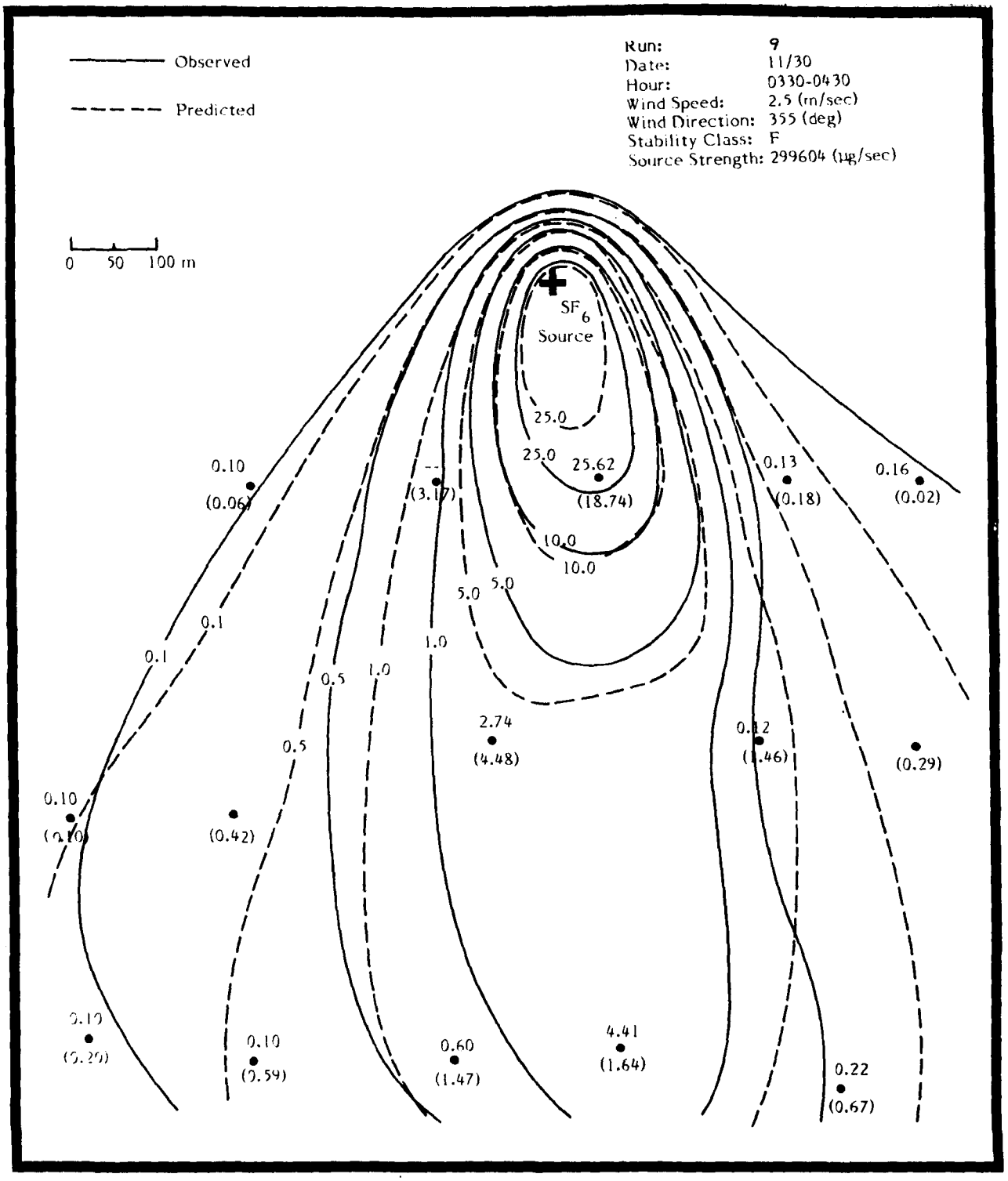

FIGURE 2. Comparison of observed (solid line) and predicted (broken line) SF_6 isopleths; at each receptor point observed and predicted (parenthses) SF_6 concentrations are also given in $\mu g/m^3$.

TABLE 2. Comparison of measured and calibrated meteorological data.

Run No.	Mean Wind Speed (m/sec)	Mean Wind Direction (deg)		a_v(m/sec)		λ	
		Measured	Calibrated	Measured*	Calibrated	Measured**	Calibrated
1	1.6	340	360	0.07	4.4	0.35	10.0
9	2.5	335	355	0.02	2.5	0.11	3.5
10	2.0	330	350	0.01	1.8	0.06	3.0
11	2.2	340	355	0.01	2.5	0.07	5.5
15	1.6	330	355	0.09	2.5	0.46	5.0

* assuming $a_v = \sigma_w$

** calculated as follows:

$$\lambda = \frac{a_v}{a} = \frac{\sigma_w}{(u)(\sigma_\theta)}$$

where σ_θ in radian

5. HYDROCARBON EMISSION ESTIMATION AND DISCUSIONS

One of the initial tasks in determining the method for predicting hydrocarbon emission rates was to define the sources. The oil refinery chosen for this study has many fugitive hydrocarbon emission sources. Each source is characterized by the particular process associated with it.

After the emission source is defined, the next step was to determine the most practical and accurate way of modeling them. The AVQUAL model, discussed in Section 3, was then utilized to calculate the relative impacts of the different source types. Finally, a numerical technique was devised to give a best fit emission rate in accordance with the data. The method is as follows:

1. For the emission source definition, it was decided to classify the numerous sources into areas of similar source types. Five source types were chosen for their distinct emission charateristics, namely, process areas and emulsion plant (Q_1), asphalt plant (Q_2), loading-unloading facilities (Q_3), fixed roof storage tanks (Q_4), and floating roof storage tanks (Q_5). Because not all of the sources of any of the five types occurred together in a centralized area, smaller areas were defined which contained only a particular category. The final scheme consisted of 31 defined areas: five areas of Q_1 type, two areas of Q_2 type, two areas of Q_3 type, fourteen areas of Q_4 type, and eight areas of Q_5 type. A diagram of the defined areas and their source category is depicted in Figure 3.

2. The defined area is further subdivided into a number of point sources whose optimum spacing was determined. It was found that by reducing the spacing of point sources the error of estimating downwind concentrations from an area source was reduced. However, it was also found that further reduction from a spacing of 25 feet would not result in a significant improvement in the model output. For example, reducing the spacing from 25 feet to 12.5 feet would increase the accuracy by less than 2%. Thus, a spacing of 25 feet was used in the emission prediction. Within the unit area of 25 feet square, it was assumed that the source points were distributed evenly. This approach allowed for equal emphasis of the different source categories on an area basis.

3. The AVQUAL model can be expressed in a simplified form as follows:

$$\chi/Q = f(\text{meteorology, coordinates})$$

Thus, χ/Q can be calculated solely from the meteorological data and the coordinates. For a unit area k of emission type j, its contribution to the ambient concentration at receptor point i (χ_{ijk}) is identical to $Q_j f_{ijk}$. Thus, the total contribution of refinery emissions to receptor i can be expressed as

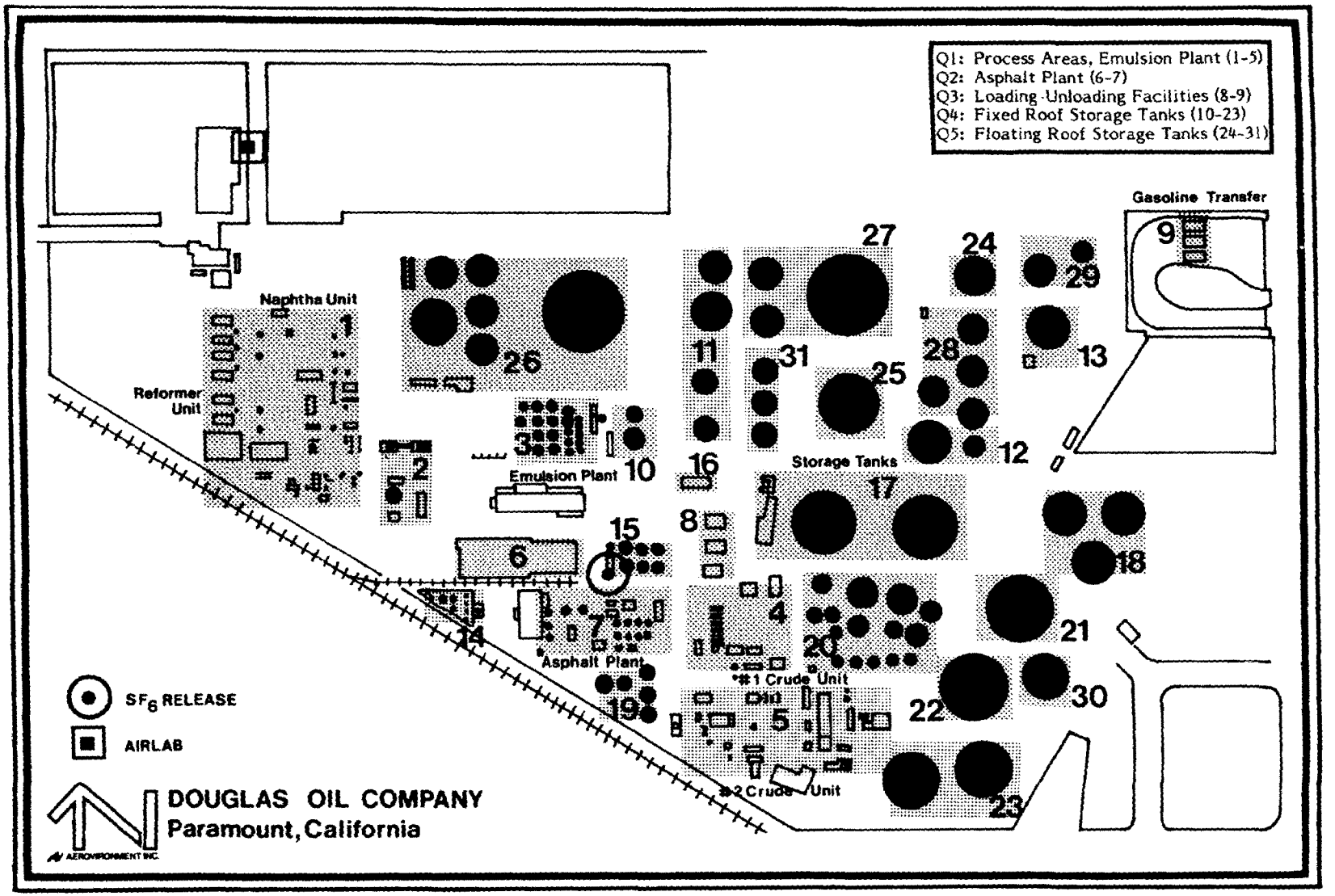

FIGURE 3. Source definitions used for the refinery emission prediction.

$$\chi_i = \sum_{j=1}^{5} \sum_{k=1}^{k} Q_j f_{ijk}$$

In Equation (2), χ_i was measured and f_{ijk} was calculated from the calibrated meteorological data; with Q_j's the only unknowns to be solved. Therefore, theoretically, only five downwind receptor points were needed to solve the equation since only five source types were assumed.

4. Realistically, however, more receptor points were needed for the accurate prediction. In this study as many as 15 downwind receptors were used. A numerical analysis technique was therefore necessary to solve the over-determined set of equations.

5. A linear programming approach with a non-negative Q constraint was used for solving the over-determined case. This technique, however, was very sensitive to the variation of receptor concentrations; a noise of ± 5% in receptor concentrations was about its tolerable limit. Eventually it was necessary to smooth out the receptor concentration data by fitting the receptor points with the same downwind distance to a 2nd-order polynomial equation. The hydrocarbon emission prediction thus obtained gave the most consistent results.

6. The hydrocarbon concentration correlation factor obtained in Section 4.1 was applied to the predicted emission rates.

Table 3 presents the results of the emission rate predictions. Emission rates from five different source categories as well as the total rate are tabulated for each run. Run 1 indicates the Q_5 source type was the predominant source, while the rest of the emissions came from the Q_1 category; Q_2, Q_3 and Q_4 were negligible sources. Run 9, however, shows that Q_1 and Q_5 types were approximately equally weighted as far as contribution to the total emission rate, whereas the contribution from source types Q_2, Q_3 and Q_4 were still negligible. Runs 10, 11, and 15 predicted that the emissions were predominantly from the Q_1 type.

One would not expect to observe a change in the predominant source type from hour to hour, such as the case observed for Runs 9, 10 and 11 which were conducted on November 30. Also, one would not expect the change from day to day because of the fairly steady refinery operations. Therefore, the average of the five runs should be more significant as far as interpretation. As shown in Table 3, the arithmetic average of the source type emissions indicates that Q_1 contributed about 60% of total emissions, while Q_5 contributed about 40%; Q_2, Q_3 and Q_4 were essentially negligible. The average total emission rate from the Douglas refinery was predicted to be 26 lb/hr.

TABLE 3. Hydrocarbon emission predictions for various source categories of the refinery in lb/hr.

Run No.	Q_1	Q_2	Q_3	Q_4	Q_5	Total
1	8	0	0	0	39	47
9	14	0	0	0	10	24
10	12	0	0	3	0	15
11	18	0	0	1	0	19
15	23	0	0	0	0	23
Avg.	15	0	0	1	10	26

Q_1 = process areas and emulsion plant

Q_2 = asphalt plant

Q_3 = loading-unloading facilities

Q_4 = fixed roof storage tanks

Q_5 = floating roof storage tanks

This predicted value is about one order of magnitude lower than that in the KVB source testing results, presented in Table 4 (KVB, 1977). Also presented is the KVB estimation of the Douglas refinery hydrocarbon emissions by using the Southern California APCD and AP-42 emission data. The KVB source testing was conducted at the same time periods as the ambient testing was performed to formulate a coherent testing program.

Compared to these values, AV's prediction appears to be too low quantitatively. Qualitatively, however, it is sensible. A major part of the emissions came from the process areas and the floating roof storage tanks, The asphalt plant contributed little, if any, to the total emissions. Fixed roof storage tanks normally should contribute to the source term. Nevertheless, the ambient testing was conducted during the early morning hours when the fixed roof storage tanks were "breathing in." Thus, the prediction of negligible emissions from this particular source type is reasonable.

Possible causes of the discrepancy between the AV prediction and the KVB source testing results include hydrocarbon layering under the extremely stable atmospheric conditions, and heavier hydrocarbon retainment within the refinery because of the boundary wall. Source testing data clearly indicated CH_4 emissions from the refinery (KVB, 1977). However, AV's ambient testing data showed little or no difference for the upwind and downwind CH_4 data. It is possible that under the very stable conditions most of the CH_4 emitted would diffuse upward, and thus was not detectable in the ground level. Heavier hydrocarbons (in reference to air) under the same stable atmospheric conditions would tend to diffuse downward. They were, however, more or less contained in the refinery by the existence of the wall in the downwind side of the refinery. The results of the above possible causes would be the reduction in the ambient air hydrocarbon concentrations; and thus, the underestimation of emissions.

TABLE 4. Comparison of KVB's preliminary hydrocarbon emissions source testing results with the results calculated by using the SCAPCD and AP-42 data for the refinery; in lb/hr.

			KVB/ARB	
	SC APCD	AP-42	24-Hr. Av.	Night
Stack Emissions	11	26	13	13
Fugitive Emissions	84	123	54	54
Tank Storage and Transfer	90	95	95	67
TOTAL	185	244	162	134

6. CONCLUSIONS AND RECOMMENDATIONS

The following conclusions may be drawn as a result of the ambient testing of the refinery:

1. The AVQUAL model can be adequately applied to the Douglas refinery environs.

2. Emission prediction results for various source types are qualitatively sound. The predominant sources during the test hours were the process areas and the emulsion plant (source type Q_1) and the floating roof storage tanks (source types Q_5).

3. Quantitatively, the prediction is one order of magnitude lower than predictions by accepted emissions factors or from source testing. This is possibly caused by the hydrocarbon layering in the immediate atmosphere, and also by the existence of a wall at the refinery boundary in the downwind side.

For future hydrocarbon source emission predictions using the ambient testing approach, the following are recommended:

1. Meteorological parameters should be measured at a site which is representative of the source area. The possible heat output from the area should be taken into consideration in the site selection.

2. Receptor locations should be set up close to the source area in order to observe maximum impact.

3. Instead of extremely stable atmospheric conditions (Class F), the testing can be done under less severe conditions, such as Class D, to provide better air mixing. Alternatively, if the study has to be performed under Class F conditions, the collection of air samples at three different heights would be desirable to detect any layering problem.

4. More receptors should be arrayed crosswind for the same downwind distance to facilitate the data smoothing technique.

5. Avoid selection of source area with wall perimeters so that no boundary type problem will exist. In so doing, the source-receptor relationship would be solely governed by atmospheric diffusion.

In summary, hydrocarbon emission prediction through diffusion modeling represents a unique approach to the inventory problem. No statistical analysis is needed, as is with the case of direct source testing. Also, theoretically, every hydrocarbon source is accounted for since the mixed refinery plume is collected at the downwind locations.

The apparent weakness of this approach lies on the fact that the measurement system might provide too much of an error for the model to handle. A variation of $\pm$ 5% for the downwind concentrations proved to be intolerable. Eventually, it was necessary to smooth out the data to cut down the noise.

One way to improve the sampling accuracy would be to arrange the receptor locations closer to the source so that maximum impact can be observed, and thus the percent error of the receptor point concentrations can be minimized. Another way would be to utilize real time, continuous hydrocarbon monitors for all receptor points. This way the handling of air samples is cut down to minimum, and thus the most accurate results are achieved. However, this approach is expensive. The methods employed in this study, i.e., on-site analyses of integrated samples, should be the most economical and acceptable procedures.

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MEASUREMENT OF FUGITIVE EMISSIONS FROM PETROCHEMICAL PLANTS

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ABSTRACT

Fugitive hydrocarbon emissions from petrochemical plants were measured as part of an emission assessment program being conducted by Monsanto Research Corporation under contract with the U.S. Environmental Protection Agency (EPA). The objective of this study was to provide accurate data on fugitive emissions which would reflect current technology and operating practices in the petrochemical industry. Field sampling and analysis was conducted at four plants producing monochlorobenzene, butadiene, ethylene oxide/glycol, or dimethyl terephthalate.

Fugitive sources at petrochemical plants include pumps, valves, flanges, process drains, compressors, agitators, sample valves and relief devices. A random sample of potential sources at each plant was selected and screened with an organic vapor analyzer to determine sources having hydrocarbon emission rates in excess of 0.5 g/hr. These sources were enclosed in a flexible plastic tent and sampled using a portable sampling train. On-site hydrocarbon sample analysis was performed using a gas chromatograph with flame ionization detection to identify and quantify each organic component. Sampling data showed the average hydrocarbon emission rates from pumps, valves, flanges, and compressors to be 22 g/hr, 4.5 g/hr, 1.4 g/hr and 30 g/hr, respectively. Of the total number screened, the estimated percentages of pumps, valves, and flanges having significant (≥0.5 g/hr) emission rates were 23%, 7%, and 2%, respectively. The number of such sources at petrochemical plants having significant emissions is ∿20% lower than the number of such sources at petroleum refineries.

INTRODUCTION

BACKGROUND

Hydrocarbon emissions from petrochemical plants can be divided into two broad categories: stack and fugitive emissions. Stacks and/or vents identified as principal hydrocarbon emission points are considered to be controlled sources. Hydrocarbon emission points other than stacks and/or vents are considered to be fugitive sources. Fugitive emissions may occur due to accidents, inadequate maintenance, or poor planning, although many fugitive emissions occur even in the absence of such conditions and are unavoidable characteristics of some process operations.

The emission factors generally employed for fugitive hydrocarbon emissions are based on a study conducted to determine miscellaneous hydrocarbon emissions from petroleum refineries in the late 1950's by Los Angeles County (Ref. 1). These emission factors are being used by both industry and air pollution agencies to estimate emissions from fugitive sources, with little or no modification to reflect current equipment, technology, and operating practices. Current data on the amount and composition of fugitive hydrocarbon emissions from petrochemical plants also are not available; they are at best estimated. Therefore, sampling efforts were necessary to obtain data of the required quality and accuracy and to identify and quantify emissions of potentially toxic chemical substances.

The EPA is currently conducting a study on fugitive hydrocarbon emissions from petroleum refineries to determine the adequacy of established emission factors in light of current refinery technology and equipment. Information on fugitive emissions from petrochemical plants generated in this study will be used by the EPA to compare fugitive emissions from such plants with those from petroleum refineries.

SCOPE OF STUDY

Fugitive hydrocarbon emissions were measured at plants manufacturing monochlorobenzene, butadiene, ethylene oxide/glycol, and dimethyl terephthalate. Hydrocarbon emission rates were determined for pumps, valves, flanges, process drains, compressors, agitators, sample valves, and relief devices. On-site analysis was performed to identify individual organic components. Sampling results were extrapolated to evaluate fugitive emissions from all potential point sources within each process.

Data on fugitive emissions from petrochemical plants and petroleum refineries were compared. Petroleum refinery data were acquired from the study conducted by Los Angeles County (Ref. 1).

SAMPLING AND ANALYSIS

SOURCE SELECTION

Potential fugitive sources at a petrochemical plant are numerous and varied. Due to budget and time constraints it was not feasible to determine whether each source type was emitting fugitive hydrocarbons. In order to define the number of sources required to achieve statistically relevant data three factors were required. Two of the factors are the desired confidence level and the corresponding error. On this program , these factors were established as the 95% confidence level and a corresponding error of ± 70%. A third factor needed to define the number of samples required is the standard deviation of the measurements. Assuming that sampling data follow a normal distribution with no bias, the following relationship exists:

$$S_x = \frac{\Sigma\sqrt{n}}{t}$$

where: S_x = estimated relative standard deviation

Σ = error (difference between true emissions and mean of measurements, ±70% for this program)

n = number of samples

t = value from statistical tables for "t" distribution

A plot of S_x versus n is given in Figure 1 which was used to predict the minimum number of sources required from estimated relative standard deviations to obtain ±70% uncertainty.

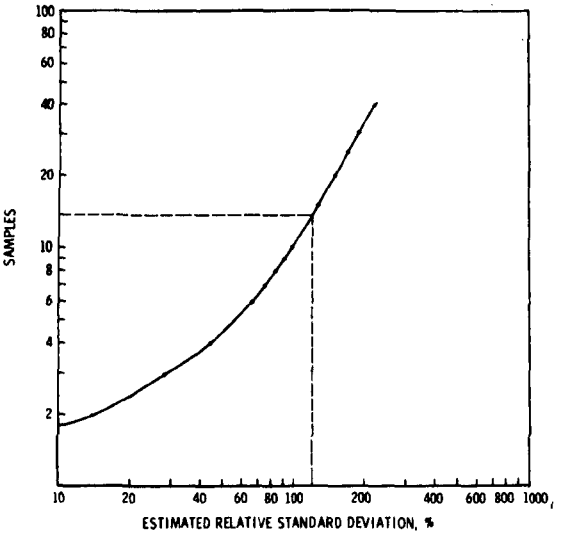

Figure 1. Number of samples required for fugitive sampling.

Fugitive source types which were found at the various petrochemical plants included pumps, valves, flanges, compressors, relief valves, sample valves, process drains, and agitators. Each plant site was inspected prior to sampling to determine the existence of any unusual process configurations. Unusual configurations, such as open vessels containing hydrocarbons, were not found at the plants sampled.

To guarantee that a sufficient number of samples was obtained, approximately 40 samples of each source type were tested whenever possible. Sources were selected randomly throughout each plant to insure that fugitive emissions sampled were representative of and resulted from the process being studied.

Source selection was randomized by one of two methods. The preferred method was to utilize process instrumentation diagrams which depict all pumps, valves, compressors, etc. The depicted sources were sequentially numbered on the diagram, and a random number table was used to determine which numbered sources were to be tested.

Many process instrumentation diagrams were, however, either not sufficiently detailed or were outdated, rendering the above method impractical. In this instance potential fugitive sources were selected by placing a plastic grid over a diagram of each unit operation or process. Based on the size and shape of the available diagram, each square millimeter covering the area was consecutively numbered. A random number table was then used to select corresponding squares on the grid. These points were then transferred to the diagram. The diagram, marked with the number of screening sites selected, was taken into the field for source selection. Fugitive sources located nearest to the area marked on the diagram were then tagged for source screening. Figure 2 shows the selection of sampling sites in a storage tank area.

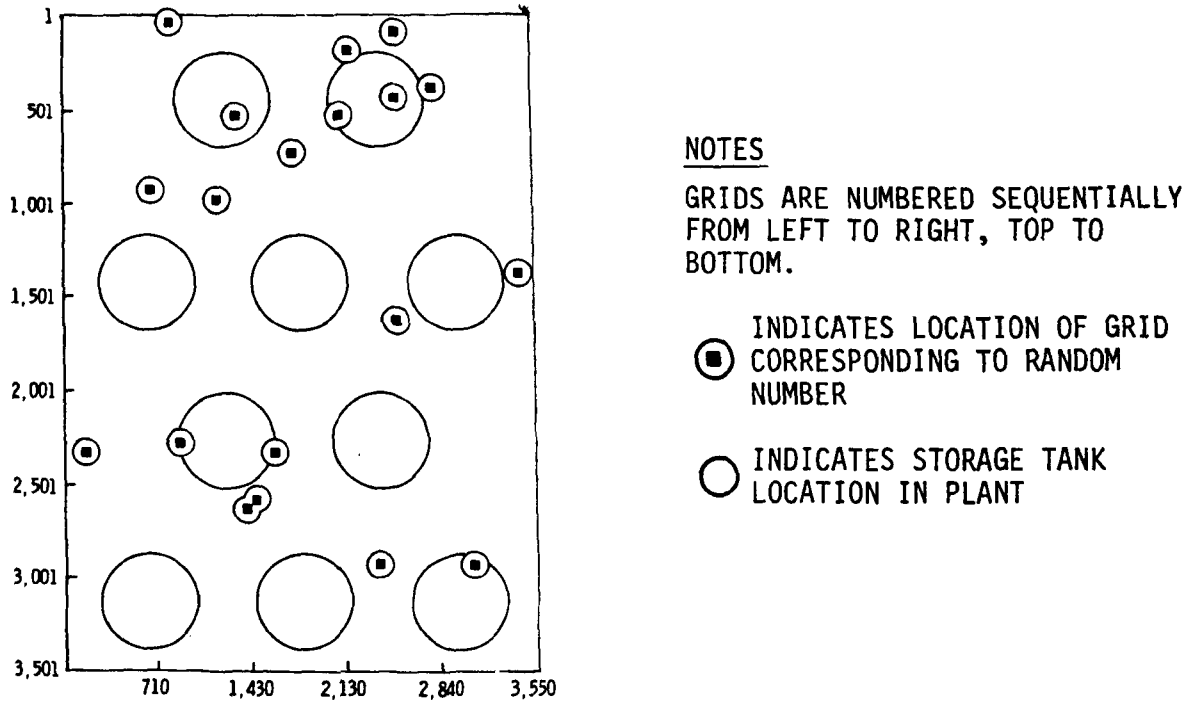

Figure 2. Selection of screening sites in plant storage tank area using random numbers.

The selection of sources for screening was supervised by the plant's engineer, who was responsible for ensuring that all selected sources were a part of the manufacturing operation being tested.

FUGITIVE SOURCE SCREENING

Fugitive emissions were detected using an Organic Vapor Analyzer (OVA), Model 128, obtained from Century Systems Corporation. The OVA-128 instrument utilizes a hydrogen flame ionization detector (FID) sensitive to 1 ppm of organic vapor and provides a direct meter readout with a 2-second response time. The instrument is available with a linear readout covering three ranges: 0 to 10 ppm, 0 to 100 ppm, and 0 to 1,000 ppm. Using a calibrated dilution probe, the instrument's readout capabilities were increased to include ranges of 0 to 10,000 ppm and 0 to 100,000 ppm.

In prior studies, the EPA has determined that for a fugitive source an OVA instrument reading of greater than 200 ppm corresponded to an approximate emission rate greater than or equal to 0.5 g/hr. For this study, it was established that only sources with emission rates greater than or equal to 0.5 g/hr would be sampled. Therefore, if a hydrocarbon concentration greater than 200 ppm was detected by the instrument at any fugitive source, that source was prepared for sampling.

Pumps, Compressors, Agitators, and Valves

The OVA-128 instrument was used for screening pumps, compressors, agitators, and valves by placing the instrument probe as close as possible to the intersection of the shaft and packing gland at four points, 90° apart. If a hydrocarbon concentration in excess of 200 ppm was detected at any point, the source was prepared for sampling.

Flanges

Fugitive emissions from flanges were screened using one of two techniques depending upon the condition and accessibility of the flange.

The first method, used for screening large, relatively cool vessel and exchanger flanges, involved taping the outside perimeter of the flange interface, thus effectively sealing the interface. A small hole was then punctured into the tape seal. Hydrocarbons leaking from any point around the flange were emitted through the puncture, and they were screened in a manner similar to that discussed for screening of valves.

The second method, applicable for smaller and/or hot flanges, involved placing the OVA-128 instrument at 50-mm intervals all around and against the outside perimeter of the flange interface. All instrument readings of 10 ppm or greater were recorded. If the sum of all the readings taken around the perimeter exceeded 200 ppm, the flange was prepared for sampling.

Relief Devices, Process Drains, Sample Valves

Relief valve emissions were vented to the atmosphere through a large diameter pipe, normally located at a high point on the unit which it served. Relief valves were screened by inserting the OVA-128 instrument probe into the stack

and measuring the hydrocarbon concentration at this point. The ambient concentration was also determined. If the difference between the stack and ambient concentration exceeded 200 ppm, the relief valve was prepared for sampling.

Fugitive emissions from process drains occurred at points where the drains are vented to the atmosphere. Emissions are vented through a perforated steel plate built into the ground directly above the drain. Process drains were screened by placing the OVA-128 instrument probe near the steel plate outlet. If the instrument detected a hydrocarbon conentration greater than 200 ppm, the process drain was prepared for sampling.

Process stream samples are collected from sample valves at regular intervals by plant personnel during normal operation at a petrochemical plant. Sample valves were screened for fugitive emissions by placing the OVA-128 instrument probe near the open spout of the sample valve. If a total hydrocarbon concentration greater than 200 ppm was measured, the sample valve was identified for sampling.

SAMPLING OF FUGITIVE VAPORS

Field sampling of fugitive organic vapors was accomplished using the dilution or flow-through method. Two separate sampling trains, the vacuum sampling system and the pressurized sampling system (shown in Figures 3 and 5), were used. The trains were mounted on a portable cart for easy maneuverability from source to source.

Vacuum Sampling System

This sampling train, shown in Figure 3, was equipped with a pneumatic vacuum pump, an orifice meter and a sampling syringe. A cold trap was placed in the system to condense water and heavier hydrocarbons and to prevent condensation in downstream lines and equipment. The fugitive emission source identified through screening was prepared for sampling by tenting. In the tenting procedure, the leaking source (e.g., a valve) was enclosed in a flexible Mylar® tent as shown in Figure 4. Ambient air was drawn across the emission source for approximately 20 minutes before sample collection in order to achieve equilibrium conditions inside the tent. Steady-state conditions were determined by using the OVA-128 instrument to measure exit gas concentration.

Samples were obtained by drawing a portion of the gas from the main flow using a 500-ml glass syringe and then injecting the gas sample into an evacuated Teflon® bag. The vacuum pump was capable of maintaining a flow rate approximately 0.037 m^3/min to 0.034 m^3/min during sampling. The source emission rate was determined from the air flow rate and the hydrocarbon content of the collected sample. Ambient air samples were obtained in the vicinity of the leaking source prior to sampling and analyzed for hydrocarbon content to determine background concentrations. If the background hydrocarbon concentration was high (>10 ppm) or if the source being sampled was in close proximity to another fugitive source the pressurized sampling train was used as described below rather than the vacuum system.

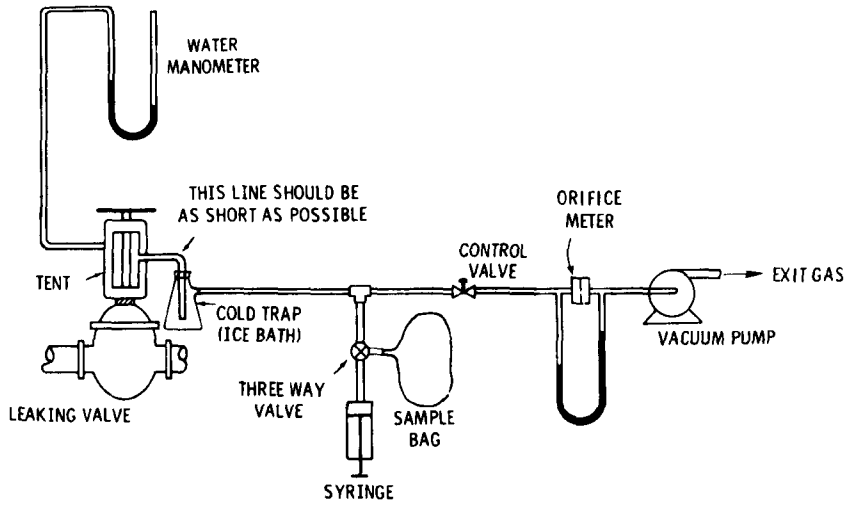

Figure 3. Portable sampling train, using a vacuum pump for sampling of fugitive hydrocarbon emissions.

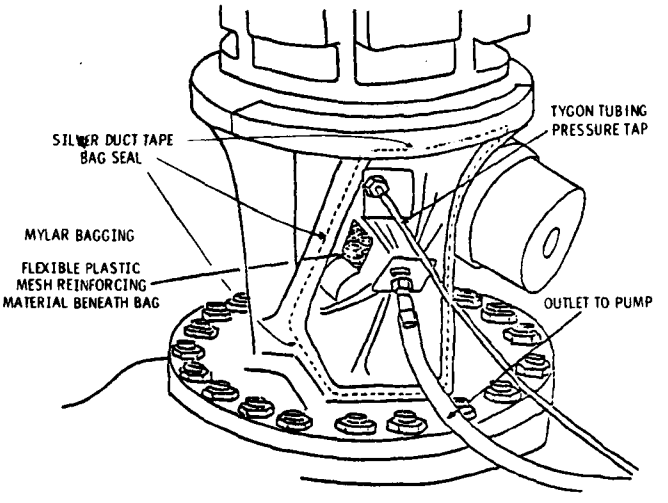

Figure 4. Tent construction around the seal area of a vertical valve.

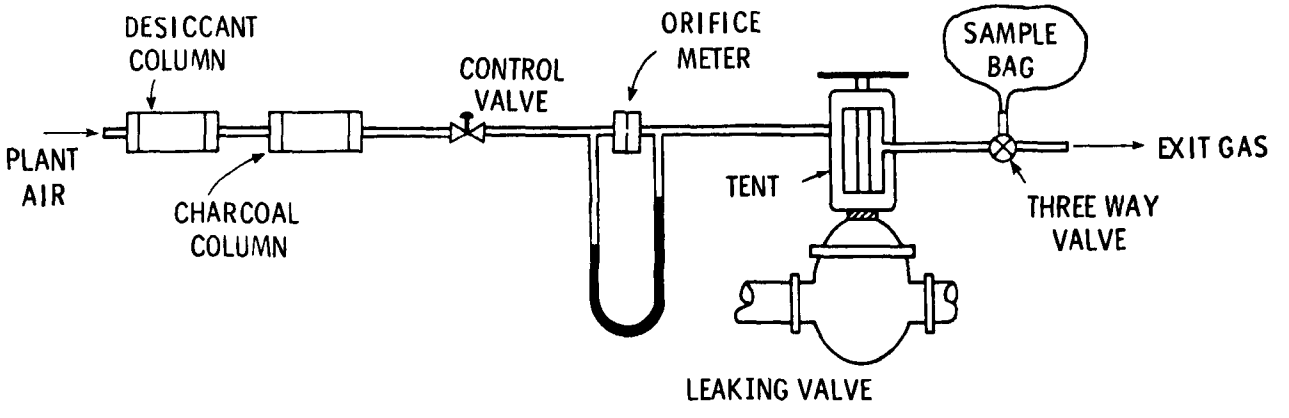

Figure 5. Portable sampling train, using plant air for dilution and sampling of fugitive hydrocarbon emissions.

Pressurized Sampling System

The pressurized sampling train shown in Figure 5 employed plant air for dilution and sampling. Plant air under pressure was first passed through a charcoal filter and desiccant to remove hydrocarbons and moisture, and then directed through the train and into the tent. After steady-state conditions were established, a sample bag was connected to the tent and filled by means of the incoming dilution air.

The vacuum system was the preferred sampling method because filling the sample bag was not dependent upon air flow through the tent. This permitted less complicated tenting procedures to be used and resulted in a substantial savings in sampling time. Also, the vacuum system was equipped with a cold trap to condense heavy hydrocarbons. If heavy hydrocarbons were to condense inside the sample bag of the pressurized system prior to GC analysis, a significant error could be introduced into the data.

The pressurized sampling system was used to sample sources located in areas where high ambient hydrocarbon concentrations existed or when fugitive sources were close to each other. In these cases it was felt that the magnitude of and variations in the background concentration would introduce errors into the emission rate calculations for the vacuum system. Dilution air in the pressurized sampling system was essentially free of hydrocarbons and water since it was passed through a desiccant and activated carbon prior to mixing with hydrocarbons from the tented source.

SAMPLING OF LIQUID LEAKS

Liquid leaks were defined as those leaks from which liquid was observed to escape. If the liquid vaporized rapidly and completely in the vicinity of the escape point, the emission was treated as a vapor leak and tented.

In order to measure the liquid leak rate at the source, the material was collected in a graduated container, which was externally cooled with ice and fitted with covers to contain most of the material. Flow rates were determined at the leak site by measuring the change in volume with respect to time.

ANALYTICAL PROCEDURES

Analysis for Individual Organic Species

After completion of a sampling run, the sample bags were transported to the field analytical laboratory. Analysis of bag contents was carried out using either a Hewlett Packard Model 5750 or a Varian 1400 gas chromatograph, equipped with flame ionization detectors. The gas chromatograph was interfaced with a Varian CDS-111 microprocessor to convert the chromatograph detector output into a digital integrated output. The system allowed storage of standard sample data; it provided data output for each sample in the form of retention times and peak areas; and it identified and quantified each component found in the samples.

Analysis for Total Hydrocarbons

If unknown hydrocarbon components were found during sample analysis, the sample was rerun and the total hydrocarbon content was measured. Total hydrocarbons were analyzed in the field by bypassing the GC column and injecting the gas sample directly into the flame ionization chamber.

Laboratory Analytical Procedures

All gaseous samples collected were analyzed in the field. Selected sample bags were returned to the laboratory for GC/MS analysis. This was done to verify field measurements and to identify any unknown components.

Liquid samples collected in the field comprised liquid leaks and condensate collected in the cold trap. These samples were returned to the laboratory for identification and quantification of each organic component present by gas chromatography/mass spectroscopy.

RESULTS

Petrochemical plants manufacturing monochlorobenzene, butadiene, dimethyl terephthalate, or ethylene oxide/glycol were selected for fugitive emission sampling since they are representative of a variety of operating conditions and unit operations. Characteristics of each process are described in Table 1.

TABLE 1. CHARACTERIZATION OF CHEMICAL PROCESSES SAMPLED IN FUGITIVE EMISSION PROGRAM

	Process			
Characterization	Monochloro-benzene	Butadiene	Dimethyl-terephthalate	Ethylene oxide/glycol
Operating temperature/pressure range	low/low	high/high	high/low	low/high
Unit Operations Utilized				
Distillation	x	x	x	x
Absorption		x		
Scrubbing/washing	x		x	
Extraction		x		
Evaporation		x		x
Crystallization			x	x
Drying			x	x
Quenching		x		
Storage	x	x	x	x
Unit Processes Utilized				
Oxidation			x	
Hydrogenation			x	
Pyrolysis		x		
Esterification			x	
Hydration				x
Chlorination	x			

Average hydrocarbon emission rates are given in Table 2 for source types in each process. These emission rates are based on sources that were found to have hydrocarbon concentrations in excess of 200 ppm as determined by screening. The estimated fugitive emission rate for all potential sources is also given; it is based on the mass of fugitive emissions divided by the total number of sources. An analysis of variance was performed to determine the effect of operating temperatures, pressures and fugitive compound vapor pressures on the variability in emission rates from each pump, valve and flange sampled. Results of this analysis indicated that the factors tested had no significance in relation to the corresponding emission rates from each sampled source.

Table 3 gives the estimated annual mass of fugitive emissions from each process. This information was derived from the number of potential fugitive sources at each plant and the estimated average emission rates for all potential sources, as shown in Table 2.

In butadiene production valves were the most significant fugitive source, contributing 90% by weight of the total fugitive emission burden from the plant. In monochlorobenzene production flanges contributed 91% of the total fugitive emissions and thus were considered the most significant source of fugitive losses at the plant. For ethylene oxide/glycol and dimethylterephthalate production data on the total number of valves and flanges were not available, and consequently no emission mass estimates were made.

Factors which determined the magnitude of fugitive emissions from various source types (i.e., pumps, valves, flanges, etc.) included 1) total number of potential sources in each process, 2) average emission rate for significant fugitive sources, and 3) percentage of sources having significant emission rates.[a] The total number of each source type was found to have the most impact on the overall fugitive emission burden for a petrochemical plant. This was due to the high variability in the total number of potential sources for different source types. For example, in the butadiene production plant sampled there were 174 pumps and 6,700 valves. Variations in the average emission rates and in the percentage of sources having significant emissions for these two source types were small compared to the differences in number of sources. Thus the total emissions contributed by valves is very much larger than the total fugitive emissions from pumps. Since the total number of valves and/or flanges was large compared to the number of other source types (pumps, compressors, etc.),at each plant sampled, valves and/or flanges are considered the major sources of fugitive emissions from petrochemical processes.

Fugitive emissions were identified and quantified in the field by gas chromatographic analysis. Table 4 lists the organic compounds detected along with their weight percent contribution to the total mass of fugitive emissions determined for each process. On-site analysis was verified by laboratory mass spectroscopy; it was determined that greater than 98% by weight of the fugitive emissions sampled were identified. Potential human health effects from exposure to various compounds detected are described in footnotes in Table 4.

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[a]Significant fugitive sources are those having a hydrocarbon emission rate greater than or equal to 0.5 g/hr.

TABLE 2. HYDROCARBON EMISSION RATES FOR FUGITIVE SOURCES BY PROCESS
lb/hr (g/hr)

Source type	Average emission rate for significant fugitive sources[a]				Average emission rate for all potential sources[b]			
	Monochlorobenzene	Butadiene	Dimethyl-terephthalate	Ethylene oxide/glycol	Monochlorobenzene	Butadiene	Dimethyl-terephthalate	Ethylene oxide/glycol
Pump seals	5.1×10^{-2} (23)	3.7×10^{-1} (160)	4.5×10^{-2} (20)	1.8×10^{-1} (82)	1.7×10^{-2} (7.7)	1.4×10^{-1} (63)	7.4×10^{-3} (3.3)	2.9×10^{-2} (13)
Compressor seals		1.3×10^{-1} (59)		2.5×10^{-2} (11)		1.2×10^{-1} (54)		1.3×10^{-2} (5.9)
Valves	3.4×10^{-3} (1.5)	2.5×10^{-1} (120)	7.0×10^{-2} (32)	3.5×10^{-3} (1.6)	1.0×10^{-4} (0.05)	3.7×10^{-2} (17)	3.3×10^{-3} (1.5)	1.6×10^{-4} (0.07)
Flanges	1.8×10^{-1} (82)	0 (0)	2.4×10^{-1} (110)	2.0×10^{-3} (1.0)	4.9×10^{-3} (2.2)	0 (0)	7.6×10^{-3} (3.4)	6.7×10^{-5} (0.03)
Relief devices		3.0×10^{-2} (14)	0 (0)	0 (0)		1.4×10^{-2} (5.0)	0 (0)	0 (0)
Process drains				1.5×10^{-1} (68)				8.9×10^{-2} (40)
Agitator seals	4.4×10^{-1} (200)		4.8×10^{-1} (218)		4.4×10^{-1} (200)		3.2×10^{-1} (145)	
Sample valves			2.0×10^{-1} (91)				8.6×10^{-2} (40)	

Note.—Blanks indicate source type nonexistent in process.

[a] Significant fugitive sources are those having an emission rate greater than or equal to 0.5 g/hr as determined by sampling and analysis.

[b] Emission rates were determined by calculating the mass of fugitive emissions from the emission rates for significant sources. The mass of emission was divided by the total number of sources screened to arrive at an average fugitive emission rate for all sources.

TABLE 3. MASS OF FUGITIVE EMISSIONS FROM PETROCHEMICAL PROCESSES

Process source	Number of sources present	Estimated mass[a] of emissions, metric tons/yr	Total mass of fugitive emissions, metric tons/yr
Monochlorobenzene			31.9
Pumps	25	1.0	
Valves	640	0.2	
Flanges	1,500	29	
Other	-[b]	1.7	
Butadiene			1,112
Pumps	174	96	
Valves	6,700	1,000	
Flanges	26,000	0	
Other	218	16	
Ethylene oxide/glycol			-[b]
Pumps	69	8.0	
Valves	-[b]	-[b]	
Flanges	-[b]	-[b]	
Other	26	7.9	
Dimethylterephthalate			-[b]
Pumps	67	2.0	
Valves	-[b]	-[b]	
Flanges	-[b]	-[b]	
Other	37	8.6	

[a]Determined from average emission rate from all potential sources as shown in Table 2.

[b]Data not available.

TABLE 4. PERCENT COMPOSITION BY WEIGHT OF FUGITIVE EMISSIONS FROM PETROCHEMICAL PLANTS

Compound	Monochloro-benzene, %	Butadiene, %	Dimethyl Terephthalate, %	Ethylene Oxide/glycol, %
Acetaldehyde[a]				11
Acrolein[a]				12
Benzene[b]	41			
i-Butane		0.5		
n-Butane		1.0		
1,3-Butadiene[a]		28		
1-Butene		11		
2-Butene		1.5		
Dichlorobenzene[a]	0.1			
Diisobutene[a,c]		33		
Ethane				<0.1
Ethene				2
Ethylene oxide[b]				73
Furfural[a]		4		
Isobutene		21		
Methane				2
Methanol[c]			65	
Monochlorobenzene[a]	59			
Xylene			35	

[a]Human health effect - irritant

[b]Human health effect - suspected carcinogen

[c]Human health effect - narcotic

Table 5 provides a comparison of fugitive emission rates for petrochemical plants and petroleum refineries. Emission rates for petroleum refineries were derived from emission factors obtained during the sampling of fugitive sources by Los Angeles County (Ref. 1-4). The percentage of sources having emission rates in excess of 0.5 g/hr for both petrochemical plants and petroleum refineries is given in Table 6.

For significant fugitive sources, average emission rates at the petrochemical plants sampled were 3% to 90% lower than corresponding emission rates for petroleum refineries. Average emission rates for all potential sources were also found to be lower at the petrochemical plants than the established emission rates for refineries. Of the total number of sources sampled, the percentages of significant sources were lower for monochlorobenzene, dimethyl-terephthalate, and ethylene oxide/glycol production when compared to refineries. For butadiene production, however, the percent of sources having significant emission rates is essentially the same as that for refineries.

TABLE 5. COMPARISON OF FUGITIVE EMISSION RATES FOR PETROCHEMICAL PLANTS AND PETROLEUM REFINERIES
lb/hr/source (g/hr/source)

Source type	Significant fugitive sources[a]		All potential sources	
	Petrochemical Plants	Petroleum Refineries (Ref. 1-4)	Petrochemical Plants	Petroleum Refineries (Ref. 1-4)
Pump seals	1.6×10^{-1} (73)	4.8×10^{-1} (218)	4.8×10^{-2} (22)	1.7×10^{-1} (78)
Compressor seals	7.7×10^{-2} (35)	6.5×10^{-1} (300)	6.6×10^{-2} (30)	3.5×10^{-1} (161)
Valves	8.2×10^{-2} (37)	8.5×10^{-2}[b] (38)	1.0×10^{-2} (4.5)	1.1×10^{-2}[b] (5)
Flanges	1.0×10^{-1} (45)	1.1[b] (500)	3.1×10^{-3} (1.4)	1.1×10^{-2}[b] (5)
Process drains	1.5×10^{-1} (68)	-[c]	8.9×10^{-2} (40)	-[c]
Relief valves	3.1×10^{-2} (14.3)	4.0×10^{-1} (183)	1.1×10^{-2} (5.1)	1.0×10^{-1} (45)

[a] Significant fugitive sources are those having an emission rate greater than or equal to 0.5 g/hr.

[b] Data shown arebased on an average emission rate of 0.27 lb/day/source. EPA currently uses a value of 0.15 lb/day/source for valves and flanges; however, this excludes sources with large leaks.

[c] Data not available.

In Tables 5 and 6 petrochemical plant fugitive emissions are shown to be generally lower in both magnitude and in the number of significant sources when compared to data on refineries. Two factors which may affect the variations in fugitive emissions are given below:

- process size - the petrochemical processes sampled were smaller in size than refinery operations and contained a fewer number of potential fugitive sources.
- product value - products from petrochemical plants have a higher value per unit than refinery products. Thus potential losses are better controlled.

TABLE 6. COMPARISON OF SIGNIFICANT FUGITIVE SOURCES IN PETROCHEMICAL PLANTS AND IN PETROLEUM REFINERIES[a,b]

	Percent of sources having significant emission rates				
	Monochloro-benzene	Butadiene	Dimethyl-terephthalate	Ethylene oxide/ glycol	Petroleum Refineries (Ref. 1-4)
Pump seals	20	39	16	16	36
Compressor seals		95		50	54
Valves	3	15	5	4.5	13
Flanges	3	0	3	3	1
Process drains				59	-[c]
Relief valves		36	0	0	23

[a] Significant fugitive emission sources are those having an emission rate greater than or equal to 0.5 g/hr.

[b] Blanks indicate source type does not exist in process.

[c] Data not available.

CONCLUSION

This study has identified and quantified fugitive emissions from various petrochemical plant processes. Fugitive emissions from petrochemical plants are generally lower in quantity when compared to EPA data on fugitive emissions from petroleum refineries.

Physical differences in operating conditions and process materials showed no relationship on variations in emission rates from individual sources.

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DEVELOPMENT OF COATINGS TO REDUCE FUGITIVE EMISSIONS FROM COAL STOCKPILES*

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ABSTRACT

The use of coatings to prevent dust emissions and leachates from coal stockpiles was studied under sponsorship of the Department of Energy, Division of Environmental Control Technology. The coatings considered included compositions formed of a filler such as pulverized coal and a binder consisting of a combination of waxes and plastics. Various latex emulsions, both with and without fillers, were also evaluated.

The purpose of coal coatings is to seal the surface of the stockpile. This prevents water penetration into the coal. By sealing the surface of the pile, dust losses are prevented and leachate formation is minimized. Air circulation through the stockpile is also greatly reduced. This yields the added benefits of reduced oxidation of the coal and prevention of spontaneous ignition. Cold weather handling characteristics of the coal are also improved.

Physical properties of the coatings were measured. Included were such characteristics as resistance to water penetration and degradation as a result of thermal cycling. Application techniques were also evaluated. Both hot-applied and cold-applied coatings were considered.

Protective coatings appear to be an attractive means of prevention of fugitive emissions from coal stockpiles. The economic benefits from reduced oxidation and improved handling more than off-set the cost of application. Use of coatings on unit trains or barges may also be attractive.

INTRODUCTION

Coal from mines is transported to its ultimate destination by rail, truck, barge or pipeline. Once it arrives at its consumption point or a processing facility, it is moved over short distances by rubber-tired loaders, conveyors or rail shuttle cars. During the transportation process, the coal is often exposed to the elements. This leads to dust emissions and losses through wind effects. Moisture absorption and oxidation also can occur, causing deterioration of the coal and loss in fuel value (ERDA, 1976).

Once at the destination, the coal is either used or stored. Most users maintain stockpiles comprising a 90-day supply at their normal use rate. This is done primarily to provide a reserve in the event of temporary loss of supply. This stockpile may stand for a year or more before it is necessary to use it.

In the stockpile, the coal is subject to weathering and oxidation. Heavy dust emissions from stockpiles are common. Absorbed moisture from rain or snow causes leachate formation and may decrease the fuel value of the coal by 1 to 8% depending upon initial moisture level and coal rank. Wind and rain erosion results in coal migration and exposes new coal surface to weather. Moist coal can also freeze solid in cold weather making normal handling impossible. Oxidation may reduce the heating value up to 5% over a one-year storage period (ERDA, 1976). In addition, internal oxidation can lead to spontaneous combustion in low rank fuels (ERDA, 1975; Paulson et al., 1975).

This work presented here addresses the use of protective coatings for coal to resolve the environmental problems associated with transport and storage. These coatings not only minimize fugitive emissions, but can also be shown to provide dollar savings substantially greater than the costs of application.

TECHNICAL APPROACH

The overall objective of this work was to demonstrate the feasibility of economical coal coating systems. To accomplish this, the following factors were considered:

- Identification of suitable materials for coal coatings
- Development of formulations which are capable of reducing the adverse environmental effects of weathering of coal stores
- Identification of acceptable application techniques
- Tests to determine coating properties and provide data for economic analysis
- Economic analysis of coatings compared to unprotected coal
- Cost/benefit analysis

The first task was intended to provide information to guide the experimental development of coal coatings. The factors considered are listed below:

- Evaluation of candidate filler-binder materials
- Review of current coal protection techniques
- Application considerations
- User motivation

Filler-Binder Materials Evaluation

A list of materials was generated which could be considered as candidates for inclusion in coal coating formulations. The candidate materials were screened to establish their characteristics with respect to:

- cost
- availability
- fuel value
- physical properties
- environmental pollution potental

The filler materials considered included coal, paper, sawdust, bagasse, rice hulls, cottonseed hulls, and fly ash. Binder materials considered fall into two categories. The first included the materials useful for the hot-melt type of coating such as waxes and plastics. The second type of binder material was the various latex emulsions. This type of material was considered for use in filled latex coatings.

Pulverized coal and fly ash emerged as the best filler candidates. Both of these materials are available at coal user sites in sufficient quantities. They are both available at little or no cost, taking into account the credit for fuel value of coal. Neither of these materials would cause additional environmental problems upon combustion, as both are normally present in coal combustion systems. The other filler material candidates were either too costly, limited in availability or presented potential environmental problems.

The best binder materials identified were slack wax, polyethylene, atactic polypropylene for the hot-melt composition, and polyvinyl acetate copolymers for the filled latex formulations.

Current Coal Protection Techniques

The most common current means of protecting coal is to carefully build and compact the stockpile, then monitor it for hot spots. At this time, it is estimated that less than 5% of the users of coal provide additional protection in the form of coatings. Coal is shipped by rail car in as-dumped condition. It is estimated that 5-10% of the shippers use latex crusting compounds to prevent wind loss and dust emissions in transit. A discussion of these current protection methods is provided below.

Stockpile Formation

Large users of coal generally maintain a stockpile sufficient to provide coal for 90 days in the event of a strike or other loss of supply. A typical user requires 4,000 tons per day. Thus, a stockpile of 360,000 tons would be required.

Stockpiles must be properly constructed; otherwise, the risk of spontaneous combustion is high. Coal stores have been known to ignite spontaneously within six days after pile formation (Wilson, 1975). A typical method for constructing a coal stockpile is given below:

1. The coal is dumped from the rail cars either by bottom hopper or car inversion.

2. The coal falls onto a conveyor and passes through a mill where it is ground to <2-inch size pieces.

3. The coal is transported to the stockpile areas either by conveyor or by rubber-tired vehicle.

4. A bulldozer spreads the coal and compacts it. Typical piles are in the form of a truncated cone, 100-200 feet wide at the base and 25-50 feet high. The length of the pile may be up to thousands of feet.

Coal as dumped has a bulk density of about 50 lb/ft^3. After compaction, the bulk density is about 65 lb/ft^3 (Paulson, et al., 1975).

The stockpiling technique varies with coal source and rank. Higher rank coal may be formed into larger piles of greater height and stored longer than low rank coals (ERDA, 1975). East coast coals may be stored longer than west coast coals as they are less reactive. The finer the coal, the more reactive it is as a result of greater surface area.

Small coal stockpiles may be stored as dumped for short periods. Such piles should not exceed 15 feet in height (Wilson, 1975). Longer term storage of uncompacted coal is risky in terms of spontaneous combustion danger. Uncompacted coal piles should be used within a few days of dumping.

Stockpile Protection

Stockpiles are formed solely by the action of a bulldozer in shaping and compacting the pile. Thermal probes are sometimes buried in the coal to monitor pile temperatures. The probes set off an alarm if the pile temperature reaches 130-140°F (54-60°C).

Dust emissions from coal stockpiles are a problem of increasing concern. Some users reduce dust formation by washing the coal before piling and transporting it wet. Other users wet the pile after formation or spray it with oil to reduce dust emissions.

Leachate formation is also a serious problem except in the case of very high grade coal. Water runoff from the pile is collected in a trough

around the pile. It is then sent to a holding tank or pond where it is treated before discharge to the environment. The treatment generally consists of introducing a flocculant or precipitating agent, then allowing the solids to settle out before discharge.

Few users currently protect their stockpiles against oxidation and the resultant fuel value losses. ERDA (1976) reports that losses up to 5% may occur in the first year of storage. Leonard (1968) found that properly stored coal will lose only 1-2% per year of its energy value due to oxidation. However, improper storage was found to result in losses of 3-5% per year.

Latex Crusting Compounds

The only protective coating of significance in current use is the latex crusting compound. This material is essentially a paint base, and it is produced by paint manufacturers. Numerous small firms purchase this material and resell it as a protective coating for coal or as a surface stabilizer for earth, sand or other materials stored in stockpiles. The latex compounds are normally applied after dilution by a factor of 3/1 to 20/1. They are sprayed onto the material to be protected using any equipment capable of spraying water.

Formulation and Evaluation of Coatings

During this work, 119 hot-melt formulations and 39 latex mixes were produced and evaluated. The types of tests conducted were as follows:

Visual observation	Viscosity
cracking tendency	Density
surface texture	Thermal expansion
adhesion to substrate	Water permeability
Compressive strength	Rheology
Tensile strength	Grindability

Not all tests were run on all formulations. Some tests, such as tensile and compressive strength, were run on a limited number of specimens to establish the order of magnitude of the results. No means of converting strength data into a useful parameter for characterizing the utility of coatings was identified. Thus, the data are of interest primarily as a means of estimating cracking potential.

Other tests, such as water permeability, effects of thermal cycling and coefficient of thermal expansion relate directly to the quality of the coatings. Tables I and II summarize the results of these tests on formulations that were near the optimum compositions for both the hot-melt and filled latex coatings.

Test Panels

Those formulations which appeared to best meet the above criteria were cast onto frames, 12" x 12", with about 3-4" of coal as a substrate.

Table I. Physical Properties of Hot-Melt Formulations

ENTRY NO.	COMPOSITION	VISCOSITY Avg. KP RPM 1	2.5	5	TENSILE PROPERTIES (-30°F) At Rupture Stress (PSI)	Strain (%)	Tangent Modulus (PSI)	COMPRESSIVE STRENGTH, (70°F) at Failure (PSI)		STRAIN IN COMPRESSION At Failure (%)	DENSITY GM/cm^3 (70°F)
2-16-4	85.0% Coal							333		1.9	
	12.0% SW							392	Avg 342	1.3	
	3.0% PE							302		1.6	
2-17-1	85.0% Coal							248		1.8	
	12.0% SW							278	Avg 258	2.1	
	3.0% PE							249		2.1	
2-17-2	85.0% Coal							148		2.2	
	12.0% SW							179	Avg 173	1.9	
	3.0% PE							192		2.3	
5-2-3	77.5% Coal	8	7	4	210	0.4	106,000				1.1
	14.9% SW				79	0.7	18,200				
	3.8% PP										
	3.8% PE										
6-1-2	80.0% Coal				222	0.4	84,800				1.08
	15.2% SW				233	0.6	68,200				
	2.4% PP				241	0.5	83,600				
	2.4% PE										

SW = Slack Wax

PE = Polyethylene

PP = Polypropylene

Table II. Characteristics of Hot-Melt and Filled Latex Compositions

REFERENCE NUMBERS	LEAK RATE, % OF SPRAY 70°F	+160°F	-30°F	36" CTE STRIP Shrinkage	Reheat Shrinkage	AVERAGE CTE	RESTRAINED ENDS GAP	GRINDABILITY
5-1-2 80.0% Coal 13.4% SW 3.3% PE 3.3% PP	3.3	8.1	11.0	0.335"	-	7.6×10^{-5}	-	Fair
5-2-3 77.5% Coal 14.9% SW 3.8% PE 3.8% PP	1.4	5.2	8.0	0.305"	-	6.6×10^{-5}	-	Fair
6-1-2, 5-30-1 80.0% Coal 15.2% SW 2.4 PE 2.4 PP	14.0	20.0	78.0	0.285"	0.278"	6.2×10^{-5}	0.118"	Fair
48 50.0% Coal 23.0% Vinyl acetate copolymer 27.0% Water	7.7	1.8	1.2					Good

SW = Slack Wax

PE = Polyethylene

PP = Polypropylene

The frames were constructed with wooden pegs spaced at 3/4" intervals around the periphery. The pegs served to restrain the coating from shrinkage to enhance crack formation and present a more realistic test than would an unrestrained coating. The coating was applied in thicknesses ranging from 1/8" to 1/2". Figure 1 shows two views of a test frame with a hot-melt coating in place. Figure 2 shows a cross-section view of the coating on a coal bed.

The test frames were subjected to a "rainfall" test. In this test, the equivalent of 2" of rain was sprayed on the surface of the test panel. The quantity of water adhering to the surface or passing through the coating was measured. For the best coatings, less than 10% of the water was retained or passed through the coating. The remainder was repelled and ran off the coating surface. The water permeability tests were generally run 3 times, at ambient temperature and after exposure to temperatures of +160°F and -30°F.

Thermal Expansion Tests

Thermal expansion or contraction is an important characteristic of hot-melt coatings. If the degree of contraction upon cooling exceeds the strain-to-failure of a coating material, cracking will result. Similarly, if the thermal expansion upon heating exceeds the compressive strain-to-failure, the coating will shatter.

The coatings listed exhibited Coefficients of Thermal Expansion (CTE) ranging from 6×10^{-5} to 14×10^{-5} in/in°F. Thus, for a 100°F temperature change, the degree of expansion or contraction would be about 0.6% to 1.4%. The tensile strain to failure was measured to be about 0.5%. Thus, most coatings would be expected to crack upon cooling by 100°F or more. Heating does not appear to be a problem. Compressive strain-to-failure was measured to be about 2%, and none of the coatings tested expanded that much.

The CTE is not the only factor in cracking of the hot-melt coatings. The materials were generally cast at 200-250°F. In general, the coatings remained fluid until they cooled to below 200°F. They then start to shrink, if unrestrained. Total shrinkage upon cooling to 70°F ranged from 0.6% to 1.7%. The cooling behavior was a function of the composition, however. The CTE curves for typical compositions are shown in Figures 3 and 4. Formulations with CTE values similar to that shown in Figure 4 would probably be satisfactory for stockpile applications.

Latex Formulation Results

A total of 39 latex formulations were prepared and evaluated during this work. Included in this number were four tests of commercially available latex crusting compounds for comparison purposes. The remainder of the formulations were filled latexes, in which pulverized coal was used as an extender to improve the waterproofing character of the latexes.

Some of the latexes tested were not compatible with coal. In the presence of coal, they coagulated or solidifed rapidly. Daratak SP-1065, Everflex GT and DLR-H resins yielded satisfactory coating films. All others tested were incompatible with the coal.

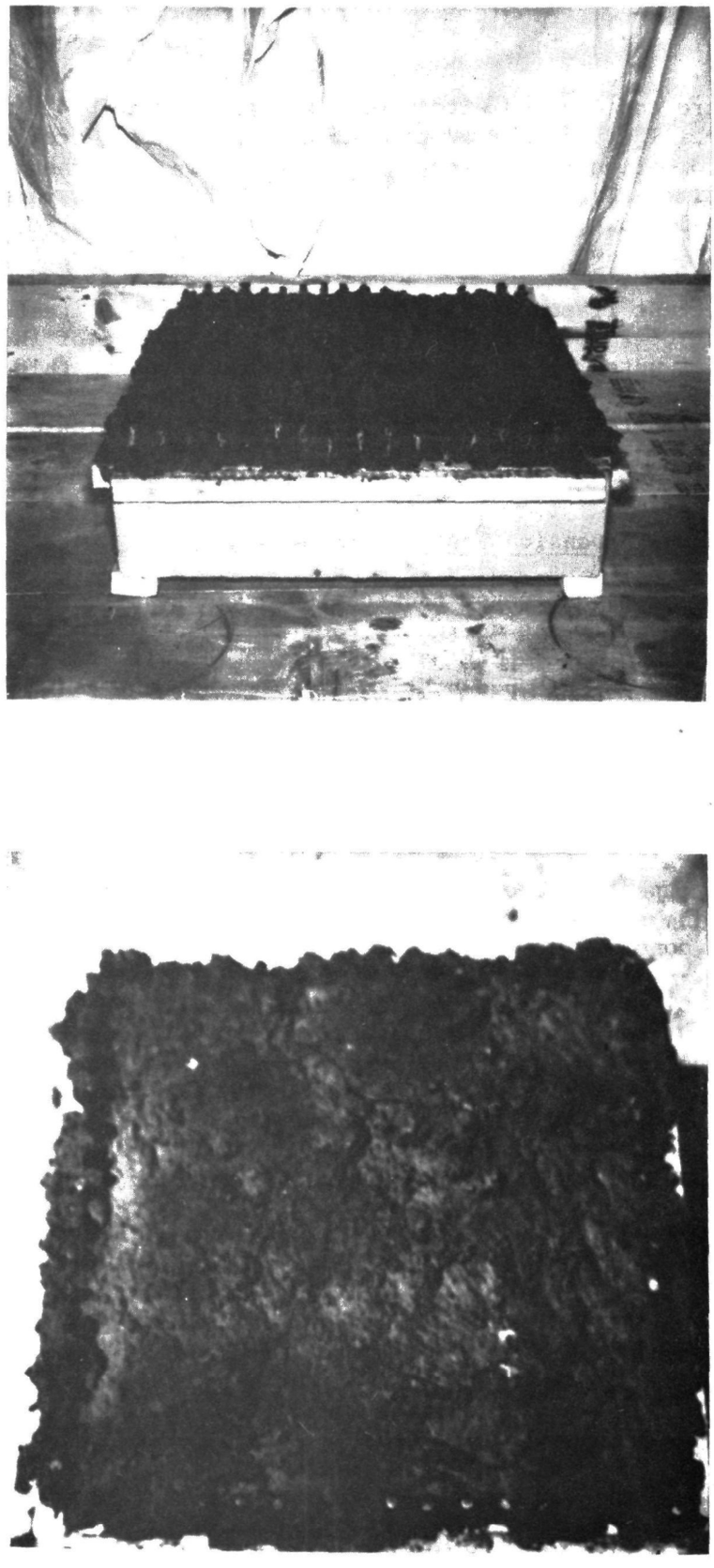


Figure 1. Hot-Melt Coating on Test Frame

Figure 2. Cross-sectional View of Hot-melt Coating on Coal Bed

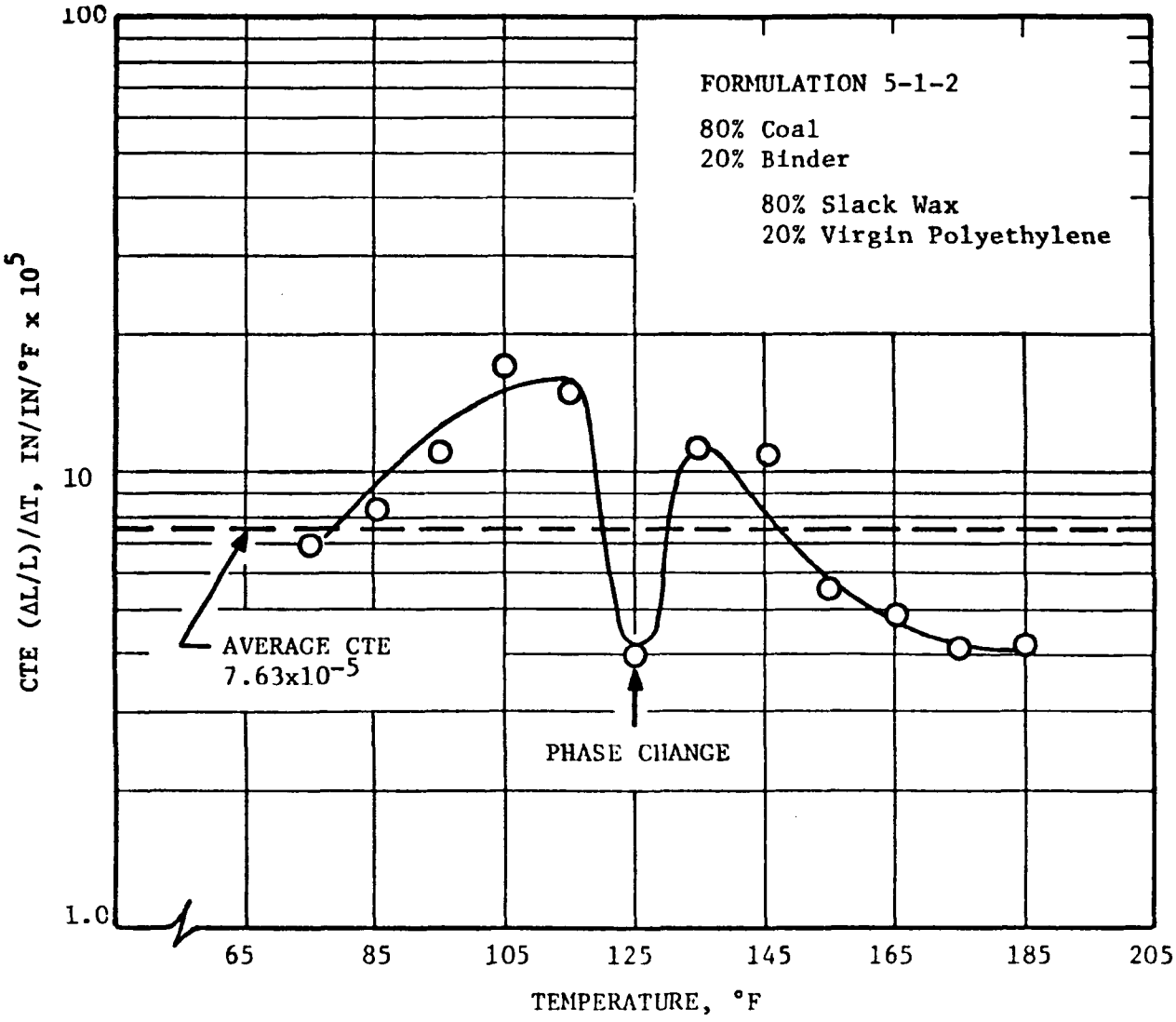


Figure 3. Coefficient of Thermal Expansion Curve for Formulation 5-1-2

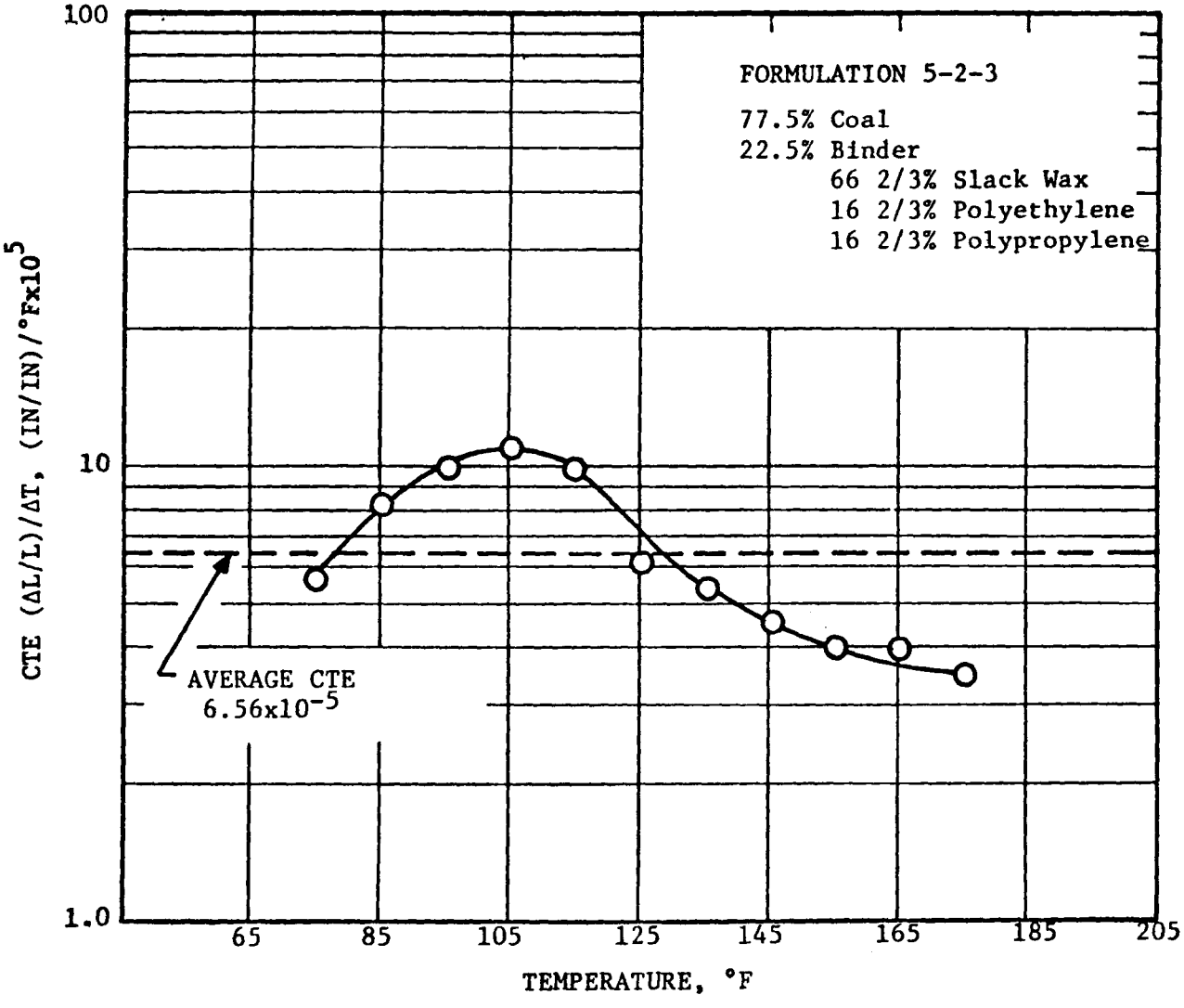


Figure 4. Coefficient of Thermal Expansion Curve for Formulation 5-2-3

The probable reason for the observed incompatibility is de-emulsification of the resin by chemical reaction with ionic components of the coal. Another possibility is instability resulting from pH change because of acidic substances in the coal.

Filled latex coatings made with polyvinyl acetate resins were applied to 1-ft x 1-ft panels similar to those previously described for the hot-melt coatings. All of the coatings were leakers in the "rainfall" test. The best of these coatings was an Everflex GT mix with 50% coal, which exhibited only a 13% leak rate.

To improve on the leakage rate with latex coatings, several test panels were made using a specially prepared surface finish on the substrate coal. One-half inch of fines, with a particle size of <1/8" was placed upon the surface before the coatings were applied. Both commercial crusting compounds and an Everflex GT filled latex coating were tested. The unfilled latex sprays did not waterproof the surface even with special preparation. In all cases, 50-90% of the "rainfall" water penetrated the coating. The filled latex coating did form a cohesive film on the surface, however. Less than 8% of the "rainfall" water adhered to the surface or passed through the coating, even after exposure to a temperature range of -30°F to +160°F. In the economic analysis, presented later in this paper, filled latex coatings on both normal and specially prepared surfaces are considered for comparison purposes.

Tests on commercial latex crusting compounds showed that they do not provide a significant degree of waterproofing to coal. The minimum leak rate noted was 78%. One commercial product was tested at the recommended concentration and again at 5 times the recommended amount. The leakage rate was similar in both cases. The unfilled latexes cannot be considered as a satisfactory waterproofing agent for coal stores even when applied at substantially higher levels than recommended by the manufacturers for dust control and surface stabilization.

Application Techniques

Application experiments were conducted to assess characteristics of hot-melt materials. The principal objective was to assure that the materials could be pumped without problems due to viscous effects or vapor lock. Initial experiments showed that compositions containing 80% coal were easily transported through a one-inch diameter tube with less than 2 psig pressure drop.

A system capable of applying hot-melt materials comprises the following elements:

a) crusher/grinder

b) polymer melt vessel

c) coal preheater

d) metering system

e) mixing vessel (heated)

f) slurry pump (heated)

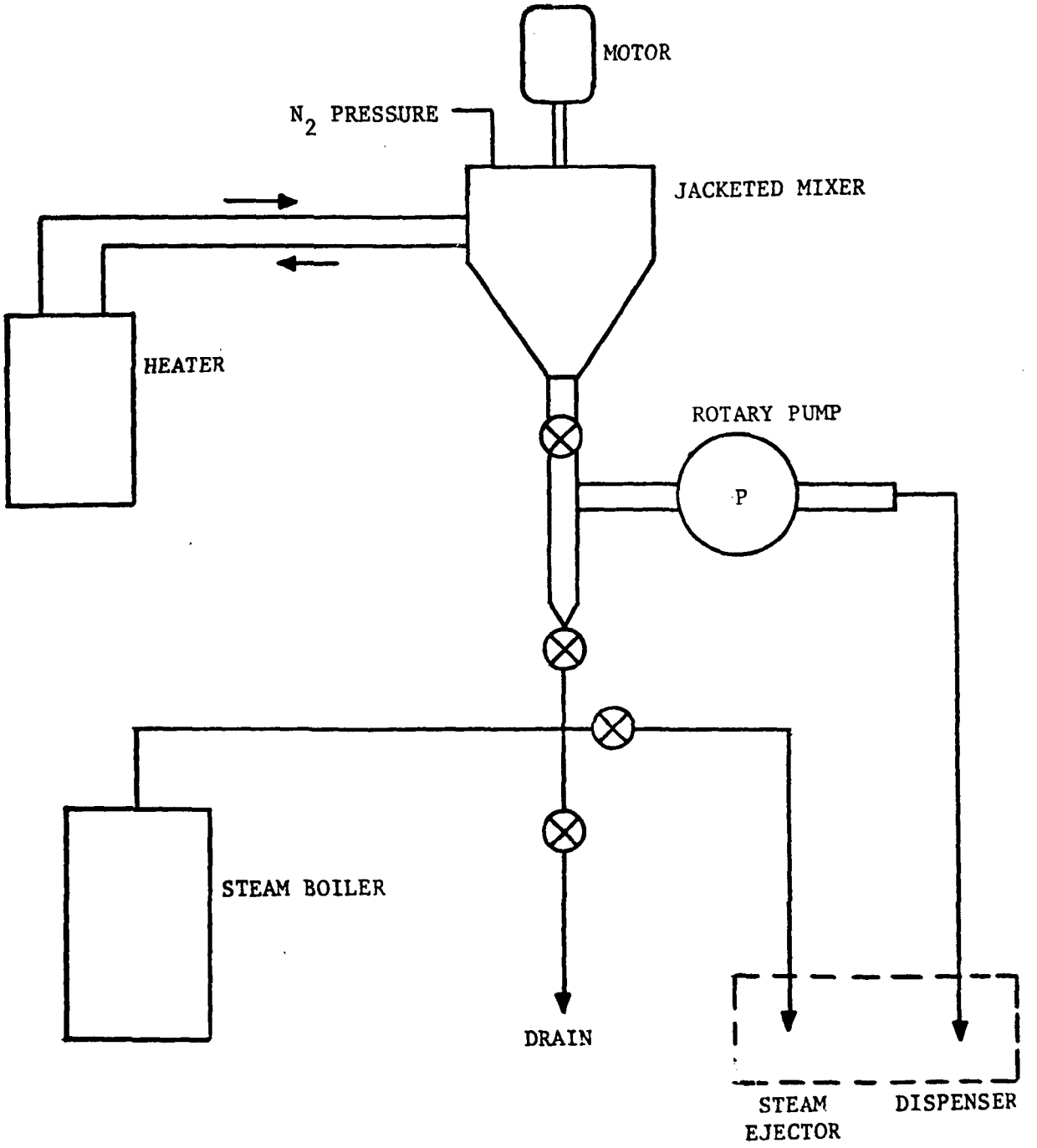


Figure 5. Schematic Diagram: Coal Cover Application System

g) service lines (heated)

h) steam boiler

i) dispensing system

A schematic of the system is shown in Figure 5. As shown, a mixing vessel is included in the current subscale apparatus. This mixer is used as a batch heater and dispenser. For larger-scale applications, a continuous system would be more desirable.

Hot flow application resulted in an excellent layer of coating on an inclined 4 x 8 foot panel. The material thickness varied from 1/8- to 1/4-inch thick. Figure 6 shows a technician in the process of coating a 4' x 8' panel.

ECONOMIC ANALYSIS

The following section is an evaluation of the costs and benefits which would accrue from the use of protective coatings for coal. Uses of latex crusting compounds, filled latex coating and hot-melt coatings are considered. Both stockpiles and rail car applications are evaluated. The steps involved in forming and protecting a stockpile are shown in Figure 7.

Summary of Stockpile and Rail Car Protection Costs

Table III shows a summary of the costs of building and protecting a coal stockpile. A 250,000 ton stockpile was used as a basis for comparison. Table IV is a summary of the costs of various techniques for protecting coal in rail cars.

Cost/Benefit Analysis - Stockpile Applications

This section illustrates the benefits to be derived from coal protection and the estimated return on investment. This analysis is based upon protection of a 250,000 ton stockpile for a period of one year. The value of the coal is assumed to be $20 per ton.

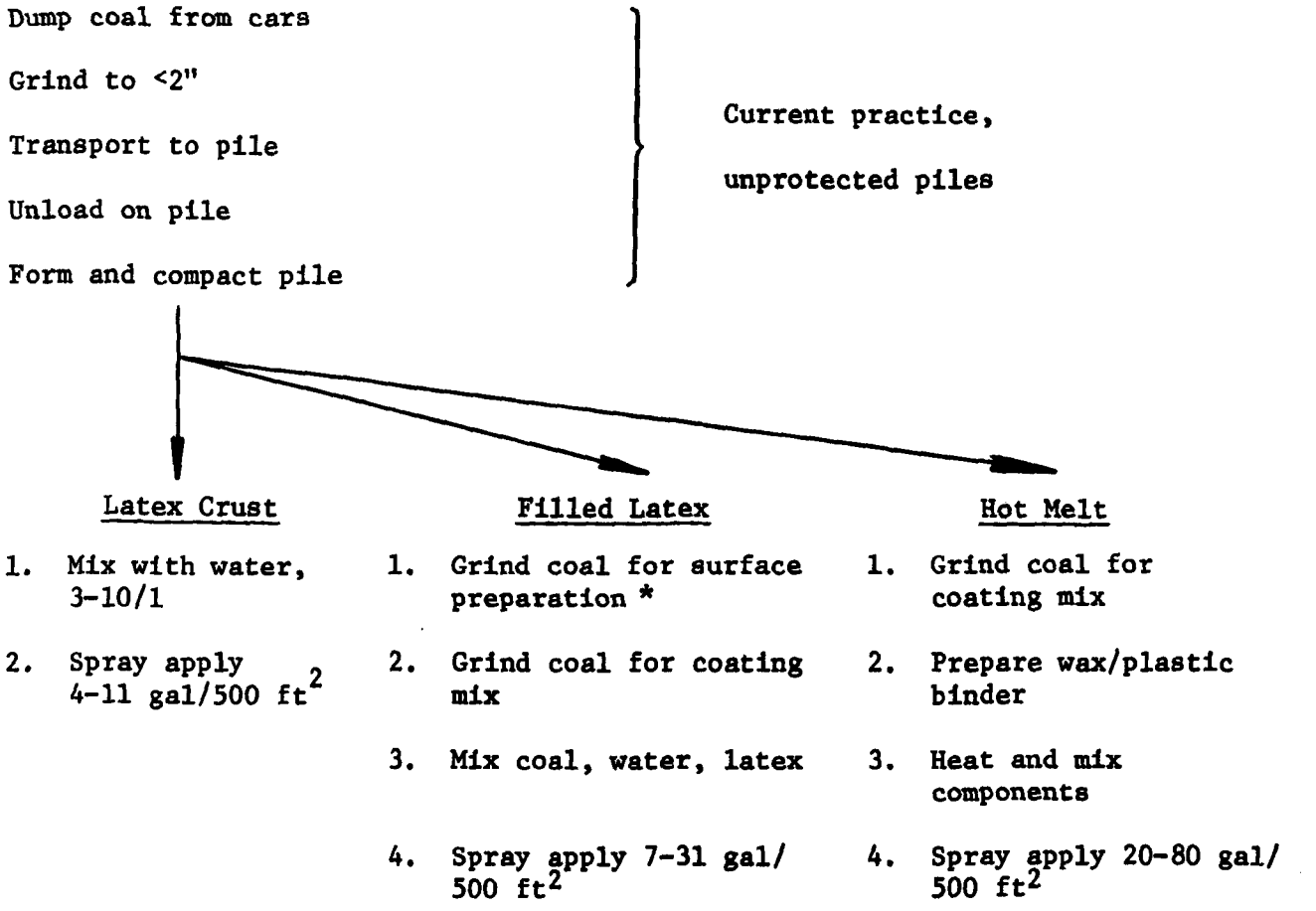
The cost factors used in determining treatment costs are listed below.

	Cost, $/Ton
Build and compact pile	0.08
Apply latex crusting compound	0.034
Apply filled latex coating, 0.025"	0.078
Apply hot-melt coating, 1/8"	0.051

The benefit factors used in determining the return on cost of applying particular coatings are:

Reduce or eliminate dust emissions.
Reduce or eliminate leachate formation.
Maintain or reduce moisture content (fuel value effect).
Minimize freezing of coal into large agglomerates.
Prevent increase in moisture content (grinder operation).
Prevent stockpile migration.
Reduce or eliminate oxidative energy loss.

Figure 6. Application of Hot-Melt Coatings



* optional

Figure 7. Coal Stockpile Formation and Protection

Table III. Summary of Stockpile Protection Costs

Basis: 250,000 ton stockpile

Treatment	Time Required Days	Total Costs $	Cost/ton $
Build and Compact Stockpile	50	55,000	0.22
Monitor Stockpile for 1 year	365	22,750	0.091
Latex Crusting Compound, 1 gal/ 200 ft^2	3.5	8,400	0.034
Filled Latex on Surface Fines, .025"	13.3	19,500	0.078
Filled Latex on Surface Fines, .050"	26.6	25,908	0.107
Filled Latex on Surface Fines, .100"	53.2	42,000	0.168
Filled Latex on Normal Surface, 1/8"	11.7	33,506	0.134
Hot-melt on Normal Surface, 1/8"	11.7	12,788	0.051
Hot-melt on Normal Surface, 1/4"	23.4	25,576	0.102

Table IV. Summary of Rail Car Protection Costs

Basis: 1 unit train = 100 rail cars
containing 100 ton/car
treatment rate = 100 cars/day

Treatment	Total Cost $	Cost/ton $
Latex Crusting Compound, 1 gallon/500 ft^2	700	0.07
Filled Latex on Surface Fines, .050"	3,844	0.38
Filled Latex on as Dumped Surface, 1/8"	4,320	0.43
Hot-Melt on as Dumped Surface, 1/8"	1,355	0.14

Reduce or eliminate spontaneous ignition.
Prevent wind or rain erosion.
Reduce need for snow removal.
Reduce monitoring costs.
Reduce fire prevention and extinguishment requirements.

The basis for determining the dollar value or equivalent for these benefit factors is described in Kromrey, et al., (1978). The results are summarized below.

Treatment	Treatment Costs, $/ton	Direct $ Savings, $/ton	Total Benefits Value, Equivalent $/ton
Compaction	0.08	0.275	0.61
Latex Crust*	0.114	0.65	1.09
Filled Latex Coating*	0.158	1.04	1.65
Hot-melt Coating*	0.131	1.04	1.90

* includes compaction

From this table, the highest return per unit cost occurs by use of the hot-melt formulation, i.e., return of benefits valued at $1.90 per ton at a cost of 13.1¢ per ton.

Cost/Benefit Analysis - Rail Car Applications

This section illustrates the benefits to be derived for protection of coal in rail cars and the estimated return on investment. The results are based upon protection of a unit of 100 cars containing 100 tons each of coal. The value of the coal is assumed to be $20 per ton. The benefit factors considered are listed below:

Reduce or eliminate dust emission.
Reduce wind losses.
Prevent moisture increase (fuel value effect).
Prevent moisture increase (grinder operation).
Prevent freezing of coal into large agglomerates.
Reduce or eliminate spontaneous combustion.

Again, the basis used to convert these factors into an equivalent dollar value is described in Kromrey, et al., (1978). The results are summarized in the following table.

Treatment	Treatment Costs, $/ton	Direct $ Savings, $/ton	Total Benefit Value, Equivalent $/ton
Latex Crust	0.07	0.54	0.74
Filled Latex Coating	0.38	0.61	0.81
Hot-melt Coating	0.14	0.61	1.01

This table indicates the benefit return per unit cost to be highest for the latex crusting compounds with $.74 per ton in benefit value resulting from costs of $.14 per ton. For rail car applications, the latex crust appears to be most cost-effective.

CONCLUSIONS

Based upon the results of this technical effort, the following conclusions are presented:

1. Commercially available latex resins used as coal crusting compounds can prevent dust loss and wind erosion, but do not waterproof the coal surface.

2. Hot-melt formulations consisting of about 77.5% coal, 15% slack wax, 3.75% polyethylene and 3.75% polypropylene are capable of sealing a coal surface against water penetration.

3. A filled latex formulation consisting of 50% coal, 23% Everflex GT resin and 27% water is capable of sealing a coal surface against water penetration provided the surface is coated with fines of less than about 1/8" particle size.

4. Hot-melt coatings appear to be most cost effective for application to coal stockpiles.

5. Latex crusting compounds appear to be the most cost effective means of protecting coal in rail cars.

6. The anticipated return in dollars and intangible benefits by use of the coal protection methods described herein is in the range of 10-15 times the cost of such protection.

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NEW CONCEPTS FOR CONTROL OF FUGITIVE DUST

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ABSTRACT

NEW CONCEPTS FOR CONTROL OF FUGITIVE DUST

by

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In the vast majority (92%) of air quality control regions not meeting total suspended particulate (TSP) standards, fugitive sources exceed point sources. In rural areas, unpaved roads and agricultural activities are important sources; in urban areas, paved roads and construction sites. In industrial areas, both process and open sources of fugitive particles must be controlled not only for TSP but also for lead air quality standards. Hence, the need for control of fugitive dust requires technology to span a variety of source types and control requirements.

Current technology involves improvement in gathering fugitive process emissions and prevention of open source fugitive emissions. One new concept under study by EPA does neither of these but rather attempts to capture particles without hooding and evacuation to a control device. Instead the particles are caused to settle out by introduction of another aerosol with appropriate charge and chemical properties. Another new concept under study uses a different approach for prevention of emissions from unpaved roads. Rather than using wetting, oiling, or chemical treatment, the nature of the roadbed is modified. Testing of these concepts is in an early stage but preliminary results show 50 to 93% control is to be expected.

NEW CONCEPTS FOR CONTROL OF FUGITIVE DUST

Introduction

Among the standards used by the Environmental Protection Agency (EPA) to prevent adverse health and ecological impacts of air pollution, is the National Ambient Air Quality Standard (NAAQS). The NAAQS is currently expressed in a number of ways involving the limitation of total suspended particulate (TSP). Unfortunately, the TSP standard is not being met in many air quality control regions. Fugitive dusts account for most of this problem. In more than nine out of ten control regions, fugitive emissions are greater than ducted point source emissions. In one out of three control regions, fugitive emissions are more than 10 times greater than ducted point source emissions.[1] Within cities the contribution of point sources is expected to be smaller than fugitive emissions. In Philadelphia, more than one-third of the TSP was attributed to reentrained street dust and vehicular traffic while point sources were responsible for less than one-fourth of the TSP.[2]

Because of the importance of fugitive sources in meeting NAAQS, EPA is involved in a program of control technology research and development. Re-entrained dust emissions and fugitive process emissions, both described below, are two ongoing projects which have been labeled "new concepts." This term does not imply that such technology has not been previously investigated but rather that the technology has not been fully utilized and will remain new until it is accepted into conventional practice.

Re-entrained Dust Emissions

Fugitive emissions come from uncontained process emissions or from the re-entrainment of dust. For the latter, the problem can be solved if the surface of the dust can be stabilized and kept free of unstabilized dust. The surface may be stabilized by the application of water, oil, or chemical surface agents. In the case of unpaved roads, application of such materials is less cost effective than paving the roads.[3] Advances in fabric development now provide another possibility to stabilize a surface; especially, unpaved and haul roads. Using this approach, an unpaved road would be covered with fabric and then coarse aggregate.

The fundamental concept behind use of a fabric roadbed stabilizer, or "road carpet," for control of fine particle emissions from unpaved roads is prevention of vortex entrainment by separation of fine roadbed materials from the coarse aggregate where the traffic movement occurs. Large aggregate is held from settling, while newly deposited fines (< 70 μm) are filtered by gravitation and hydraulic action down to a zone away from vortex entrainment. Road carpet can be made from spunbonded, thin-film polypropylene on nylon sheet (Celanese), continuous filament polyester fibers needled to form a highly permeable fabric (Monsanto), or other spun or needle-punched synthetic materials. The mechanical interlocking of fibers makes a formed fabric with the required durability

and toughness. Designed for road construction use, this fabric is laid over poor loadbearing soils to help support and contain the overburden aggregate. It spreads concentrated stress from heavy-wheeled traffic over a wider area, siphons away ground water, and contains fine soil particles in the roadbed that could otherwise contaminate ballast or road overburden.

Capital and operating costs differ for various control options. Although initial costs for some control alternatives may be lower than for road carpet, maintenance costs for these alternatives may be quite high. Road carpets can add as much as 20% to the cost of a road construction, but the material will pay for itself in less than a year in reduced maintenance costs and driver safety. In some cases, a new road can be built for as low as 20% of the cost of conventional roads.

Treating the road surface with oil once a month is an efficient method of controlling unpaved road dust emissions. The estimated cost of such applications (reported in 1972) is $0.10 per square meter treated per year.[4] However, 70% to 75% of the oil applied moves from the surface of the road by runoff and dust transport, resulting in ecological harm caused by the oil or its heavy metal constituents.[4] Furthermore, surface oiling does not prevent potholes, a major part of road maintenance.

Roads constructed using road carpet, on the other hand, require significantly less maintenance because of the increased road stabilization imparted by the carpet. The most effective method of reducing particulate emissions is paving the road surface; but, owing to high initial cost and subsequent maintenance and repair costs, paving these temporary roads is impractical. Road carpet can be installed to give a virtually permanent, maintenance-free road.

Use of road carpet fabric results in no health or safety hazards or any other unfavorable environmental impact. In the development of these fabrics, various synthetic polymers (including nylon, polypropylene, and polyester) were evaluated. Fabrics made from any of these products generally are resistant to mildew, mild acids, and alkali, and are rot and vermin proof. Polyester was chosen by Monsanto because of the following distinct advantages:

- resistance to chemicals, including those found in soils
- constant properties over a wide range of temperatures
- high melting point
- little change in wet or dry properties
- low moisture absorption
- high abrasion resistance
- high modulus of elasticity and excellent resilience
- excellent creep resistance

Thus, use of road carpet precludes any environmental damage due to leaching of hazardous chemicals or heavy metals.

Fugitive Process Emissions (FPE)

Control of fugitive process emissions often has to overcome the disadvantages of a dispersed, low concentration emission in a large volume. If the particulate were not dispersed it could be collected in a high efficiency hood and ducted to a conventional control device. If the particulate were of a high concentration or in a building of small volume, the process building could be evacuated to a conventional control device. Assuming that the FPE cannot be hooded or evacuated to a control device, EPA is investigating the use of chemically and electrostatically treated water spray droplets. This method applies to fugitive dust emissions which cannot be controlled by stabilizing the surface of the source. Examples are transfer points, conveyor belts, charging operations, molding lines, and roof monitors. As always, water with or without chemical additives can be sprayed to stabilize exposed sources, such as stock piles which are not being processed.

Fugitive dust particles entrained in the gas stream may be collected with charged or uncharged water sprays by mechanisms such as diffusion, inertial impaction, interception, and electrophoresis. The larger size of the water drops would allow easy separation from the gas stream by methods such as gravitational settling or entrainment separators.

Figure 1 is a functional diagram for the process anticipated for controlling fugitive emissions. The functional phenomena represented in this diagram could occur concurrently or separately in several types of equipment.

Several types of chemicals have been found effective in reducing fugitive dust emissions when applied to the source. Over 100 chemical products are presently marketed or under development specifically as dust control agents. These chemicals act by several different means and are generally categorized by their composition: bituminous, polymer, resin, enzymatic, emulsion, surface-active agent, ligninsulfonate, latex, etc. Dust stabilizing agents have been reported in the literature[5] and are not being considered in this investigation.

In addition to the use of surface active agents, the concept of using electrostatic sprays involves the controlled disposal of fugitive dust that is entrained in a gas stream. This approach is the most permanent way to control FPE because it precludes the re-dispersal of dust which can occur if stabilization is used or if the collected particles are deposited on the ground. After the air containing the FPE is conveyed to the apparatus, it is necessary to bring the spray into contact with the fugitive dust. The water sprays would be produced with single- or two-fluid nozzles. Nozzle systems are currently available on the market which produce charged water drops. Contacting may be obtained by projecting the water drops into the dusty gas or moving the dusty gas into the vicinity of the water drops. The water drops will be formed by hydraulic pressure sprays. An ejector type of spray system is a likely candidate for contacting with minimum energy usage.

Containing fugitive emissions with a series of barriers and/or electric curtains may prove most cost effective. The electric curtains would be used to deflect the particulate, thus minimizing the volume of air to be cleaned. Because of the great variety of FPE source situations, it is not possible to use a single approach to this part of the system.

After spraying in the charged water drops, the next process step would be to remove the water spray drops after sufficient contacting time to effect capture of the initial fine particles present in the gas. At this stage the large size and mass of the water drops is utilized to separate them from the gas. An entrainment separator may be used for this step, depending on the mist elimination and pressure drop requirements. The cleaned gas stream leaves the entrainment separator at this point. The water from the entrainment separator is passed through a separation process, such as a filter, to remove the collected dust particles. The water may then be recycled and the dust may be disposed of in such a way as to prevent its re-dispersion.

The concept proposed here has some similarity to liquid scrubbers. Recent studies have shown increased scrubber efficiency can be obtained by electrostatically charging particle and collectors to opposite polarity.[6,7] Hoenig[8] has shown that naturally charged fugitive emissions may be collected more efficiently with charged drops than with uncharged drops. The results of preliminary analysis show that this technique is most effective for small particles (less than 5 μm diameter).

Development of Concepts

EPA has planned an extensive research, test, and evaluation program for both of these concepts. The research portion will identify parameters affecting the theoretical and economic limitations to utilization of the technology. Preliminary studies of these parameters will provide an understanding for optimization. In the case of road carpet, the vortex entrainment, comminution, and saltation of material on the coarse aggregate must be modeled. Size and thickness of the aggregate may affect performance. For the charged water sprays, methods of partial containment of the FPE must be studied as well as fundamentals of droplet/particle interactions.

The test and evaluation portion of the program will involve studying a prototype of each new concept. Using the road carpet, a 30 to 40 meter long section of haul road will be constructed at a quarry. A second prototype road will be constructed at a different site to evaluate subsoil differences.

The emissions from the prototype road and a comparison conventional road will be monitored at several intervals of time. Sampling will be conducted when wind conditions are favorable at two different vehicle speeds and over the course of 3 months. During this time, any changes in the mass reduction or particle size distribution will be noted and correlated with observations on the condition of the prototype road. The durability and other positive attributes of this type of road construction will be evaluated in order to reassess the economics of this method of

emissions reduction. Sampling will be conducted using the quick-reaction sampler at distances of from 10 to 12 meters from the road. Since the sampling can be conducted over short periods of time (4 to 10 minutes), desirable wind direction, speed, and atmospheric stability can be chosen for sampling (i.e., mid-day and approximately perpendicular to the road).

The test and evaluation program for the charged water sprays will consider the following parameters:

1. Air flow rate--three levels.
2. Air temperature--two levels.
3. Particle type--two levels.
4. Particle size distribution--two levels.
5. Water-to-air ratio.
6. Water pressure.
7. Nozzle type and arrangement.
8. Electrostatic charge level.
9. Surfactant concentration.
10. Confinement type.
11. Wind conditions (windy and calm).

The spray contactor will be 1.8 m high and 2.5 m wide and attempt to collect metal oxide or liquid aerosols. Sampling will be by total filters, cascade impactors, and light scattering devices.

Conclusions

Two concepts for control of fugitive emissions have been identified and will be investigated under an EPA program. One concept employs road carpet to stabilize unpaved roads by separating traffic from entrainable particles. The second employs charged water sprays for direct collection of fugitive process emissions.

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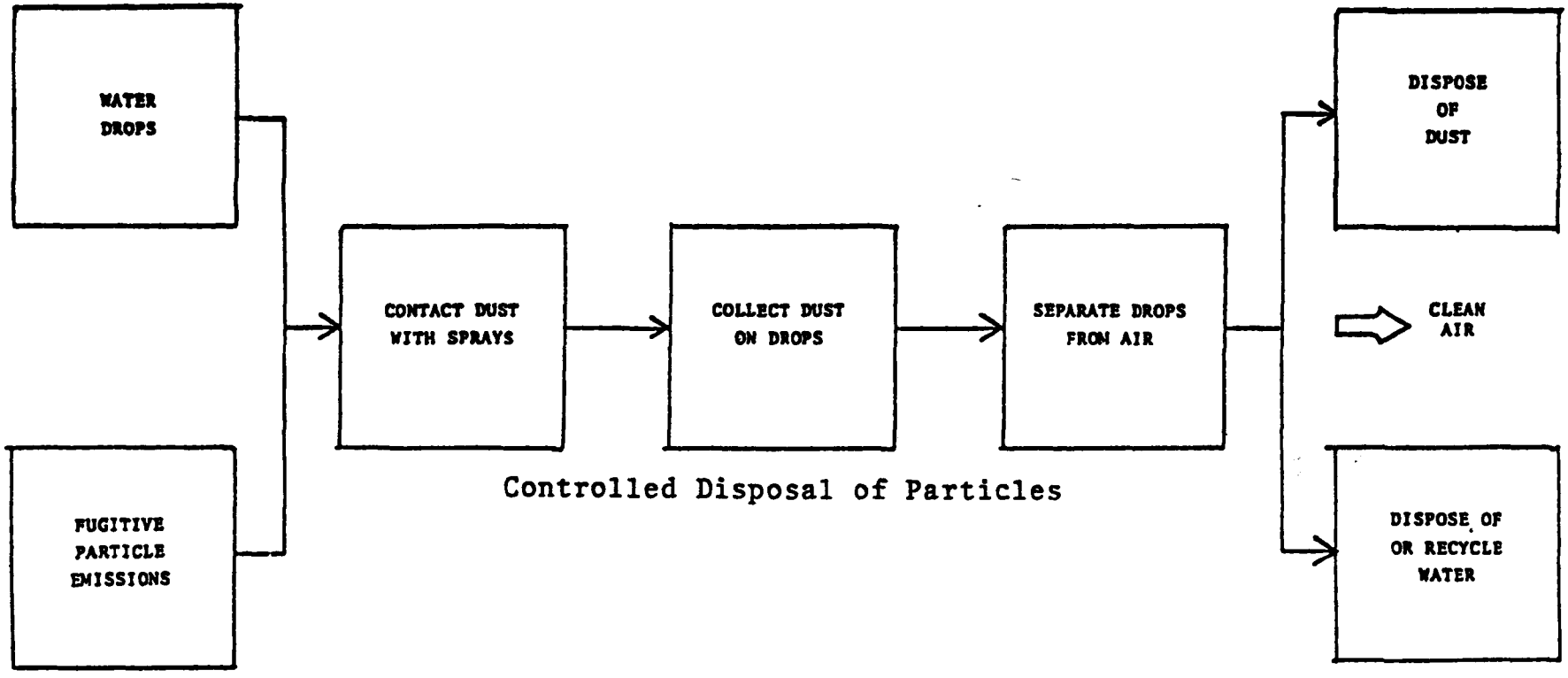

Figure 1. Functional diagram for the major process steps involved in controlling fugitive particle emissions with a charged drop system.

BEST AVAILABLE CONTROL TECHNOLOGY (BACT) FOR FUGITIVE EMISSIONS CONTROL IN THE STEEL INDUSTRY

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ABSTRACT

Technology for controlling fugitive emissions related to the various steps in iron and steelmaking have been given low priorities in the past due to light concentrations as well as the difficulty in collecting these emissions with the result that the state-of-the-art for controlling fugitive emissions in the steel industry has lagged behind the technology developed in other areas. In some cases, such as blast furnace cast house emission control, this technology is virtually non-existent in the U.S.

Since the technology for emission reduction through gas cleaning already exists and can be accomplished by any number of air pollution control devices, the limited scope of this paper will concentrate on the best available technology for collecting fugitive emissions from the major sources which include:

1) Blast furnace - cast house
2) Oxygen steelmaking - hot metal charging, tapping ladle alloy additions, reladling
3) Electric Furnace Shop - charging and tapping

The ideal solution appears to be collection of the fugitive emissions at their source preventing their escape into the atmosphere, which allows emission control with minimum volumes at lowest capital investment and operating cost.

Although collecting the emissions at their source has not always been the most practical method in the past, due to interference with normal operations, present day technology is gradually overcoming these obstacles, allowing systems that collect the emissions at their source to be installed in operating shops.

BEST AVAILABLE CONTROL TECHNOLOGY (BACT) FOR FUGITIVE EMISSIONS CONTROL IN THE STEEL INDUSTRY

INTRODUCTION

BLAST FURNACE CAST HOUSE EMISSION CONTROL

Tap Hole
Trough
Runners
Hot Metal Cars
Slag Pots

FUGITIVE EMISSION CONTROL DURING THE OXYGEN STEELMAKING PROCESS

Hot Metal Charging
Scrap Charging
Tapping
Slagging
Puffing During Oxygen Blow

HOT METAL RELADLING

FLUX HANDLING

ELECTRIC FURNACE SHOP FUGITIVE EMISSION CONTROL

Charging
Tapping

CONCLUSION

."BEST AVAILABLE CONTROL TECHNOLOGY (BACT)
FOR FUGITIVE EMISSIONS CONTROL IN THE STEEL INDUSTRY"

Introduction

Current air pollution control regulations dictate that the Best Available Control Technology (BACT) be utilized for controlling fugitive emissions in the Iron and Steel Industry. These emissions can be generally divided into two (2) categories - open dust sources and process fugitive emissions. Open dust source fugitive emissions include those sources from which emissions are generated by the forces of wind and machinery acting on exposed aggregate materials, while process fugitive emissions include uncaptured particulates and gases that are related to the various steps in the iron and steelmaking process.

Since air pollution is associated with practically all steps of iron and steel production, the related air pollution control equipment is an important factor in all of these operations. A production process may become obsolete if it is not capable of meeting today's stringent air pollution control requirements.

Technology for controlling fugitive emissions related to the various steps in iron and steelmaking have been given low priorities in the past due to light concentrations, as well as the difficulty in collecting these emissions with the result that the state-of-the-art for controlling fugitive emissions in the steel industry has lagged behind the technology developed in other areas. In some cases, such as blast furnace cast house emission control, the technology is virtually non-existent in the United States.

Since the technology for emission reduction through gas cleaning already exists and can be accomplished by any number of air pollution control devices, the limited scope of this paper will concentrate on the Best Available Control Technology (BACT) for collecting the process fugitive emissions generated by the main metallurgical production processes which include Blast Furnaces, Basic Oxygen Steelmaking and Electric Furnaces.

The alternatives for collecting fugitive emissions generated during the main metallurgical processes are generally limited and usually have many disadvantages.

The ideal solution of collecting fugitive emissions at their source to prevent their escape into the atmosphere, allowing emission control with minimum volumes at lowest capital investment and operating cost, is normally the most difficult system to install.

Collecting fugitive emissions at their source has not always been the most practical method in the past due to possible interference with normal operations; present day technology is gradually overcoming these obstacles, allowing systems that collect the emissions at their source to be installed in operating shops.

BLAST FURNACE CAST HOUSE EMISSION CONTROL

The cast house structure surrounding the blast furnace, encloses the runners and operating area and provides weather protection for the operators and equipment (Figure 1). At the same time, this enclosure also contains the fumes generated during the cast. Since environmental control efforts have been concentrated in other areas, blast furnace cast house emission control has been given a low priority with the result that little has been done to date to control these cast house emissions in the U. S.

In the U. S. today, there are 184 blast furnaces producing basic iron with 3 new blast furnaces scheduled to start up in the near future. However, none of the operating blast furnaces are capable of meeting current air pollution control regulations. It is anticipated that in the future, all new blast furnaces will be required to incorporate cast house emission control and existing blast furnace cast houses will be required to install some degree of cast house emission control.

Virtually all blast furnace cast house emission control technology to date has been developed by the Japanese. During the past ten (10) years, they have developed their systems to the point where they now have integrated their ironmaking and emission control. At the present time, one-hundred percent (100%) of the blast furnaces in Japan have some degree of cast house emission control.

Primarily, the Japanese approach is to capture the fumes at their source, preventing their escape into the atmosphere. This is accomplished by close fitting hoods and covers wherever the hot metal is exposed to the atmosphere in the cast house. At the same time, the Japanese often employ a separate secondary system consisting of canopy or monitor hoods for scavenging and building evacuation.

Particulate emissions from cast houses in the U. S. have been found to be in the range of 0.2 to 0.6 lbs./ton of hot metal cast. However, in Japan, particulate emissions have been found to exceed 1.0 lbs./ton of hot metal cast. In general, these emissions are approximately 75% iron oxide with small percentages of manganese, silicon oxides and sulphites. Differences in the levels of fume generated from cast house to cast house can be attributed to the variations in operating practices and materials used in the blast furnace cast house.

Sources of fugitive emissions in the blast furnace cast house are the tap hole, trough, runners, hot metal cars and slag pots. These areas generate practically all of the fugitive emissions associated with cast house operation. Existing cast house installations in the U. S. offer many problems in adopting Japanese technology. However, the new blast furnaces with multiple tap holes scheduled to start up in the U. S. in the near future are incorporating Japanese technology for cast house emission control (Figure 2).

Tap Hole

The primary source of fugitive emissions in the cast house is from the hot metal as it exits the blast furnace at the tap hole. Approximately 30% to 40% of the total fugitive emissions can be attributed to the tap hole. With the Japanese technology, these fumes are normally controlled with local hoods or retractable curtains (Figure 3).

Trough

The hot metal pool adjacent to the tap hole normally extending to the dam and skimmer (25' to 50' long x 3' to 4' wide x 2' deep) serving as a holding pit to separate the hot metal and slag, is also a major source of emissions in the cast house. The fumes here are effected by the area of hot metal exposed to the atmosphere, temperature of the hot metal, type of refractory used, and other factors. Operating systems collect these emissions with the tap hole fumes utilizing a retractable curtain.

Runners

Emissions from the runners are also dependent on the pool areas exposed to the atmosphere and the metal temperature. As the metal cools, carbon emerges from the saturated solution as "Kish", a form of graphitic carbon that is light and flaky. These emissions can be controlled by replaceable refractory lined runner covers.

Hot Metal Cars

Molten metal from the runner spouts is poured into the hot metal cars located outside of the cast house or in arcade under the cast house floor (Figure 4). The density of these fumes is dependent on the rate of cooling of the hot metal. Local hooding can normally be utilized to control these fumes since the hot metal cars are always in the same relative position during this operation.

Slag Pots

Fumes generated here are the result of running the slag into refractory lined slag pots or a pit adjacent to the cast house and can be readily controlled with local hoods.

Since the installation of cast house emission control systems must have a minimal impact on established operating practices and at the same time maintain high safety standards on the cast house floor, transfer of Japanese technology for blast furnace cast house emission control in the U.S. is not a simple matter due to the difference in operating practice as well as cast house geometry. Tap hole restrictions, number of pouring stations, runner layout and pouring station geometry are all limiting factors on the majority of older blast furnaces with rated capacities of 2,000 tons/day or less.

The alternative of building evacuation requiring 45-80 air changes per hour is a costly solution to the cast house emission control problem.

FUGITIVE EMISSION CONTROL
BASIC OXYGEN STEELMAKING SHOP

In the BOF Shop, a number of sources exist which generate fugitive emissions that are not captured by the main gas cleaning system during the oxygen blowing process. Today, collection of these fugitive emissions has become more of a concern than the primary gas cleaning itself. This concern for fugitive emission control is based on the difficulty of efficient fume collection at the emission source, since once the fume escapes into the building, it is almost impossible to control.

In an effort to comply with current air pollution control regulations, many systems of different design have been installed and are in operation today with varying degrees of success. However, to successfully control fugitive emissions with minimum capital expenditure, they must be collected at the source and not allowed to escape into the building.

Major sources of fugitive emissions in the basic oxygen steelmaking process are scrap charging, hot metal charging, tapping and ladle alloy additions, slagging, puffing during the oxygen blow and flux handling. Other sources such as ladle transporting and teeming also contribute to fugitive emissions, but these are low in volume and dissipate readily. They normally do not contribute to monitor emissions from the BOF Shop. In general, fugitive emissions from the BOF Shop are effected by a number of factors and can be substantial. However, the particulate loading is relatively low.

Hot Metal Charging

The worst condition occurs during charging of hot metal into the furnace which already contains scrap. The hot metal and the effect of the hot metal on the scrap both contribute to emissions during this period (Figure 5). Test results have shown that fumes generated at this time are composed of approximately 35% iron oxide, 30% kish and others, with particulate size less than 100 microns and an approximate emission rate of 0.3 to 0.4 lbs. per ton of hot metal poured.

Since the prime factors controlling fume generation during this period are the condition of scrap and the rate of hot metal poured, operating practice is a big factor in the amount of fumes generated.

Scrap Charging

Scrap charging itself is not a primary source of pollution, however, when the scrap contains a foreign substance such as dirt, paint or oil, it becomes a major pollution source when the hot metal is added. The quantities of pollution generated during scrap charging itself are minor compared to the fumes generated during hot metal charging and can be minimized by scrap selection.

Tapping

A dense fume normally results from the tapping operation itself and with ladle additions such as ferro silicon and ferro manganese, the magnitude of the problem increases. Fume composition here is dependent on the alloys employed but generally would consist of approximately 75% iron oxide with particle size less than 10 microns and an average emission rate of 0.15 to 0.20 lbs. per ton.

Slagging

Emissions from the furnace slagging operation can be a major problem depending on the type of steel being produced since it influences the slag volume and emission rate at this time. The method of killing the slag is also a factor which influences the emissions during the slagging operation. Fumes are generated inside the furnace at this time and continue with the slag as it falls inside the slag pots below the charging floor. These emissions normally have a particle size less than 100 microns.

Slagging fumes are relatively cold and do not have the necessary thermal energy to cause the fumes to rise into an overhead canopy hood making them difficult to capture.

Puffing

Puffing during the oxygen blow occurs intermittently. When this occurs, generally small quantities of fume are emitted around the mouth of the furnace. This is not a major source of fugitive emissions, however, it does contribute to the total emission problem in the shop.

Methods of Collection

With present technology, the alternatives available for collecting the oxygen steelmaking fugitive emissions are:

1. Complete or partial building evacuation.
2. Local hoods and dampers.
3. Furnace enclosures.

Canopy hoods located in the building trusses for partial or complete building evacuation systems are operating in a number of shops with varying degrees of success. In most cases, this method is normally considered as a last resort due to the high capital expenditure and operating cost involved. However, these systems usually are given favorable consideration by maintenance and operating personnel since they require minimum maintenance and do not restrict operating practices. Volume requirements to achieve the necessary collection efficiency with a canopy hood system, would normally be in excess of 1,000,000 CFM for complete building evacuation.

Local hoods appear to be more effective than canopy hoods in collecting these emissions, since they are in closer proximity to the emission source. However, relatively high volumes are still required to effectively collect fugitive emissions in this manner. Local hoods are also undesirable from the maintenance and operating point of view. Dampers utilized to direct charging emissions into the main gas cleaning hood have also experienced only limited success to date.

Collection of fugitive emissions by means of a furnace enclosure has proven to be the most economical solution, as well as the Best Available Control Technology (BACT) since it allows collection of emissions at the source and prevents their escape into the atmosphere. With properly designed furnace enclosures, it has been demonstrated that it is possible to effectively control scrap charging, hot metal charging, furnace tapping, ladle alloy additions, furnace slagging and puffing emissions with low volumes. At the present time, systems of this type are doing an effective job of fugitive emission control with volumes of approximately 350,000 ACFM.

The BOF furnace enclosure (Figure 6) essentially forms a gas tight seal when the bi-parting doors are closed. Since the furnace enclosure extends below the charging floor, the only openings are for the ladle car. If desired, the ladle car openings can be effectively reduced by air curtains or the addition of a vertical shield on the end of the ladle car, as a means to increase the efficiency of the furnace enclosure during tapping.

The furnace enclosure design is based on sound engineering principles and actual testing, taking into consideration the volume of fumes generated inside the furnace due to the reaction of pouring hot metal into the furnace, as well as the velocity and temperature of the fumes leaving the mouth of the furnace (Figure 7).

During charging, the bi-parting doors are opened while charging scrap or pouring hot metal and the fumes are collected through the secondary hood located inside the enclosure directly above the furnace mouth. For controlling emissions during all other phases of operation, the enclosure doors are closed, while the fumes are evacuated from the enclosure through the main or secondary hood.

With a properly designed furnace enclosure, it is possible to collect secondary emissions generated by the basic oxygen process with approximately 90% efficiency, provided the charging of hot metal into the furnace is done at a controlled rate and the scrap is relatively clean.

One of the more recent BOF installations with effective fugitive emission control facilities (Figure 8) incorporates individual furnace enclosures over two (2) 350 ton vessels, with the main gas cleaning systems being utilized for secondary emission control (Figure 9).

Practically all new BOF/Q-BOP vessels that have been installed in the U. S. in the past seven (7) years have included a partial or full furnace enclosure for fugitive emission control. Since the original enclosure designs had many deficiencies, these systems are operating today with varying degrees of success. At the present time, however, there are approximately ten (10) installations operating or in the construction stage which incorporate the secondary hood inside the furnace enclosure with sufficient volume for effective fugitive emission control being provided by the main gas cleaning or an auxiliary system.

The first retrofit fugitive emission control system incorporating furnace enclosures for controlling emissions has been installed in an operating BOF Shop without interrupting normal production, demonstrating that it is possible to install such equipment without disrupting normal production schedules. In this case, an existing baghouse system was utilized for gas cleaning.

Hot Metal Reladling

The hot metal reladling operation is another primary source of fugitive emissions in the oxygen steelmaking process. The fumes generated during hot metal reladling consist of approximately 55% iron oxide less than 3 microns, 42% graphite greater than 75 microns and 3% others with an approximate emission rate of 0.25 lbs. per ton of hot metal. With today's technology, collection of reladling emissions is not a problem since it is possible to utilize local hoods or close fitting ladle hoods depending on the arrangement of the reladling facility.

The main factors effecting the collection of reladling emissions is the distance of the hood from the ladle mouth, as well as the rate of pouring hot metal. In actual practice, since the handling time remains almost constant regardless of the amount of hot metal handled, the volume required to control reladling emissions with a canopy or local hood is proportional to the volume of hot metal.

In contrast, utilization of a close fitting refractory lined ladle hood for collecting the reladling emissions allows fume collection at the source. The close fitting refractory lined ladle hood (Figure 10) utilizes the "air seal" principle where the fumes are drawn through a pouring slot in the hood and the gap between the hood and the ladle with sufficient velocity to prevent fumes from escaping into the atmosphere.

Actual efficiency of the ladle hood has been demonstrated by a recent installation (Figure 11) where the reladling emissions for a 350 ton furnace are effectively collected with 125,000 CFM. In this design, the hot metal is poured through a vertical slot in the movable ladle hood which serves two (2) reladling stations. It is estimated that canopy or local hoods would require volumes in excess of 300,000 CFM to effectively collect these emissions.

Flux Handling

Flux handling also contributes to total fugitive emissions in the BOF Shop, but these emissions are minor in nature and easily controlled.

FUGITIVE EMISSION CONTROL
ELECTRIC FURNACE SHOP

Fugitive emissions associated with the electric furnace shop are mainly charging and tapping emissions which are usually heavy and difficult to capture. Today, a typical emission control system for an electric furnace would normally consist of direct evacuation or side draft hoods for the primary gas cleaning with canopy or local hoods for collecting charging and tapping emissions (Figure 12). Total emissions from electric furnaces producing carbon steel are approximately 30 lbs./ton of steel produced with approximately 3 to 5 lbs./ton associated with charging and tapping emissions, while the particulate loading for electric furnaces melting alloy steel is approximately one-half of the emissions generated by carbon steel furnaces.

Charging Emissions

Charging emissions resulting from charging a hot electric arc furnace with scrap are usually heavy and difficult to capture. The intensity of the charging emissions is a function of scrap cleanliness. Scrap containing heavy rust, oil, grease or dirt produces heavy emissions during charging. These emissions are highly carbonaceous consisting primarily of smoke and soot.

Tapping Emissions

Tapping emissions generated while pouring steel from the furnace into the ladle are primarily composed of metallic oxides resulting from contact with the air and from bath agitation. Tapping fumes, like charging emissions, are also difficult to capture (Figure 13).

Methods of Collection

With present technology, the alternatives available for collecting charging and tapping emissions are complete or partial building evacuation and local hoods, in addition to partial or full furnace enclosures.

Today, most electric furnace shops utilize canopy hoods for charging and tapping emission control. Although they offer little operating restrictions, they have the disadvantages of high volume requirements, high capital and operating costs and result in poor working conditions due to the shop atmosphere.

Local hoods have achieved some degree of success for collecting tapping fumes. However, operating practices do not allow the installation of local hoods for collecting charging emissions.

Based on the success in collecting fugitive emissions in the BOF Shop with furnace enclosures, the alternative of collecting Electric Furnace Shop fugitive emissions by means of a furnace enclosure offers a practical solution. Although the furnace enclosure offers an efficient and economical solution to the problem, objections from operating personnel appear to be a big obstacle.

While the furnace enclosure technology applied to electric furnaces is still in its infancy, an electric furnace shop consisting of two (2) 60 ton arc furnaces, utilizing complete furnace enclosures for total emission control, has been operating since 1976 without adversely effecting their operating practices.

The typical furnace enclosure (Figure 14) essentially forms a gas tight enclosure from which the fumes are evacuated.

During the charging operation, the bi-parting doors are opened to allow entry of the scrap bucket (Figure 15). After positioning the scrap bucket, the bi-parting doors are closed and the scrap is charged into the open furnace. The fumes generated at this time are evacuated from the enclosure for gas cleaning.

During the tapping operation, the bi-parting doors are closed while the fumes are evacuated from the enclosure (Figure 16).

In addition to offering the Best Available Control Technology for electric furnace shop emission control, the furnace enclosure offers many other advantages, including effective emission control with low volume requirements, collection of emissions at their source, lowest capital investment and operating costs, improved working conditions and a practical solution for sound control in electric furnace shops.

The Texas Air Control Board recently made an analysis of electric furnace shop emission control systems with total emissions control in the State of Texas since 1972. Utilizing a ventilation index developed by the Texas Air Control Board to grade the various systems, the furnace enclosure had the lowest index number proving to be the most efficient installation.

CONCLUSION

Technology for fugitive emission control has lagged behind the technology developed for main gas cleaning systems in the Iron and Steel Industry because the fugitive emissions were considered minor in comparison to the emissions generated during the actual melting periods, and the fact that regulatory authorities were willing to tolerate these emissions while concentrating on other more important areas. As a result, at the present time, system applications for fugitive emission control have not reached the same degree of reliability.

This gap in technology is gradually closing due to current pressure by the regulatory authorities to control all fugitive emissions. This is evidenced by the evolution of the total furnace enclosure for controlling fugitive emissions from oxygen steelmaking facilities and electric furnace shops and development of the close fitting ladle hood for controlling reladling emissions.

With present day air pollution control regulations, new technology will undoubtedly be developed for blast furnace cast house emission control. Proven Japanese technology with modifications to accommodate U. S. operating practices and cast house geometry appears to be a practical solution. Two (2) new blast furnaces scheduled to start up in the near future in the U. S. will incorporate Japanese technology for cast house emission control.

The Best Available Control Technology (BACT) for fugitive emission control that will be acceptable to the Iron and Steel Industry requires a technology that offers the most effective and economical type of fugitive emission control from the standpoint of working environment which will allow these emissions to be collected at their source and prevent their escape into the atmosphere.

References

1. Blast Furnace Cast House Emission Control Technology Assessment, EPA-600/2-77-231 (Nov. 1977).

2. Inspection Manual For Enforcement Of New Source Standards For Producing Electric Arc Furnaces, EPA 340/1-77-007 (May 1977).

3. A Regulatory View Of Air Pollution Controls On Electric Arc Furnaces In Foundries And Steel Mills In Texas by James C. Caraway, Texas Air Control Board, Austin, Texas.

Typical open cast house with crane parallel to iron trough

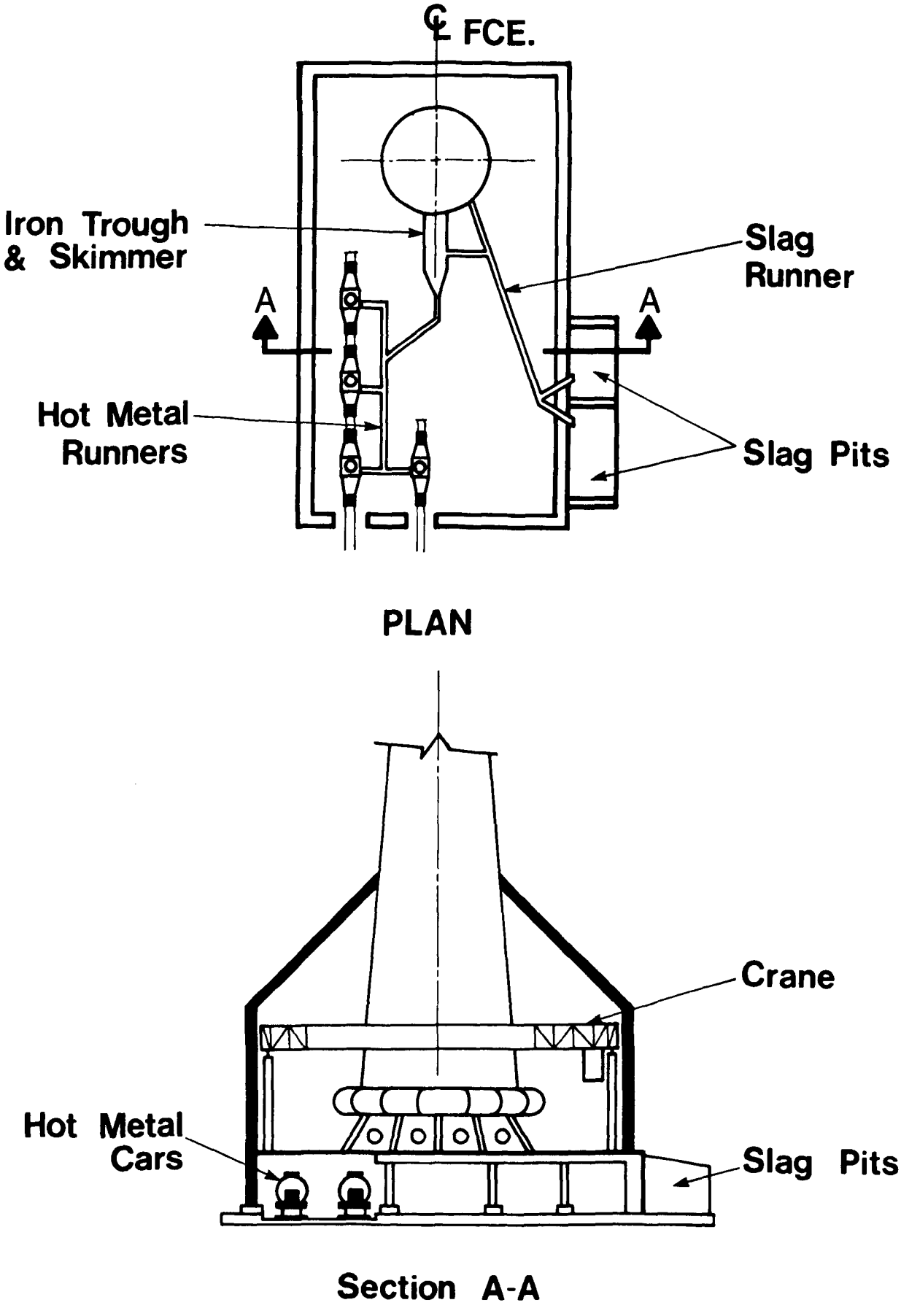
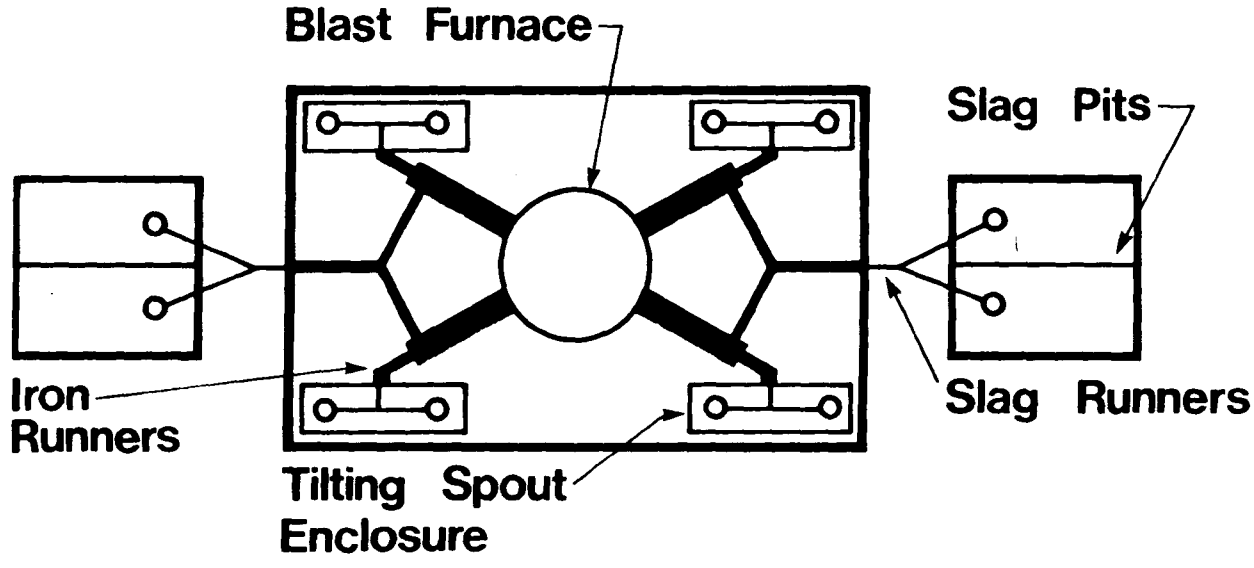
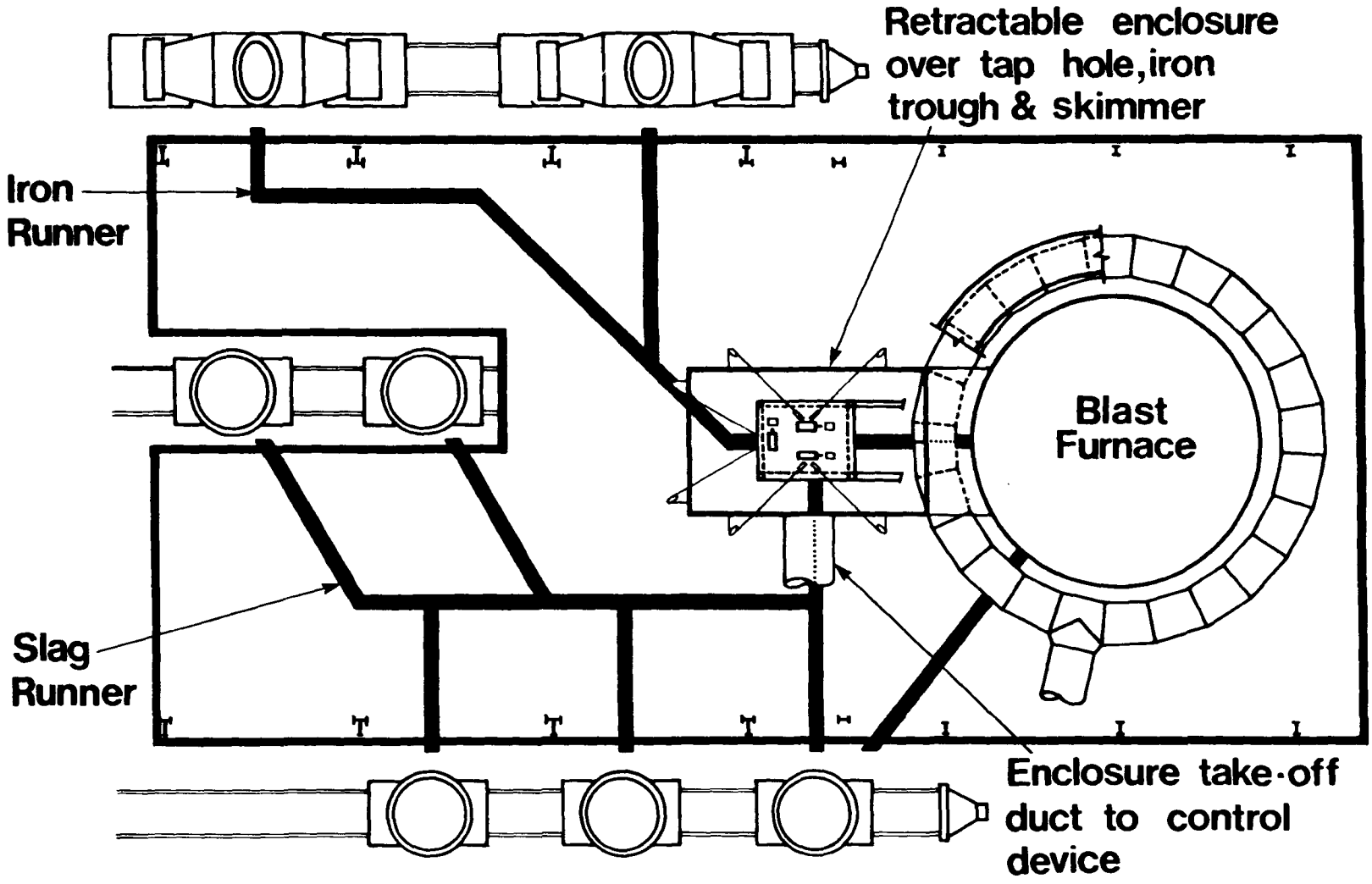


Figure 1 - Cast House Arrangement



Typical cast house 10,000 ton day furnace capacity

Figure 2 - Multiple Tap Hole Cast House Arrangement



Plan view single tap hole furnace partial emission control concept

Figure 3 - Retractable Curtains in the Cast House

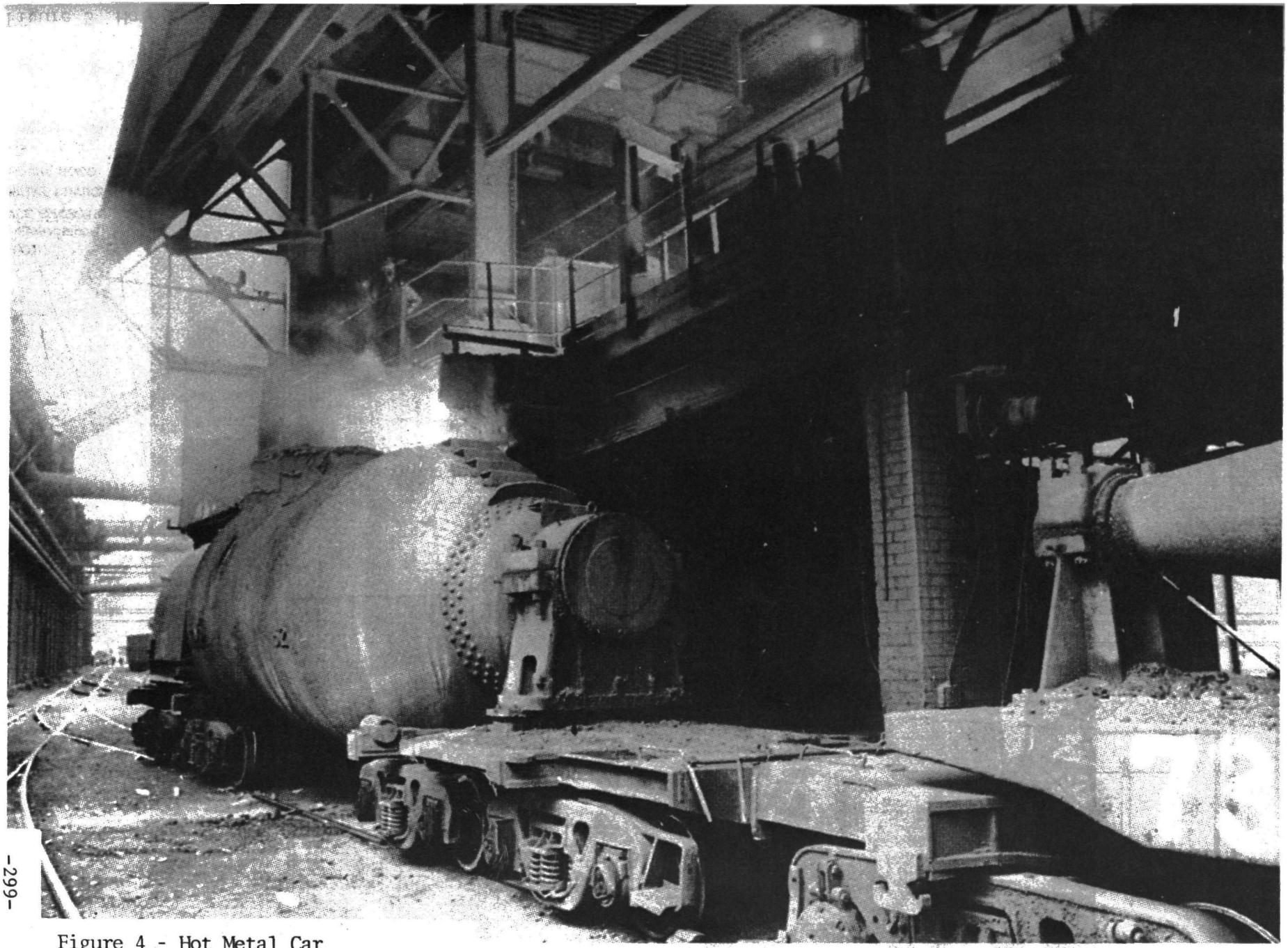


Figure 4 - Hot Metal Car

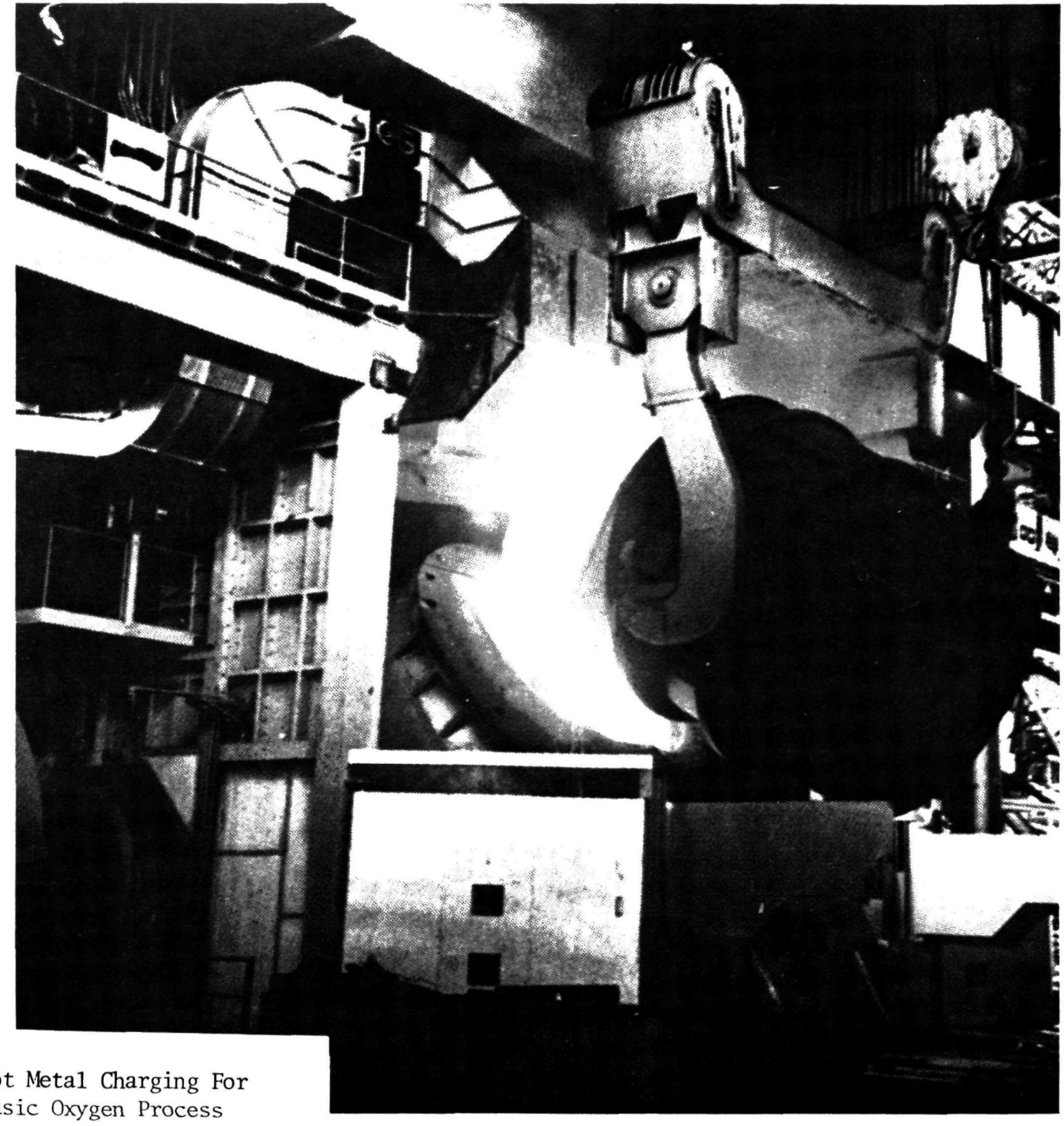


Figure 5 Hot Metal Charging For
Basic Oxygen Process

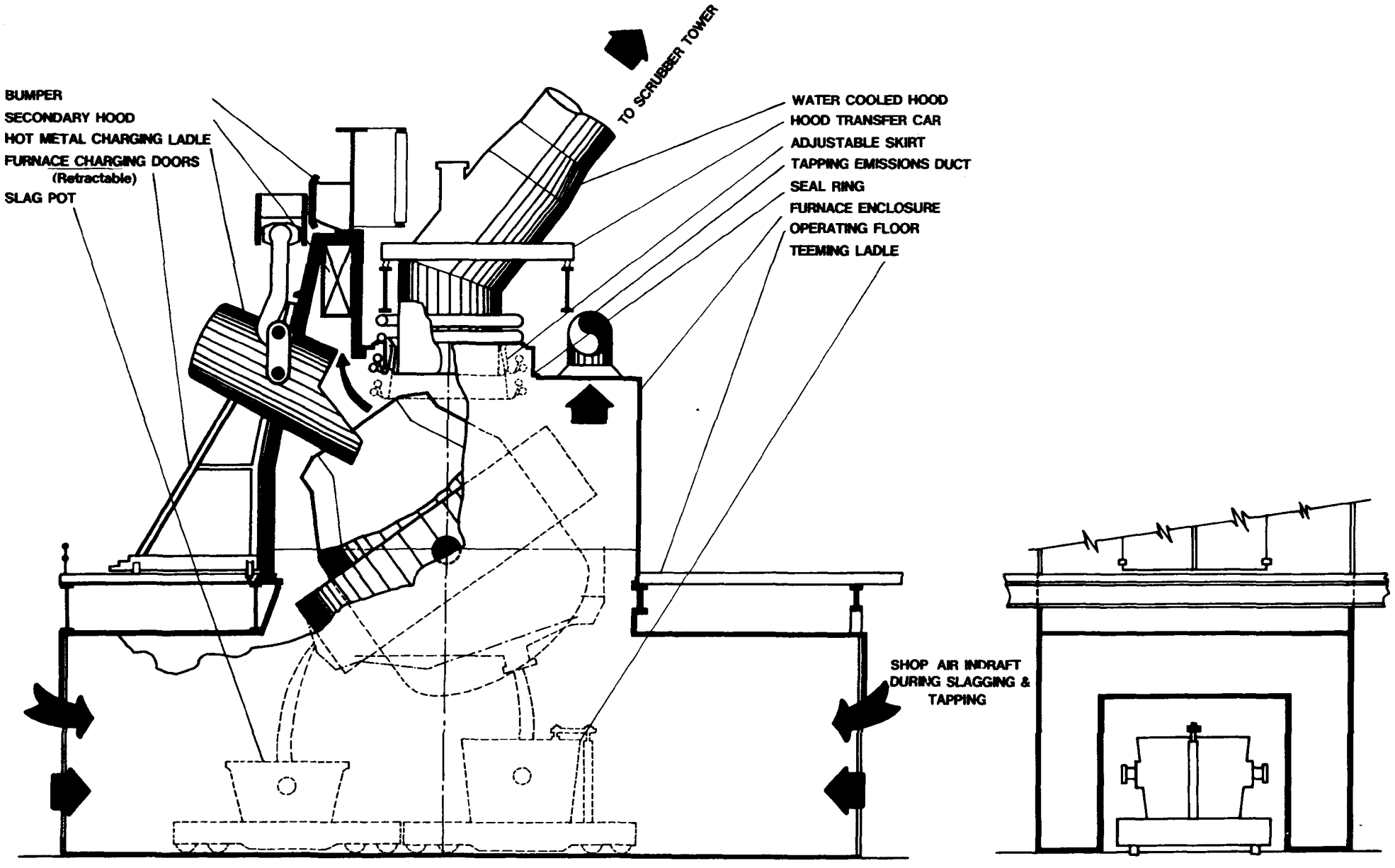
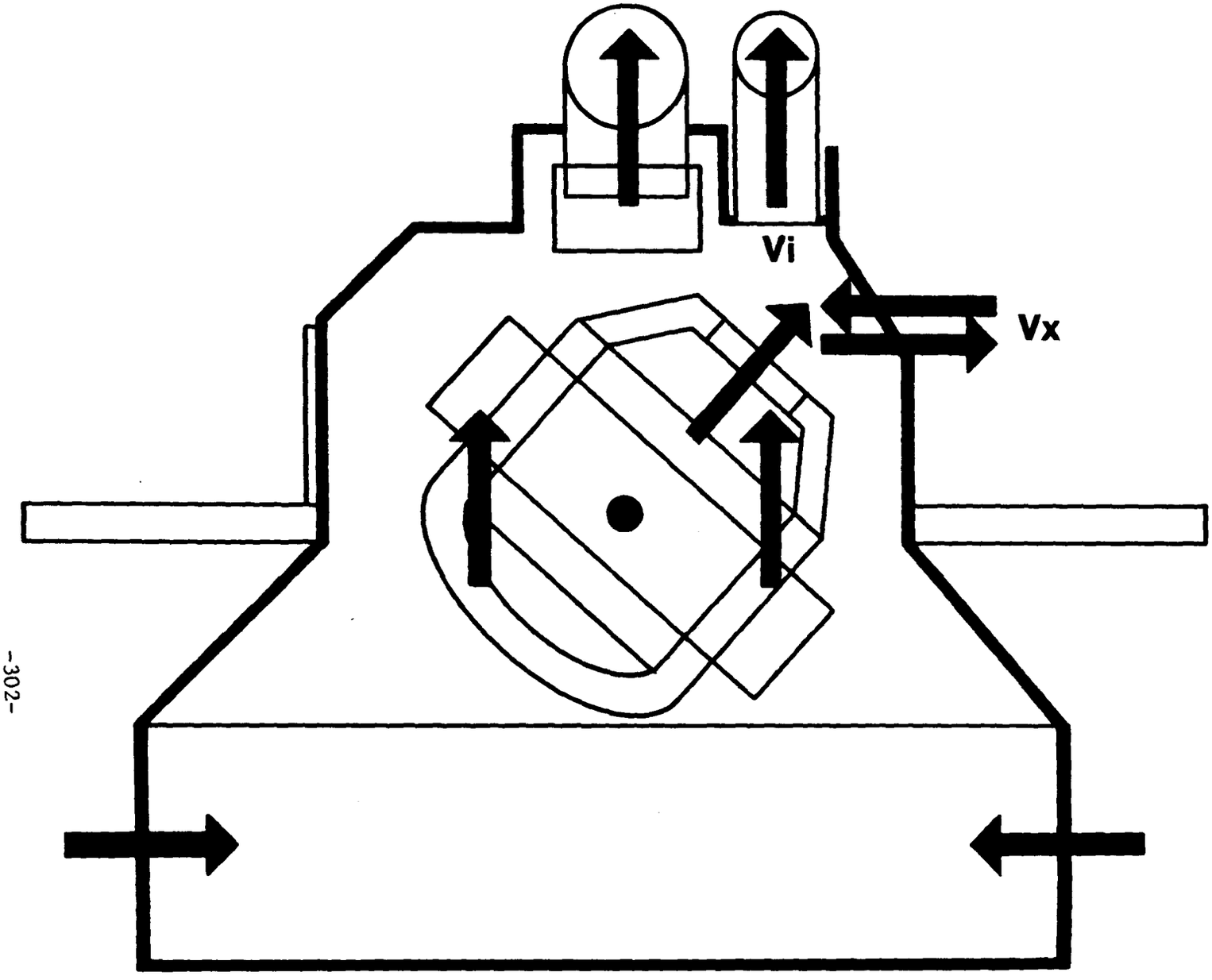


Figure 6 - Schematic Arrangement For BOF Furnace Enclosure



FURNACE ENCLOSURE

Figure 7 - Components Effecting Fugitive Emission Control With Furnace Enclosure

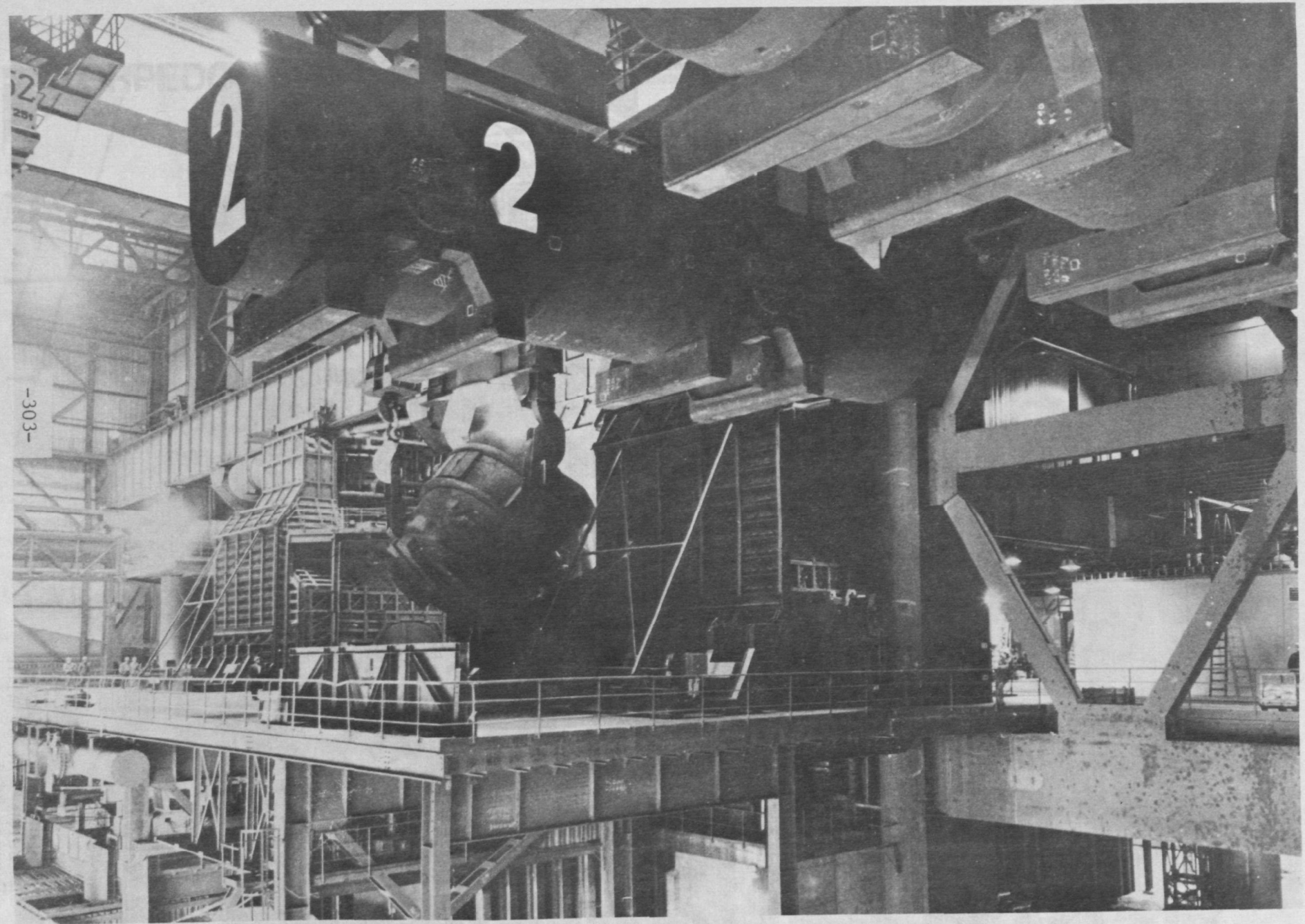


Figure 8 - Recent BOF Installation With Fugitive Emission Control

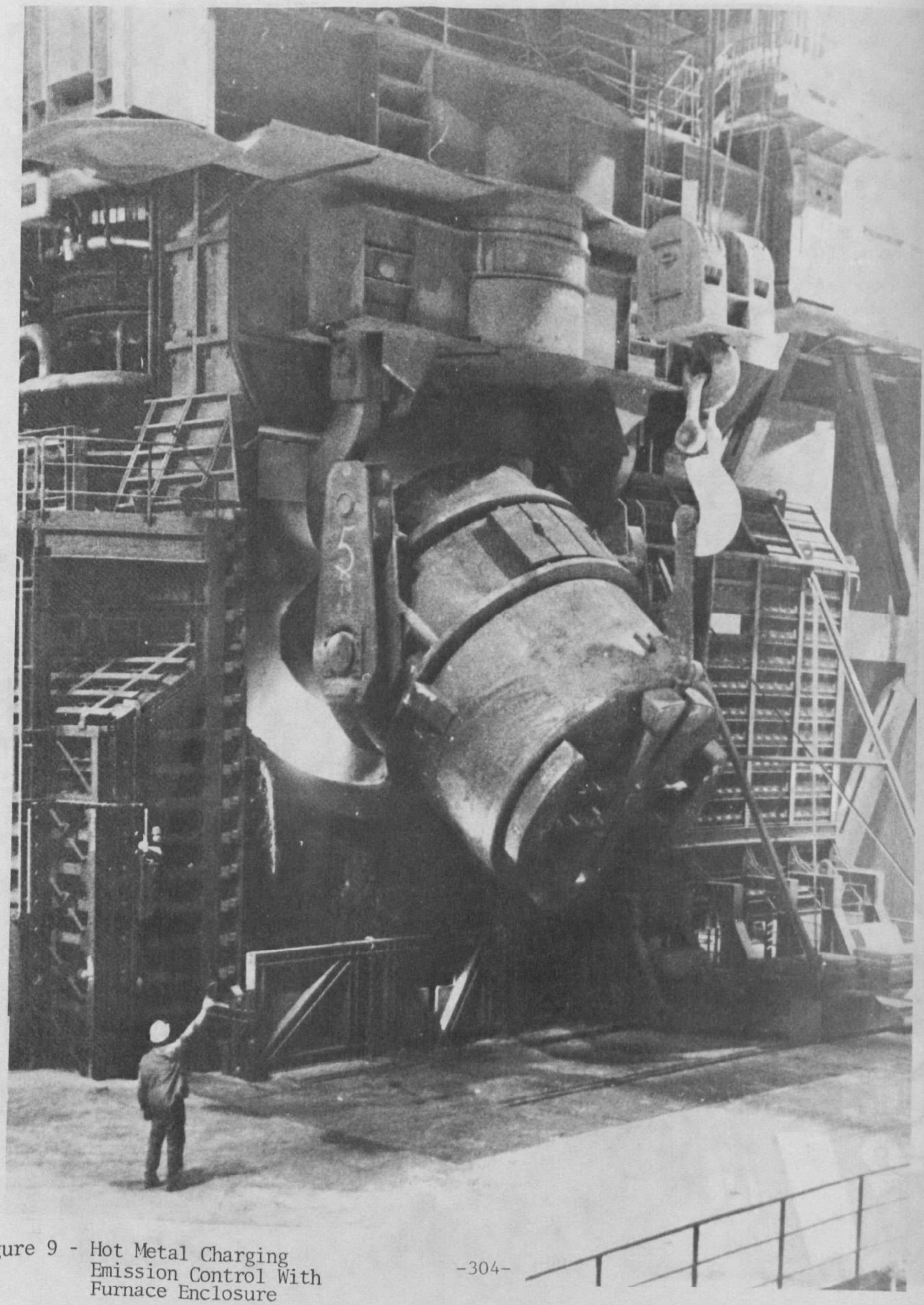


Figure 9 - Hot Metal Charging Emission Control With Furnace Enclosure

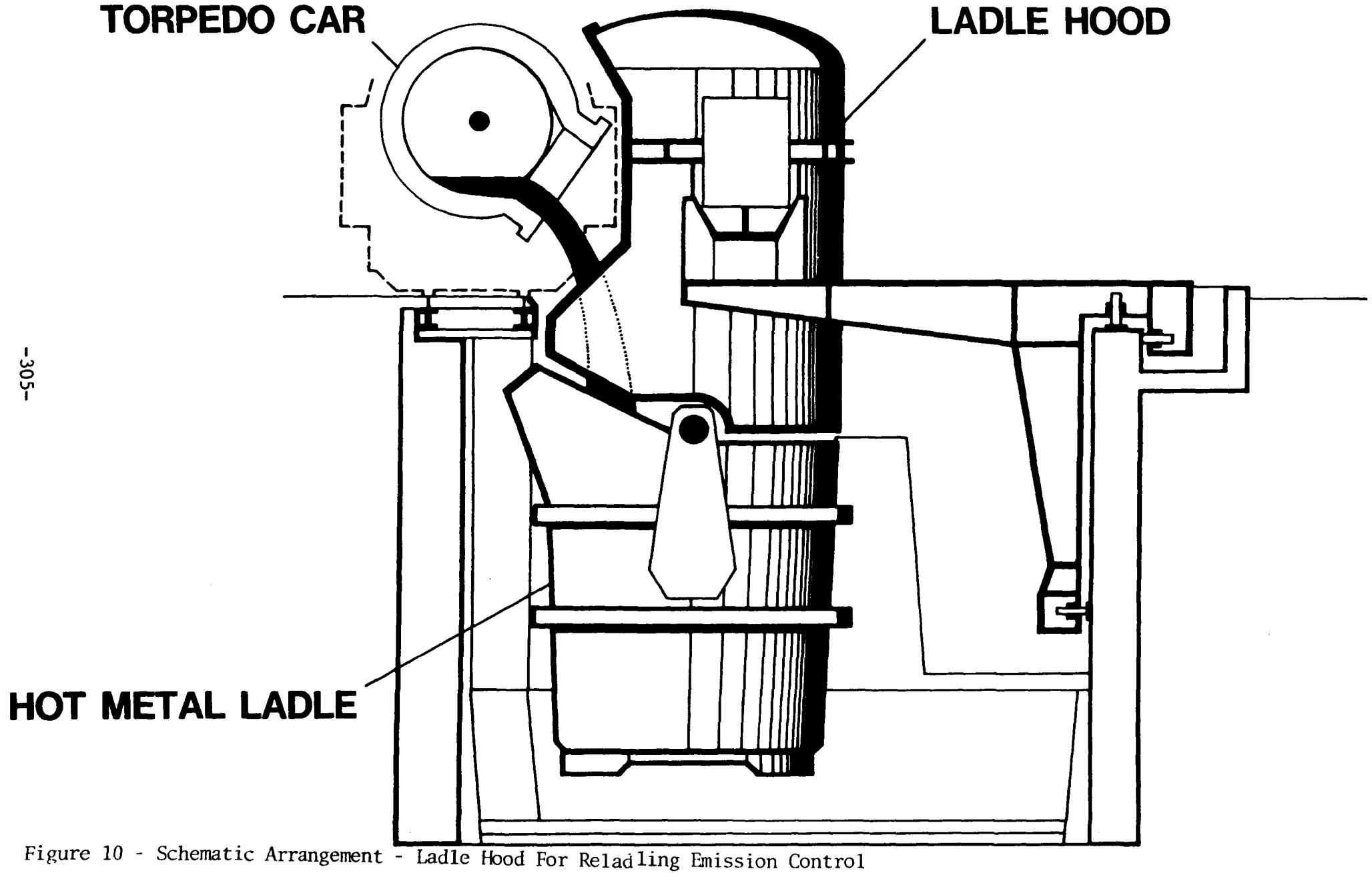


Figure 10 - Schematic Arrangement - Ladle Hood For Reladling Emission Control

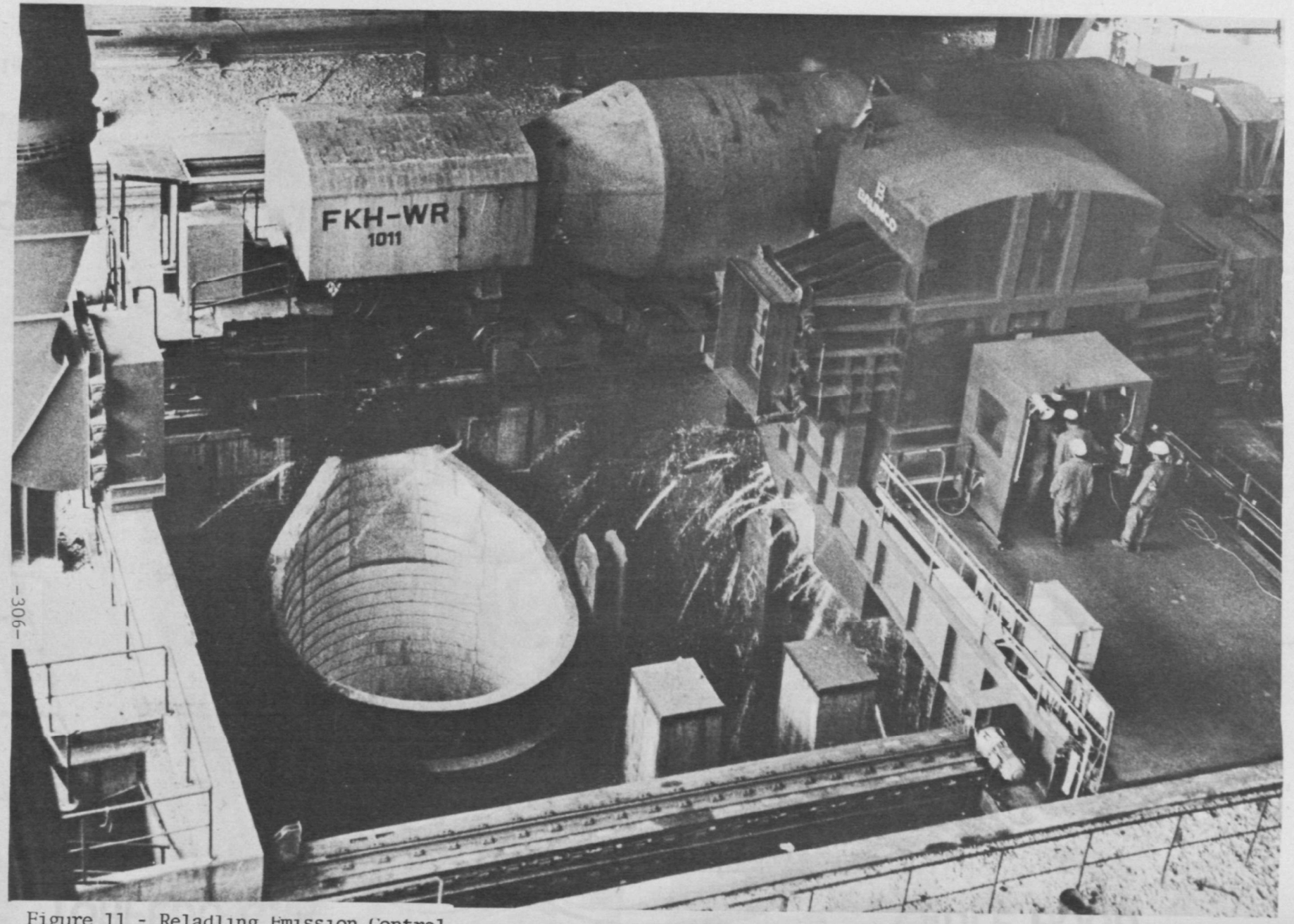


Figure 11 - Reladling Emission Control Pouring Through A Vertical Slot In The Ladle Hood

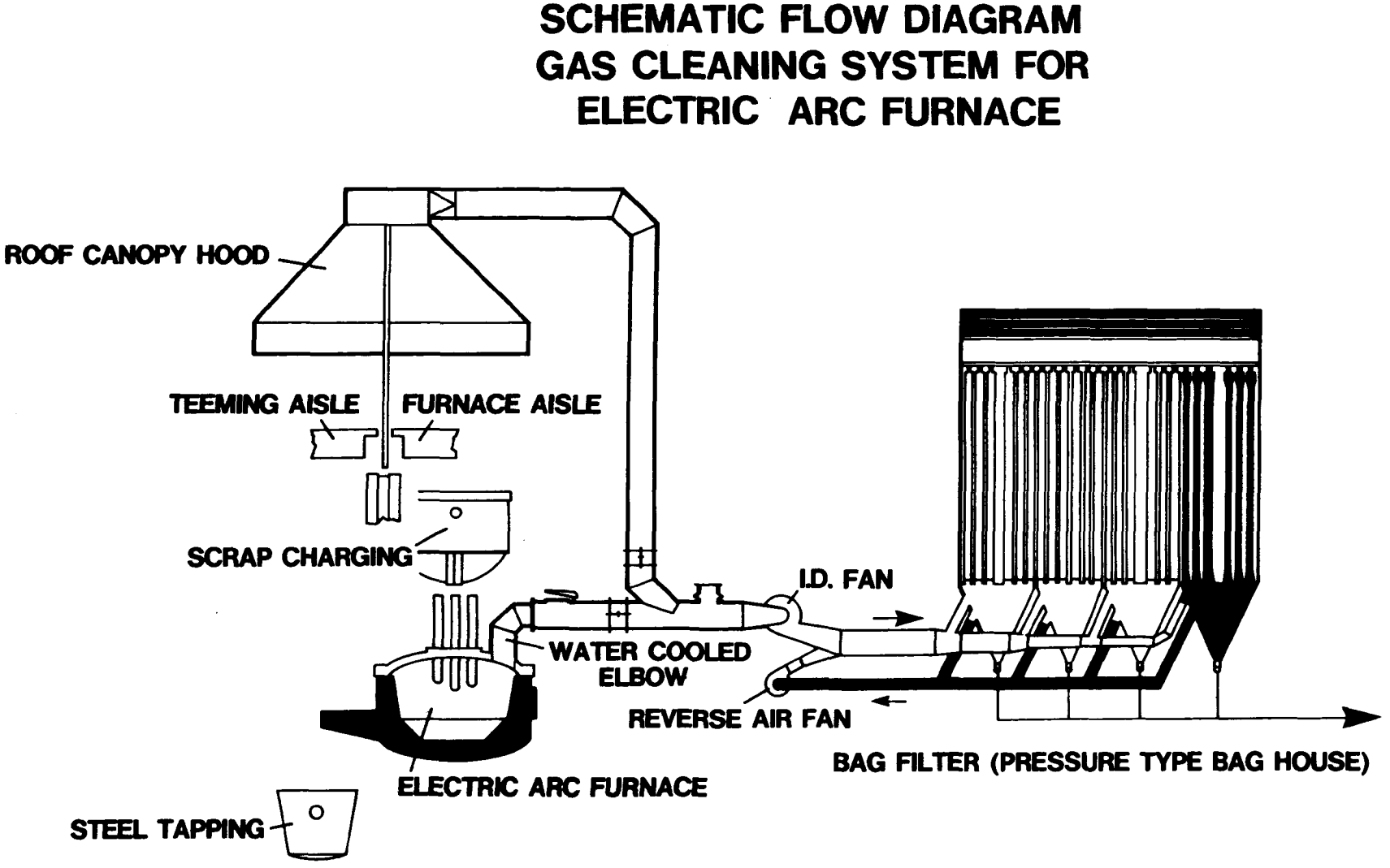


Figure 12 - Schematic Flow Diagram - Pollution Control System For Electric Furnace Shop

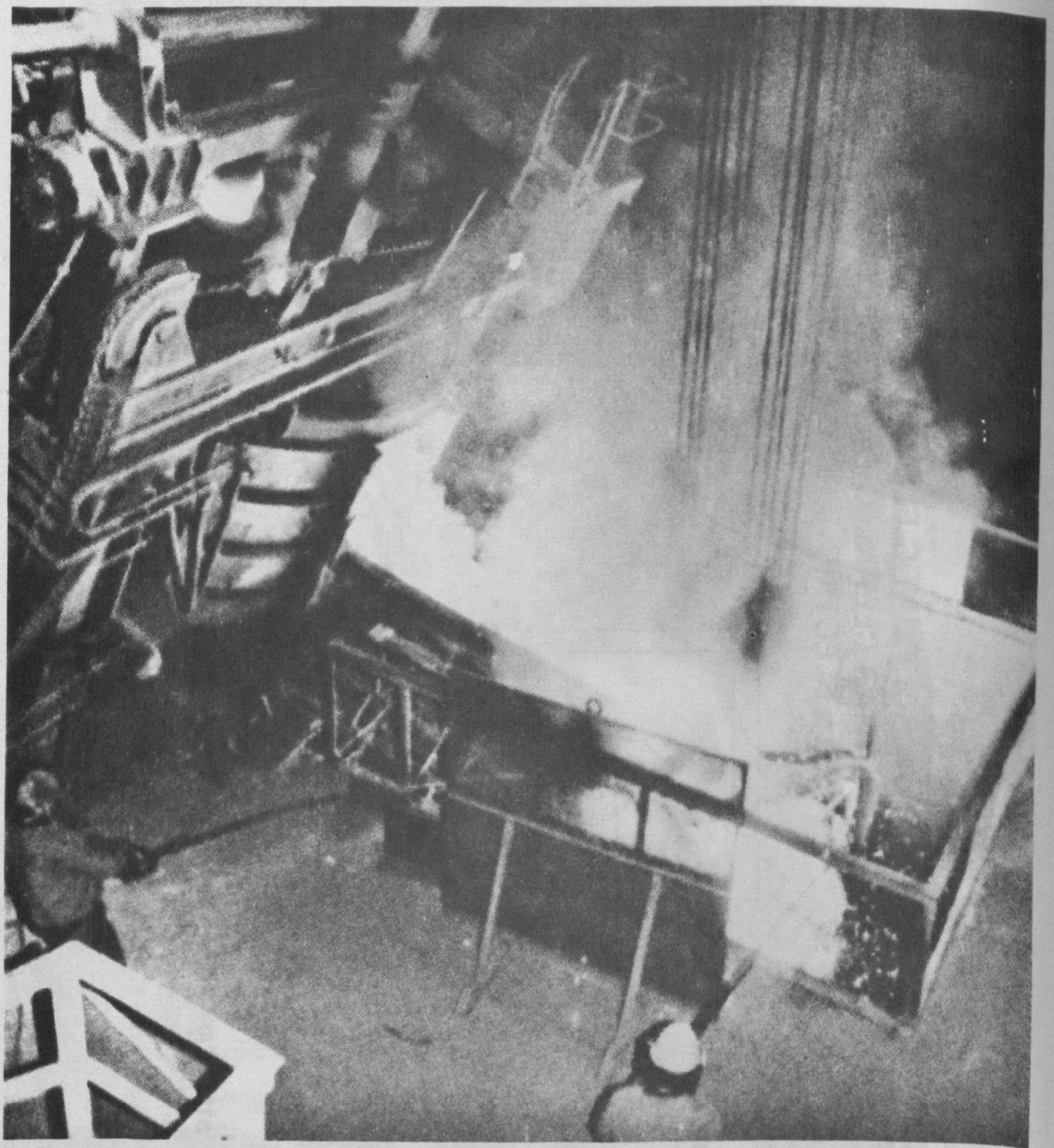


Figure 13 - Fugitive Emissions During The Tapping Operation

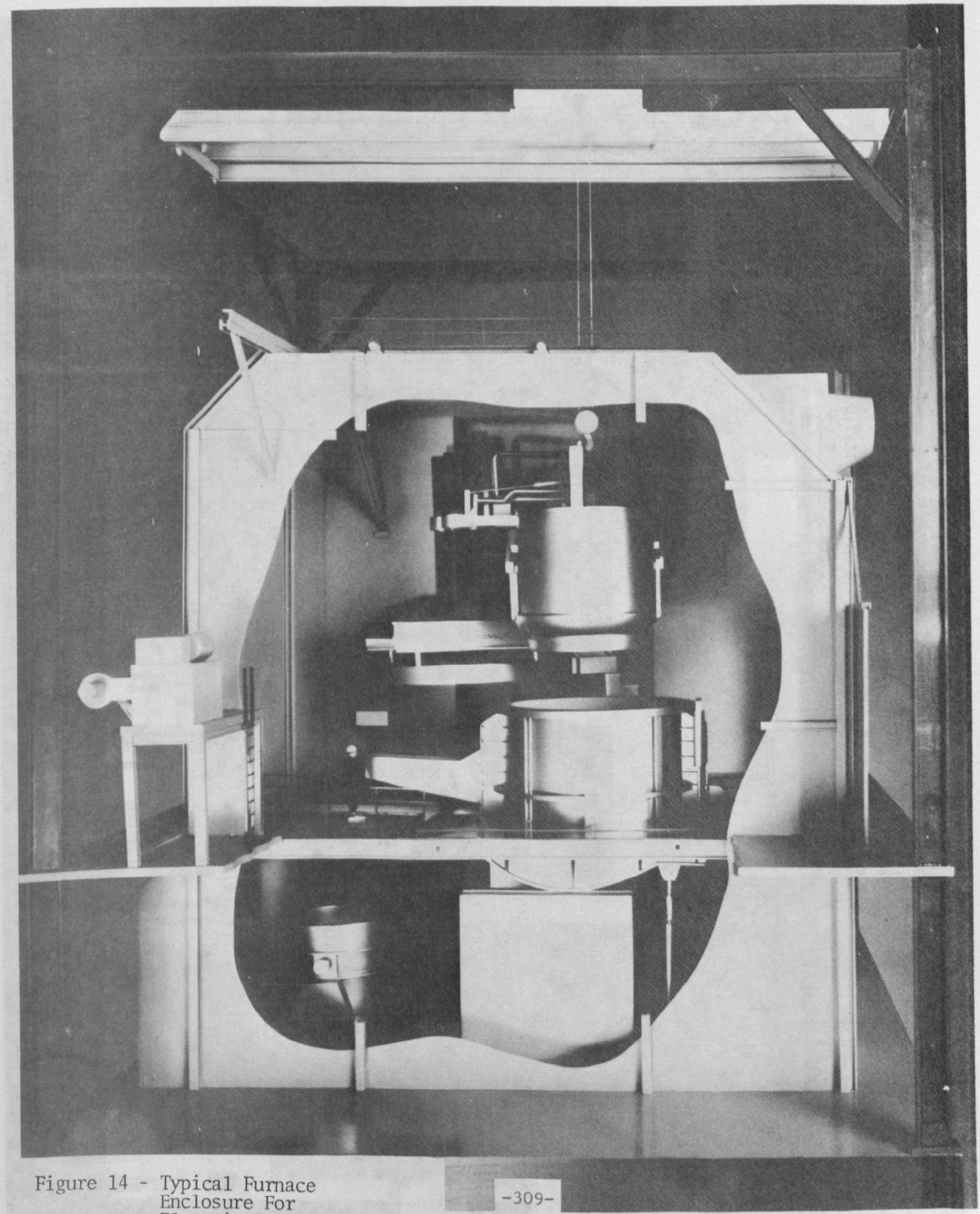


Figure 14 - Typical Furnace Enclosure For Electric Furnace

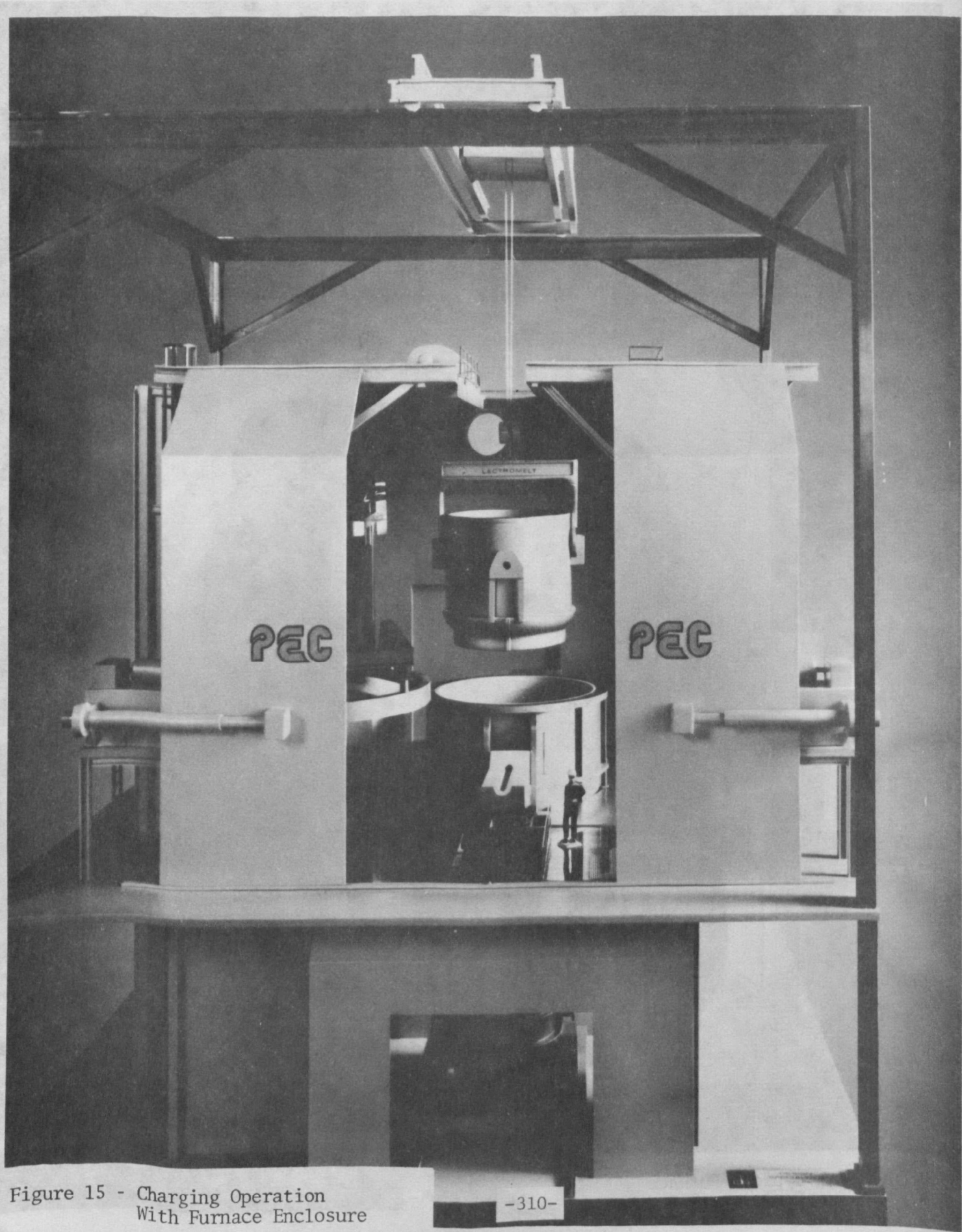


Figure 15 - Charging Operation
With Furnace Enclosure

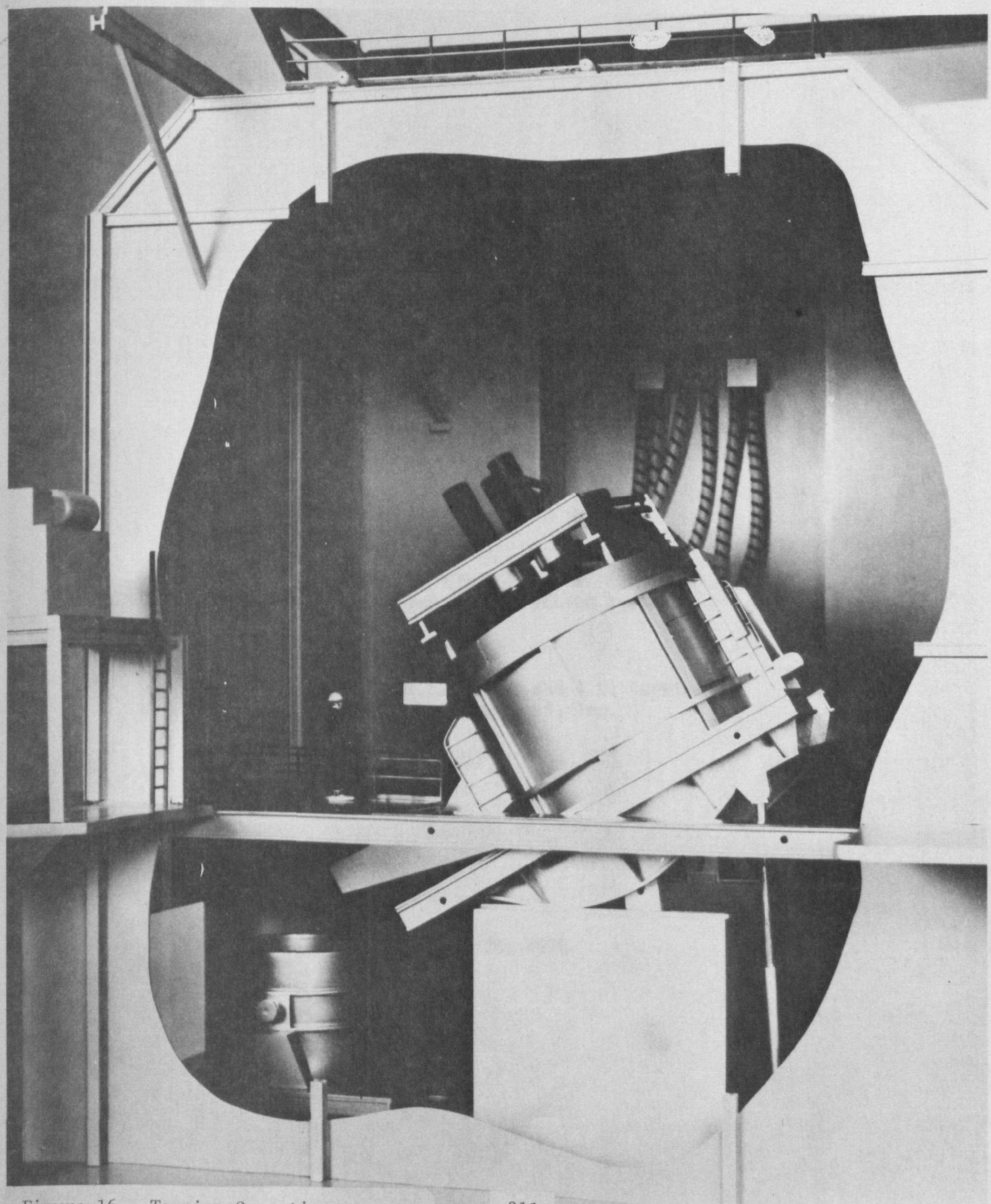


Figure 16 - Tapping Operation
With Furnace Enclosure

CONTROL OF FUGITIVE EMISSIONS AT REVERBERATORY FURNACES AND CONVERTERS

by

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DISCLAIMER

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ABSTRACT

The following are discussed in this paper: the functions of the reverberatory and converter furnaces; methods of charging, operations, slagging, tapping of each type of furnace, and their related emissions during these operations; methods of controlling the fugitive emissions during the aforementioned operations; relative merits and efficiencies of the different methods of controlling fugitive emissions and costs; types of furnaces available and their relative fugitive emissions in comparison with each other; and possible future types of furnaces to minimize emissions.

CONTROL OF FUGITIVE EMISSIONS AT REVERBERATORY FURNACES AND CONVERTERS

Summary

Introduction

A. Copper Smelting Furnaces
   1. The Smelting Process
      a. The Reverberatory Furnace
      b. The Electric Furnace
      c. The Flash Furnace
      d. The Noranda Furnace
   2. Fugitive Emissions - Smelting Furnaces

B. Copper Converters
   1. The Converting Process
      a. The Pierce-Smith Converter
      b. The Hoboken Converter
   2. Fugitive Emissions - Converters

C. Discussion of Fugitive Emission Controls/Practices
   1. Smelting Furnaces

D. Relative Merits and Efficiencies of Different Methods of Controlling Fugitive Emissions - Converters

E. Costs of Fugitive Emission Control Systems
   1. Cost Parameters - Secondary Hooding
   2. Capital Costs
   3. Operating Costs

F. Current and Possible Future Methods/Processes For Emission Control

SUMMARY

In terms of domestic consumption, copper is the largest of the three primary nonferrous metals (lead, zinc, and copper). In 1976, about 1.33 million metric tons of copper were produced in the 16 U.S. smelters from domestic ores, and 1.8 million metric tons from domestic and foreign ores. Vast amounts of particulate and sulfur dixoide (SO_2) emissions are generated in producing this tonnage of copper. Copper smelters are a major source of SO_2, emitting approximately 80 percent of the total amount of SO_2 emitted from the copper, lead, and zinc industries. The industry is implementing control methods to recover some of the SO_2 as a marketable product. All of the smelters have installed pollution control equipment to reduce emissions into the ambient air, for example, electrostatic precipitators to remove particulates from dust-laden air out of roasters, dryers, reverberatory furnaces, and converters; and many of the smelters have acid plants to reduce SO_2 emissions primarily from converters. Even with these emission controls, it is estimated that 52,000 tons/yr of particulates, an indeterminate amount of fumes that contain noncondensables, and 3.5 million tons/yr of SO_2 are still emitted into the air through the stack. (This is based on 1974 reports and does not consider new modifications to the smelters since then.) Sulfur dioxide quantities are especially high because SO_x from multiple-hearth roasters and reverberatory furnaces is dilute, and only a portion is sent to acid plants and the larger portion is emitted without control. Sulfur dioxide from converters can be sent to acid plants only when the converters are in the blowing operation since the SO_2 concentration is high. The emissions discussed so far are called primary emissions because they are either sent to control equipment or are at least ducted to a stack for dispersion into the ambient air.

Emissions of concern in this paper are secondary or fugitive emissions. These fugitive emissions are those that escape through leaks in dryers, roasters, smelting furnaces, converters, and anode furnaces or escape from primary hooding systems not designed with great enough capture velocity, or they escape because of the position of the process equipment in certain phases of operation. Particulate fugitive emissions may be high around multiple-hearth roasters where concentrate is being handled and roasted, old reverberatory furnaces, leaky waste heat boilers, and converters when the converter is not in stack or in-line with the main hood position. Sulfur dioxide fugitive emissions can be considerable from roasters, smelting furnaces, and converters. Various reports state that 2 to 25 percent of the SO_2 generated by the copper industry is fugitive emissions. The amount of fugitives varies considerably within a plant from day to day and hour to hour.

Fugitive emissions are a problem at all smelters, however, and in some plant configurations they are a difficult problem to handle efficiently. The following is a summary of the copper smelters and their process equipment:

	No. of plants	No. of units
Copper smelters	16	
Green feed	5	
Multiple-hearth roaster	4	50
Rotary dryer	7	(includes dryers at concentrator plants)
Fluid bed roaster	4	
Reverberatory furnace	11	25
Electric furnace	3	
Flash furnace	1	
Noranda furnace	1	3
Peirce-Smith converter	15	63
Hoboken converter	1	5
Anode furnace	12	28
Fire refining furnace	1	
Acid plant	13	20
Liquid SO_2	3	4
Dryer or roaster gas to acid plant	4	
Furnace gas to acid plant	6	
Converter gas to acid plant	13	
Total concentrate feed rate of 16 plants, tons/day		≈20,000
Total copper production at 16 plants, tons/day		≈ 5,700

This paper will briefly describe the smelter operations, address the effectiveness and costs of fugitive emission controls, and also discuss some ideas on possible future plant designs to reduce fugitive emissions.

INTRODUCTION

Copper produced by the domestic primary copper industry is mainly from sulfide ores comprised of a variety of minerals. Small amounts of copper ore also are recovered from oxide ores, low-grade waste, and imported ores. Because most of the domestic ore is mined in the southwestern states, the majority of the plants are located in that area. Figure 1 shows the locations of the 16 primary copper smelters in the United States. Because most copper ores are sulfides, copper recovery processes have been developed to treat these ores. These processes recover copper while removing most of the impurities present in copper ore. Remaining impurities are removed by refining.

Copper metal primarily is recovered from copper ores by pyrometallurgical processing with the remainder consisting of hydrometallurgical processes. The pyrometallurgical processes convert ore concentrate into an impure copper called blister copper. The processes consist of roasting or drying, smelting, converting, and refining. The anode copper product (as high as 99.8 percent copper) is then sent to a refinery for final purification.

Figure 2 represents a general flow sheet of the copper industry in the United States. The figure shows the pyrometallurgical steps and the refining processes.

This paper deals with the state-of-the-art in the control of fugitive emissions created in the smelting and converting departments, which will be discussed in the following paragraphs. Before beginning a more detailed discussion of the smelting process, the following brief description of the traditional and still most commonly used smelter operations will be presented.

Ore concentrate, containing about 25 percent copper, is usually railed to the smelter and stored. From the storage area, the ore is generally conveyed to a dryer, then to subsequent operations, or to hoppers above a bank of multiple-hearth roasters. Feed from the hoppers is discharged on the top hearth of a roaster where rotating arms push the concentrate to the center of the hearth where it cascades to the hearth below. From this hearth, the concentrate is directed to the periphery where it cascades to the next hearth and then is directed through several additional hearths. The multiple-hearths are gas- or oil-fired. This roasting process dries the concentrate and removes some of the sulfur in the ore so as to improve the matte grade in the following operation. (Since burning of sulfur is exothermic, the temperature in all of the hearths can be maintained at adequate temperatures with supplemental heating as required.)

The dried or partially roasted (calcined) ore concentrate is usually transferred from the roasters in larry cars to a reverberatory furnace(s).

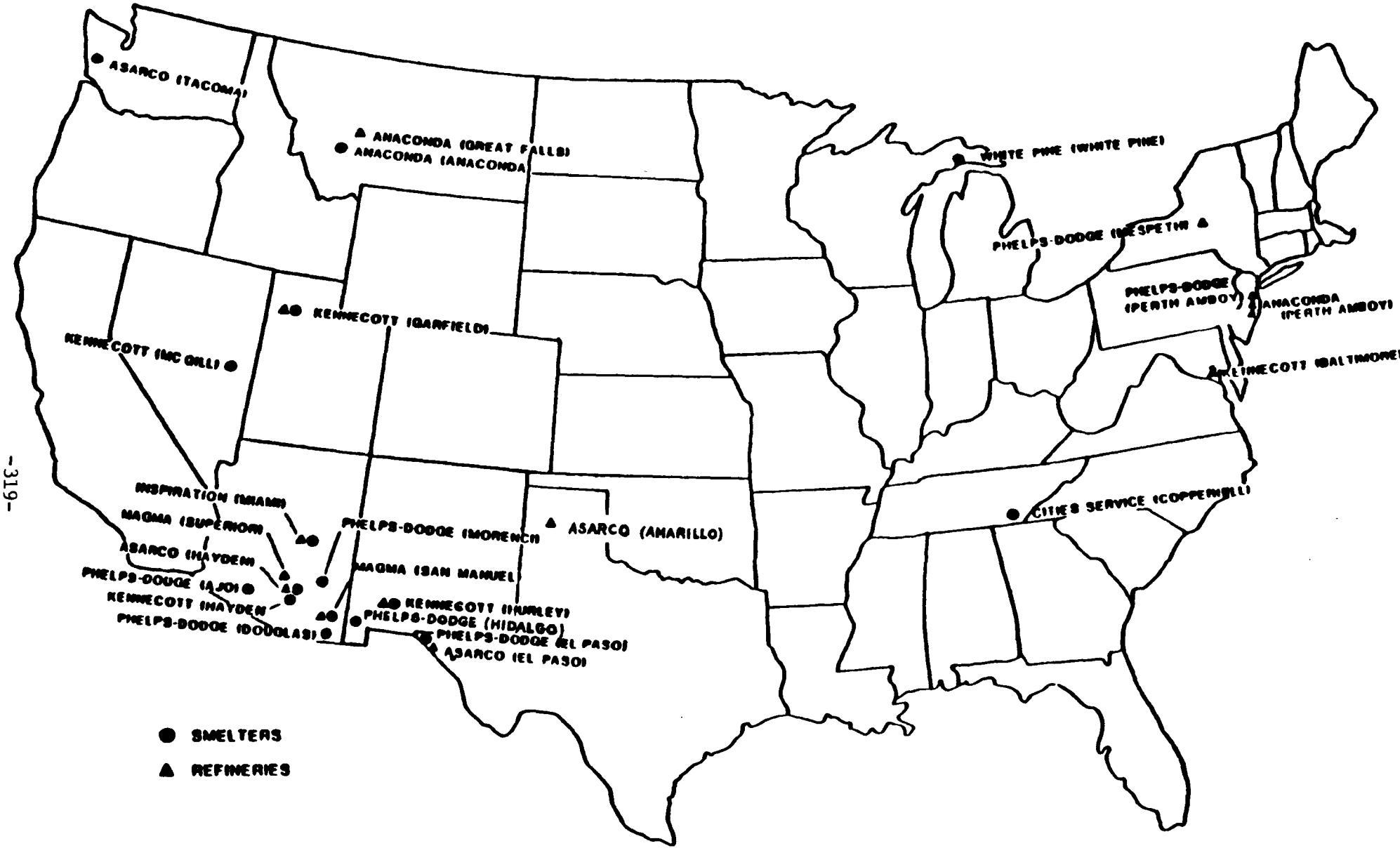


Figure 1. Locations of primary copper smelters in the United States.

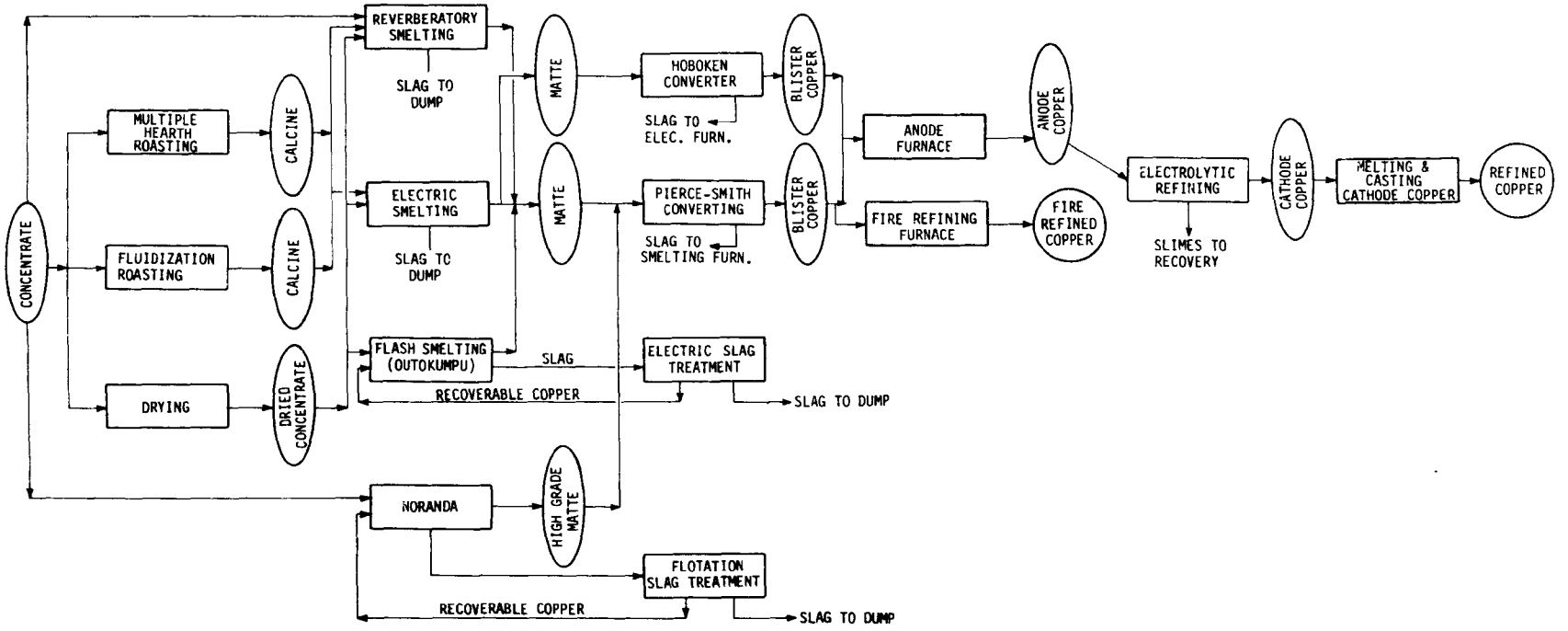


Figure 2. General flow sheet of the copper industry in the United States.

The concentrate is charged into the reverb(s) through pipes and/or hoppers located at the top or along the side walls of the reverb. The concentrate melts by reverberatory heating. The melt with the addition of a silica fluxing agent forms a copper-bearing matte layer and a waste slag layer. The matte is tapped near the bottom of the reverb. The lighter slag is tapped at a higher elevation than the matte. The slag is collected in a slag pot or ladle and carted to, and dumped at, a disposal area. The ladle, in which the matte is collected, is usually transferred by crane to a Peirce-Smith converter (horizontal cylindrical furnace). This furnace converts the matte into blister copper and slag by means of air blown into the converter below the top of the bath line. The reaction is exothermic. The slag, containing recoverable copper, is poured from the mouth of the converter into a ladle, and the slag is then returned to the reverb furnace. After final blowing, the blister copper (about 98 to 99 percent copper) is poured into a ladle and then transferred to an anode furnace. This furnace is usually fired with natural gas and completes the smelter refinement of copper (99.5+ percent copper). The anode copper is poured from the furnace into molds or a continuous casting wheel.

These anodes, varying from 460 to 1000 lb are cooled by quenching and then stored and/or loaded onto rail cars for delivery to a refinery. Figure 2 shows other alternatives to the traditional method of copper smelting.

A. COPPER SMELTING FURNACES

1. The Smelting Process. Copper smelting is the process of heating a roasted or dried sulfide ore concentrate to its melting point to separate much of the iron and undesirable impurities from the copper sulfides and other valuable metals. In the smelting furnace, hot roasted calcines or raw (green) concentrates are melted with siliceous and/or limestone flux, as well as materials with recoverable copper values such as converter slag, flue dusts, oxide ores, and copper scrap. As the temperature in the furnace increases, a complex series of reactions takes place and the charge separates into fractions. The sulfur in the charge combines preferentially with copper to form stable copper sulfides, which are mutually soluble with the molten copper metal. This fraction, known as matte, typically contains from 20 to 45 percent copper (using the reverberatory practice) as well as other impurities such as sulfur, antimony, arsenic, iron, and precious metals. Since copper has a weak chemical affinity for oxygen, very little copper oxide forms. Iron combines preferentially with oxygen to form iron oxides, which in turn react with the flux to form iron silicates. These compounds and any calcium, magnesium, and aluminum minerals that were present in the charge form a slag of lower specific gravity that is insoluble in the matte and floats on top of it. Any sulfur left over from the matte- and slag-forming reactions reacts with additional oxygen to form SO_x gas (primarily SO_2, some SO_3), which mixes with the combustion off-gases. The energy necessary for smelting can be provided by fossil fuel combustion, electrical power, and partially by heat of reaction from the oxidation of iron sulfide to iron oxide.

a. The Reverberatory Furnace. The workhorse of the U.S. copper industry is the reverberatory furnace or reverb (Figure 3), which was first introduced in 1879 and is still used in modified form at 11 of the 16 domestic smelters. The modern reverb is an arch-roofed or suspended-roof horizontal chamber, generally about 35 m in length and 10 m in width. Heat is supplied by fossil-fuel-fired burners located at one end of the furnace. A reverberatory furnace operates on the basis of the heat from the flame radiating from the roof onto the charge. Thermal efficiency of the reverb is low. However, the reverb is flexible with respect to concentrate composition and is capable of accepting as much as 1800 metric tons of material per day.

Although specific methods may vary considerably, reverbs are generally charged either through the furnace top or along the top portion of the side walls. Belt slingers (high speed conveyors), hoppers, and Wagstaff guns (inclined chutes) may be used to better distribute the charge over the molten bath. Drag chains and screw conveyors have also been used for charging. Continuous charging is not practiced because of the problem of creating local thermal imbalances that could reduce the furnace temperature below the efficient level needed for the smelting reactions. Slag is drained periodically from skimming bays at one end of the furnace and conveyed by short launders to slag pots. The slag can be cooled, solidified, granulated, or dumped molten. Matte is withdrawn periodically through tap holes in the lower furnace wall. The matte flows down launders and into ladles, which are conveyed to the converter by overhead cranes.

A. BALLOON FLUE
B. CONVERTER UPTAKE HOOD
C. ADDITIVE SYSTEM
D. MONITOR
E. E.O.T. CRANE 60/60
F. PEIRCE-SMITH CONVERTER
G. LADLE
H. CONVERTER AISLE
I. REVERBERATORY FURNACE
J. WASTE HEAT BOILER
K. REVERB. FLUE

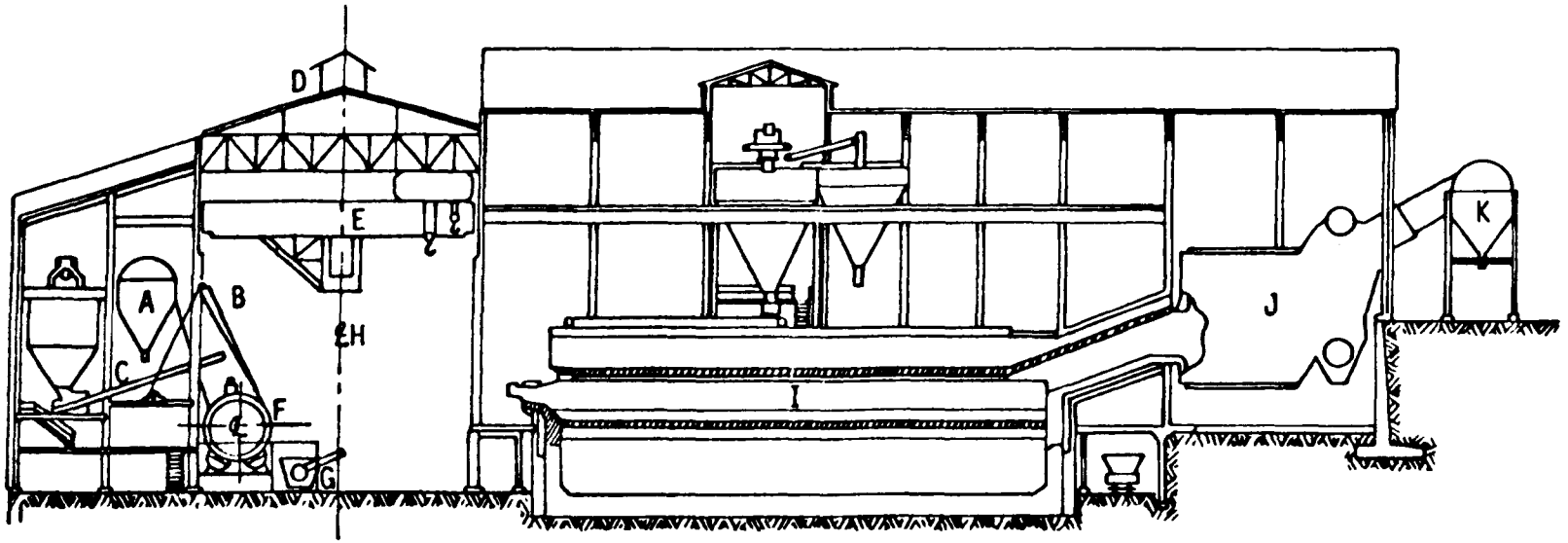


Figure 3. Typical reverberatory furnace and converter

(Adapted from Kirk-Othmer Encyclopedia of Chemical Technology, Second Edition, Volume 6, page 148.)

The outlet gases from the reverb, which may run upward of 120,000 m^3/h, usually contain between 0.2 percent and 1.5 percent SO_2. The specific SO_2 content is dependent upon the sulfur content of the ore and whether or not the concentrate was roasted prior to smelting. Although there is considerable air infiltration, the principal reason for the low-SO_2 content is the substantial quantity of combustion air that must be provided to the furnace. The outlet gases contain vapors (i.e., water vapor and some sulfurous and sulfuric acid) as well as considerable dust and fume, including trace metal compounds of arsenic, antimony, lead, and zinc. These gases are generally passed through waste heat boilers to recover much of the heat of the combustion gas, and are finally cleaned of particulate by means of hot electrostatic precipitators or baghouses and vented to the atmosphere.

b. The Electric Furnace. The electric copper smelting furnace (Figure 4) has traditionally been used in Scandinavian areas where hydroelectric power is cheap and fossil fuels are expensive. The first such furnace in the United States started operation in 1972, and two more smelters have since adopted this technology. The electric furnace is rectangular in cross-section with a firebrick sprung-arch roof. The largest are about 35 m in length and 10 m in width. Carbon electrodes are placed in contact with the molten slag, and the heat required for smelting is generated by electrical resistance of the slag to the submerged arc between electrode pairs. Electrical ratings vary up to 51,000 kVA. The chemical and physical changes that occur in the molten bath are similar to those that occur in a reverb, although the thermal efficiency of the electric furnace is much higher.

The charge of concentrate and fluxes is delivered to the roof of the furnace by drag conveyors and then fed to the molten bath through multiple-feed spouts near the electrodes and/or between the electrodes and sidewalls. As the charge materials melt, they settle into the bath and form additional matte and slag. Separate launders or chutes on the furnace end wall may be used to charge converter slag and reverts. Matte is tapped into ladles from tap holes placed in the hearth area near one end wall. Slag is skimmed from tap hole(s) in the opposite end wall and delivered by launder(s) into slag pot(s), which are usually hauled to the dump by trucks.

Although originally designed as an alternative to the use of expensive fuels in the Scandinavian countries, the electric furnace also facilitates air pollution control. Large amounts of combustion air are not required, resulting in an outlet gas about an order of magnitude less than a reverb, but containing the same quantity of sulfur oxide emissions. SO_2 concentrations of 2 to 4 percent can be expected, and particulate emissions should be less than a reverb because of the lower gas volume and more uniform gas flow. The electric furnace off-gas at all three domestic smelters is combined with other high-SO_2 gas streams and used as feed to contact sulfuric acid plants.

c. The Flash Furnace. A more recent development in copper metallurgy is the continuous flash furnace, which is more efficient in terms of energy consumption and also produces a more easily controlled stream of flue gas than the reverb or electric furnaces. Flash furnaces are of two types, the Outokumpu Oy and the Inco, which differ primarily in their use of either preheated air or commercial-grade oxygen to sustain the smelting reaction.

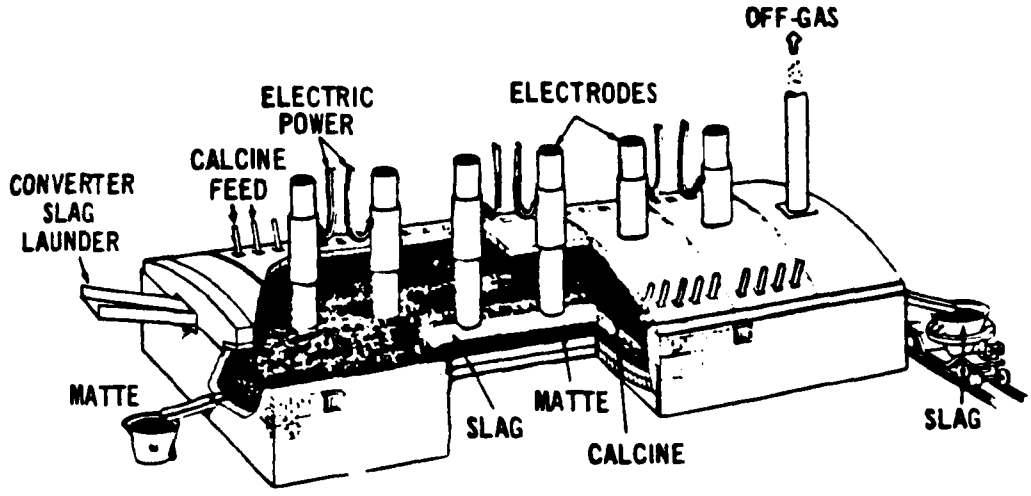


Figure 4. Electric smelting furnace.

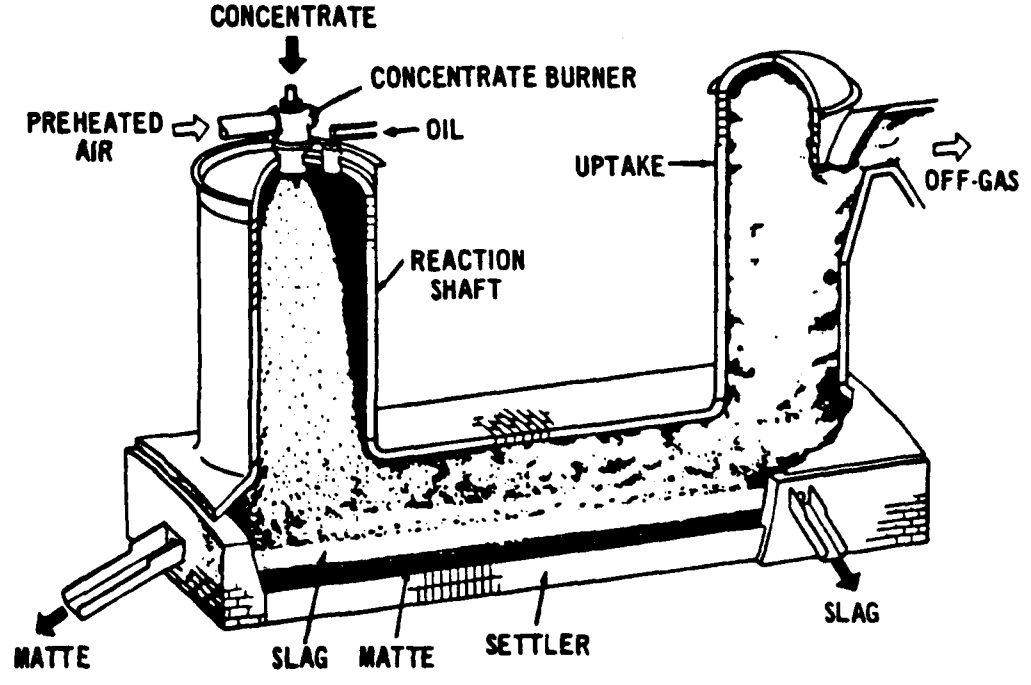


Figure 5. Outokumpu flash smelting furnace.

The flash furnace is in widespread use throughout the world, although only one is operating in this country (under license from Outokumpu).

The Outokumpu furnace (Figure 5) combines the functions of roasting, smelting, and partial converting in a single furnace with three sections--reaction shaft, settler, and uptake shaft. Dried ore concentrates are injected continuously along with flux and preheated air into the reaction shaft through concentrate burners. Oil may also be injected into the shaft. The finely divided concentrate burns in a "flash" combustion as the particles fall down through the shaft, and the heat released from the combustion of the oil and sulfur sustains the smelting reaction. The process is similar to the combustion of pulverized coal. The molten particles fall into the settler part of the furnace and separate into matte and slag layers. The matte, which contains 45 to 75 percent copper, is tapped from the settler and transferred to converters for further processing. The slag, which contains too much copper to discard, is also processed further. The flash furnace in the U.S. does not have the flexibility to recover copper from converter slags, and as a result these also require slag treatment facilities. The furnace outlet gases are withdrawn from the uptake shaft. They contain 10 to 20 percent SO_2 and considerable quantities of entrained molten or semimolten particulate matter. The gas is cooled in a waste heat boiler, cleaned of dust in an electrostatic precipitator, and used for sulfuric acid production. The Inco flash furnace (Figure 6), which requires a low-cost source of oxygen to be economically competitive, produces gases containing 75 to 80 percent SO_2, ideally suited for liquid SO_2 manufacture.

d. The Noranda Furnace. A newer type of smelting furnace used in the United States is the Noranda continuous furnace (Figure 7), in which roasting, smelting, and partial converting reactions are combined in a vessel similar to a lengthened Peirce-Smith converter. One U.S. plant has started operating Norandas within the past year. The reactor is a horizontal cylindrical furnace about 21 m long. It is fired from both end walls, and oxygen-enriched air is blown into the matte layer through side-mounted tuyeres. The furnace can be rotated on its horizontal axis, bringing the tuyeres out of the bath and stopping the smelting process. The compact design facilitates process control, and the domestic Noranda smelter is highly instrumented. The Noranda was originally developed as a one-step process that would eliminate the converter, thus significantly reducing capital costs and eliminating the need for a crane aisle. In both of its commercial applications to date, however, the use of a converter has been retained to allow better trace element control, increased production, and longer reactor-lining life.

Concentrate and fluxes are fed to the Noranda by a slinger at one end that spreads the feed over the molten bath. The high-grade matte, which typically contains approximately 70 percent copper, is periodically tapped from the side of the furnace and then transported by ladles to standard converters where it is batch-treated to remove additional sulfur and iron prior to fire refining. Slag, which contains 6 to 8 percent copper, is periodically tapped from the end of the vessel opposite the slinger. Slag is upgraded by milling, producing a concentrate (which is returned to the reactor) and a tailing (which is discarded). The off-gases leave the Noranda furnace through its mouth where they are captured by water-cooled hoods and ducted to a waste

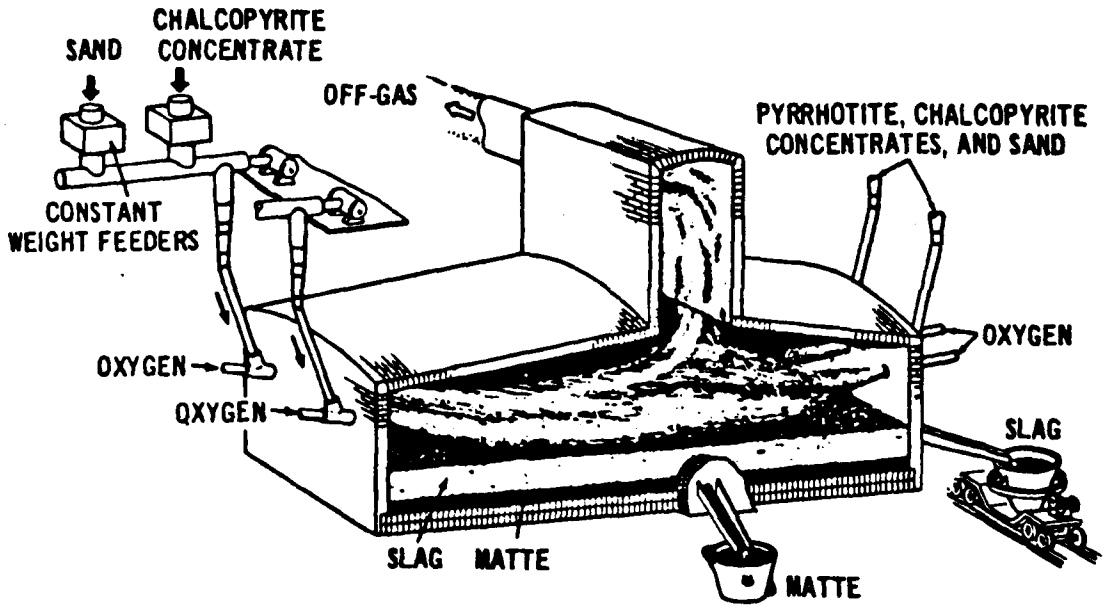


Figure 6. Inco flash smelting furnace.

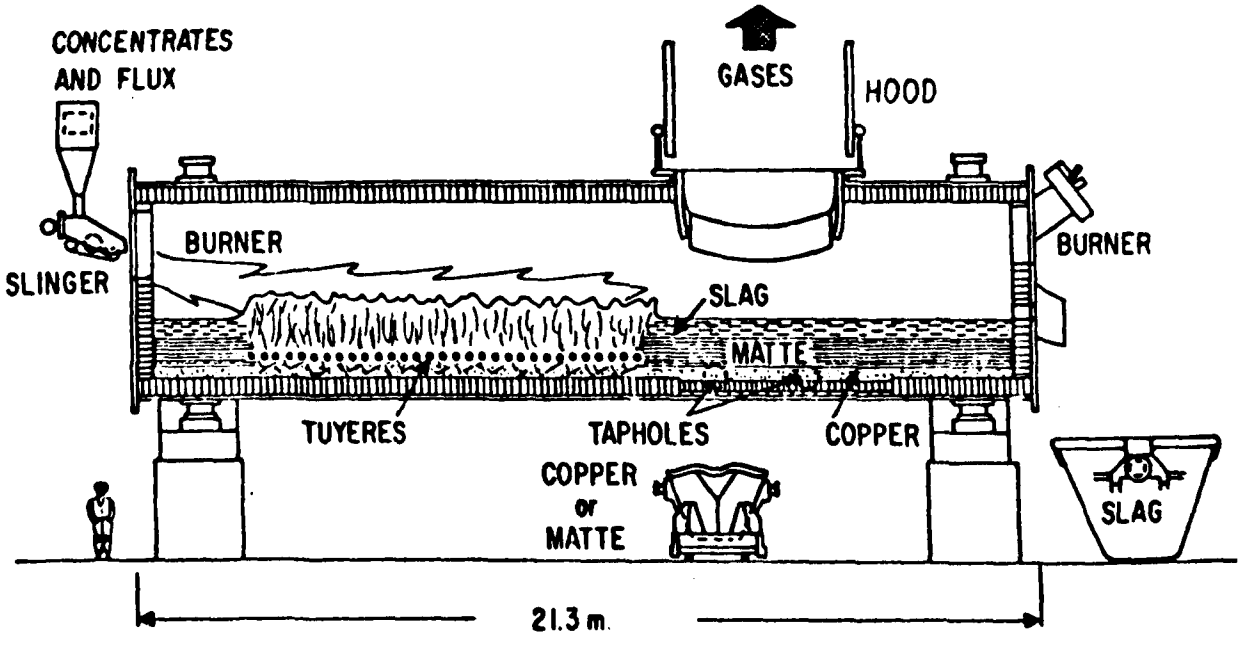


Figure 7. Noranda process reactor.

heat boiler. The gases are passed through cyclones and electrostatic precipitators to remove particulate matter, and then used as feed to a sulfuric acid plant. With 30 percent oxygen enrichment, the off-gases to the acid plant contain 6 to 7 percent SO_2. The SO_2 concentration will be only about 4 percent if oxygen enrichment is not used.

2. Fugitive Emissions - Smelting Furnaces. Fugitive emissions are those that escape from processes because of the nature of the operation involved or escape from primary control systems for various reasons (i.e., less than 100 percent efficiency; inadequate capture velocity). The emissions of concern in the copper industry consist of particulates, trace metal fumes, and sulfur dioxide (SO_2). Some fugitive emissions (fugitives) occur with all types of smelting furnaces. The emissions are present at all charging and tapping locations, as well as through leaks in the furnaces and auxiliary equipment. During the operation of smelting furnaces, the amounts of fugitive emissions will depend on:

- Charging technique
- Furnace design and age
- Amount of time required to charge feed and converter slag
- Location and efficiency of any existent secondary hooding
- Furnace maintenance and operations.

Fugitive emissions will be less variable over time with the continuous flash and Noranda furnaces than the reverb or electric furnaces. The release of materials through cracks and leaks is most serious with the reverb because of both its increased gas volume and the fact that these furnaces are generally older and in poorer repair than the other designs. The electric furnace presents the additional problem of leakage from the furnace at the roof line around the electrodes, as well as burning of the electrode paste in this area at the openings in the band containers.

With the Noranda furnace, the primary hood cannot maintain a perfectly tight seal with the reactor during smelting, and some fugitive emissions will be released. There will also be dilute gases discharged through the mouth when the furnace is rotated out from under the hooding system, even though the smelting reactions will have largely stopped.

A final fugitives source near all smelting furnaces is the ladle, which is used to transport the molten matte to the converter aisle.

B. COPPER CONVERTERS

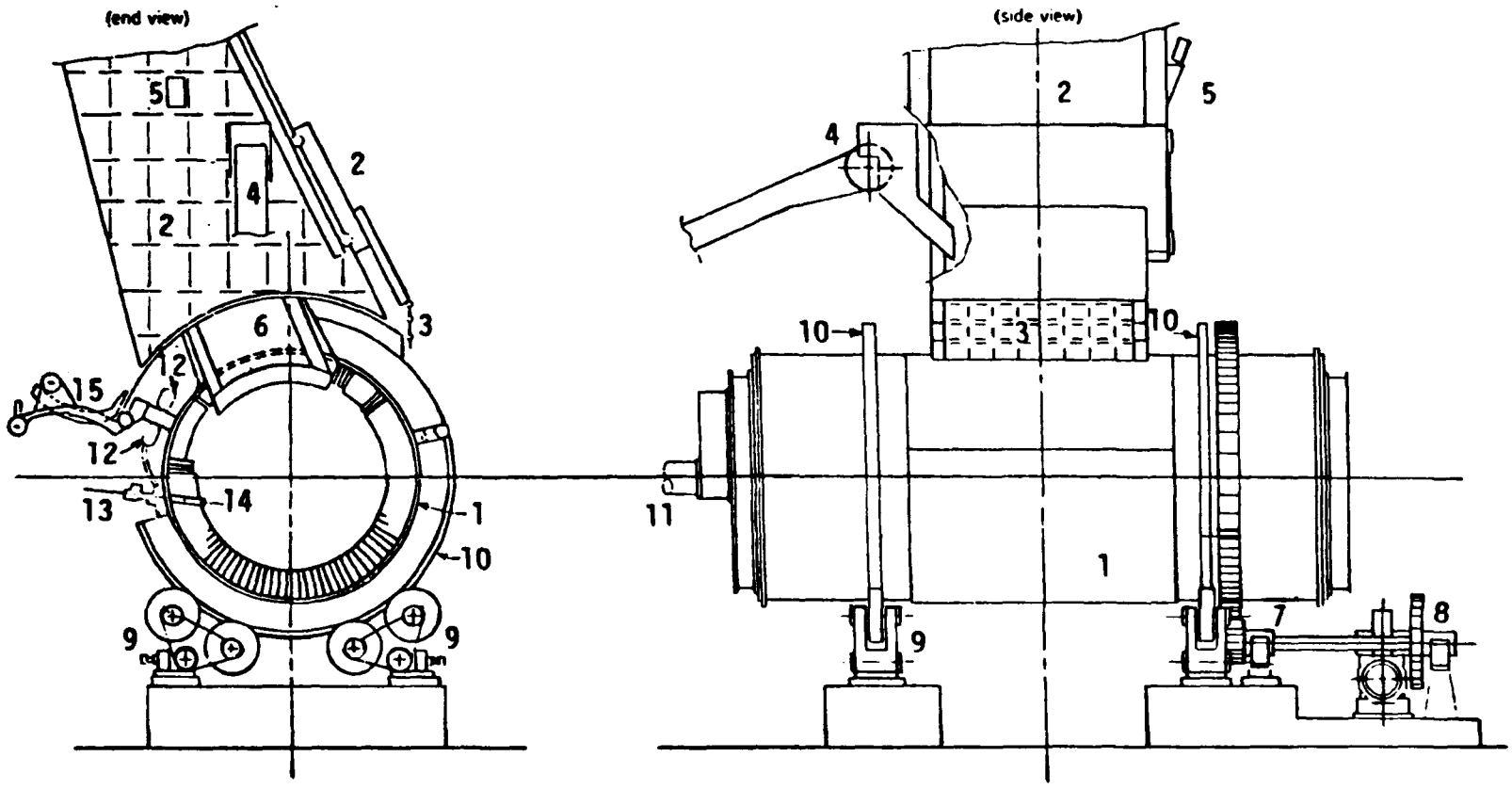
1. The Converting Process. Copper converting is the batch-type process by which blister copper is produced from the copper/iron/sulfur matte formed in a smelting furnace. It is essentially an adaptation of the Bessemer process developed by the steel industry. Molten matte and silica flux are charged to the converter, and air or oxygen-enriched air is blown through

tuyeres into the melt to oxidize the iron sulfides. This results in the formation of an iron silicate slag, which floats on top of the matte and can be skimmed from the converter. Further blowing oxidizes the copper sulfides to blister copper, which is approximately 98 percent pure. Remaining impurities can be removed by fire refining at the smelter and electrolysis at a refinery. The slag contains sufficient copper to be returned to the smelting furnace. Almost all of the sulfur remaining in the matte is removed in the converter, and the SO_2 content in the off-gas is sufficiently high that many smelters produce sulfuric acid from this feed stream. However, sulfur recovery is complicated by the cyclic nature of the converting operation, which results in major and frequent fluctuations in off-gas volumes and SO_2 concentrations. Converting is exothermic and no fuel is needed to maintain the bath in a molten state. In fact, smelter reverts, copper scrap, and, in some cases, copper concentrate may be charged to reduce its temperature and prevent damage to the refractory lining.

a. The Peirce-Smith Converter. The Peirce-Smith converter (Figure 8) is a horizontal, refractory-lined, cylindrical furnace with an opening in the horizontal side that serves as a mouth for charging feed materials and pouring slag and blister copper. It can rotate through an arc of about 120° from the vertical for operational purposes. Compressed air is supplied through a horizontal row of tuyeres along its back. The standard unit is about 4 m in diameter and 9 m in length. First developed in 1909, the Peirce-Smith converter is now used at 15 of the 16 domestic copper smelters, with as many as nine units installed at one plant. Two or three converters are generally associated with each smelting furnace. The Peirce-Smith converter is a relatively efficient furnace whose high air flows permit both large copper throughputs and the charging of bulky materials and copper scrap.

With the converter rotated partly forward, molten matte that has been transported from the smelting furnace by the overhead crane is charged through the converter mouth. The compressed air blast is put on, then the converted returned to its operating position. Silica flux can be added either before or after the converter is rotated to the upright position. The initial blow is for the formation of slag during which the iron sulfides in the matte are preferentially oxidized and sulfur is removed as SO_2 with the off-gas. Periodically, slag is skimmed from the converter and fresh matte and scrap are added. Blowing is discontinued during the skimming operation. The process is continued until a sufficient quantity of copper sulfide (white metal) has accumulated in the converter and the iron has been removed as an iron silicate slag. The copper blow then begins, and it continues until the white metal has oxidized, forming mainly SO_2 and blister copper. Upon completion of the copper blow, the blister copper is poured into ladles for transfer to anode furnace(s). A complete converter cycle lasts about 10 to 12 hours for a 40 percent matte grade.

The outlet gases from the Peirce-Smith are released through the same mouth used for charging and pouring. When the converter is in its upright operating position, the gases are drawn into a fixed hooding system situated above the converter. The hood has a retractable gate that can be lowered to reduce leakage, and the off-take is controlled by regulation of the draft on the flue system. By stoichiometric calculation, the SO_2 content in the converter off-gas can be as high as 20 percent. However, the actual SO_2



(1) Shell; (2) Hood; (3) Air baffle; (4) Stack fluxing belt and chute; (5) Radiation pyrometer; (6) Converter mouth; (7) Turning mechanism; (8) Turning motor and speed reducer; (9) Rollers; (10) Turning rungs; (11) Air-supply duct; (12) Air-distribution ducts; (13) Automatic mechanical tuyère; (14) Tuyère; (15) Puncher protection shields.

Figure 8. Peirce-Smith converter.

(Adapted from Kirk-Othmer Encyclopedia of Chemical Technology, Second Edition. Volume 6.)

concentration is in the range of 2 to 10 percent primarily because of excess air infiltration into the hood, which cannot be physically tight with the converter. Oxygen enrichment of the compressed blast air can be used to partially reduce the dilution effect of this air infiltration. Although a reduction in SO_2 content is not desirable, the dilution air does serve to cool the hot gases somewhat, which is necessary to prevent damage to downstream gas cleaning equipment such as electrostatic precipitators or baghouses. Waste heat boilers are also often employed on the converter gas. The cleaned gas is used for acid production at most smelters; however, three domestic plants release it without specific SO_2 control. To minimize the fluctuations in gas flow and SO_2 content that occur throughout the operating cycle, and ensure a relatively steady feed to the acid plant, the operation of two or more converters must be properly scheduled to ensure that one furnace is always charged with matte and blowing.

b. The Hoboken Converter. The Hoboken converter (Figure 9) is an alternative to the Peirce-Smith that was designed to largely eliminate the problem of excess air infiltration into the flue gas off-take system. First developed in the early 1930's in Belgium, a Hoboken has been operated at a single domestic primary smelter for about 4 years; it is also installed in a number of foreign copper plants. The design of the furnace itself is similar to the Peirce-Smith. However, instead of withdrawing the off-gas through the converter mouth into a hood, the Hoboken is equipped with an integral side flue located at one end of the furnace. Shaped like an inverted "U," this flue, or siphon, rotates with the converter, as does the cylindrical duct to which it is connected. A counterweight balances the siphon. The cylindrical duct is connected by an airtight joint to a fixed vertical duct that leads to the gas cleaning system. The Hoboken thus provides a direct link at all times between the converter and the gas off-take, irrespective of its operating position.

2. Fugitive Emissions - Converters. Fugitive emissions from converters are always present during charging and tapping, and during slag and copper blowing. Fugitives also occur during the rolling in and out of the converter with blast air on. During the operation of a Peirce-Smith converter (10 to 12 hours to produce blister copper), the fugitive emissions that occur are dependent on:

- Matte grade - affects blowing time
- Charging technique
- Equipment design
- Material additions of anode slag, cold material (i.e., skulls, cleanings, etc.) and flux.

The Peirce-Smith converter is a principal source of fugitive emissions in a copper smelter. Basically, the furnace operates under a negative draft, but due to the clearance required between the mouth of the converter and main hood/gate, ambient air infiltrates into the main hood. This infiltration disrupts the flow and causes some of the discharge gases from the converter to

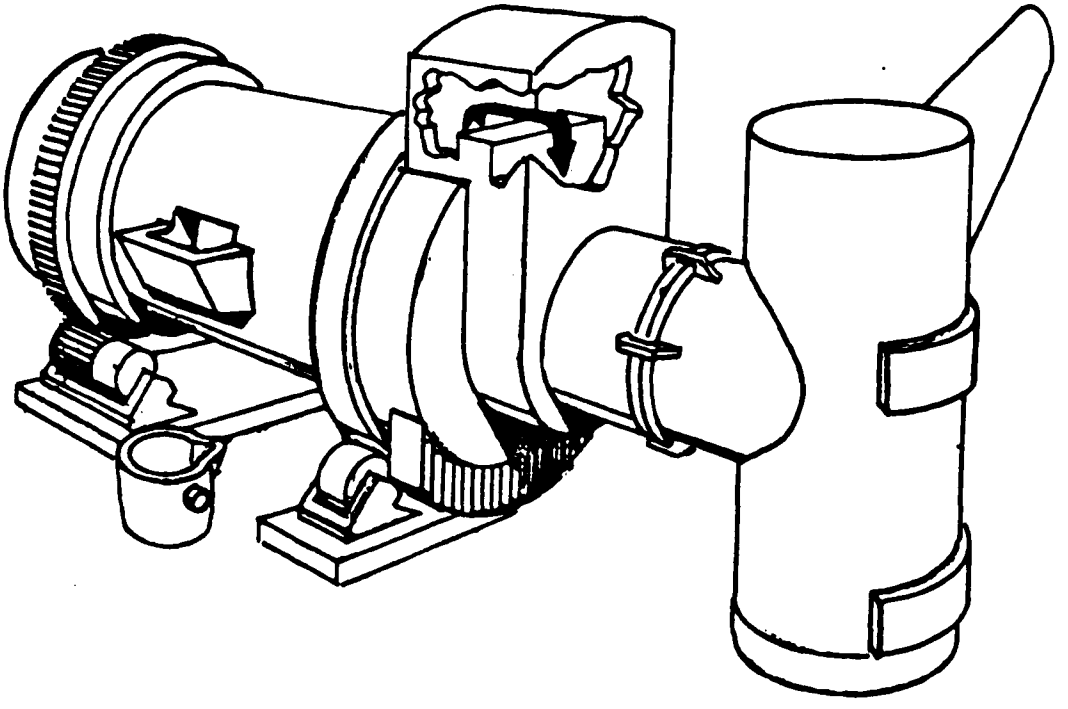


Figure 9. Hoboken converter.

escape through this opening. Fugitives increase significantly during the times in the operating cycle when compressed air is blown through the tuyeres. Substantial fugitive emissions also occur during charging and pouring operations when the converter is rolled out and the hood is no longer positioned above its mouth.

The Hoboken converter's operating cycle and charging and pouring operations are similar to the Peirce-Smith. The principal difference is that the link to the gas cleaning system is never broken, as is the case when the Peirce-Smith rotates forward from under its hood. With this in mind, no fugitive emissions theoretically should escape the Hoboken. In actual practice, under good controlled operations, the Hoboken can eliminate fugitives. A constant zero or slightly negative draft can be maintained; thus minimizing fugitive emissions at the converter mouth. Maintenance of the proper draft is complex. If excessive fan draft occurs, this will cause entrainment of the material in the molten bath, which carries over into the siphon system and forms accretions that restrict the gas flow and would disrupt the draft at the mouth of the converter and would eventually cause emissions.

Additional sources of fugitives common to both types of converters can be leaks in the off-gas ductwork and the ladles that carry matte to the converter and ladles that transport blister copper to the anode furnaces.

C. DISCUSSION OF FUGITIVE EMISSION CONTROLS/PRACTICES

1. Smelting Furnaces. Methods to decrease fugitive emissions around a reverberatory and electric arc furnace could include the following:

- Good, tight fitting refractory/maintenance of the furnace and its auxiliaries to reduce fugitive emissions through leaks in the brickwork, and around electrodes and inspection ports
- Hooding over slag and matte launders to pick up fugitive emissions during tapping and slagging operations
- Hooding over slag and matte ladles to pick up fugitive emissions during tapping
- Having adequate draft to maintain furnace under slightly negative pressure

The Noranda furnace, although a smelting furnace, is similar to the Peirce-Smith converter in design and operation. Thus, fugitive emissions can be treated in the same manner as a converter.

D. RELATIVE MERITS AND EFFICIENCIES OF DIFFERENT METHODS OF CONTROLLING FUGITIVE EMISSIONS - CONVERTERS

Control of fugitive emissions in the vicinity of Peirce-Smith converters could reduce or nearly eliminate these emissions and help improve working conditions for smelter plant personnel. Control can be achieved with varying degrees of success by the addition of fixed or movable secondary hoods or by

enclosure of the converter building. Efficiency of fugitives control ranges from 30 to 100 percent, depending upon hooding configurations as outlined in Table 1. Following are the major types of secondary hoods:

- Fixed type -- made of structural steel with an eliptical cross section. It is attached to the primary or uptake hood.

- Fixed and movable -- consist of a movable intermediate hood and a hood fastened to the gate. Both hoods are made of structural steel with eliptical cross sections so that they telescope in the retracted mode.

- Swing-away type with a fixed overhead hood -- made of structural steel, refractory lined, and supported by pivot arms with a motorized drive to permit positioning during blowing and pouring operations.

- Combination of fixed and movable and swing-away type.

- Controlled building evacuation with adequate number of air changes discharging to a fabric filter (baghouse).

The positions of the movable hoods for the various converter operations are outlined in Table 1.

Estimated fugitive emission control efficiencies are presented in Table 2.

A descriptive summary of the secondary hooding systems is shown in Table 3. This table points out some of the operational and maintenance problems involved, and factors that affect control efficiency.

E. COSTS OF FUGITIVE EMISSION CONTROL SYSTEMS

1. Cost Parameters - Secondary Hooding. This cost study includes various items that must be installed or modified to achieve control of fugitive emissions from the Peirce-Smith converter (this furnace being the worst source of secondary emissions). It does not include costs of certain operating procedures that would minimize fugitive emissions; e.g., allowing minimal clearance between the primary uptake hood and the apron of the converter, or maintaining proper matte charges to provide for direct flow of gases from the mouth of the converter to the centerline of the primary uptake hood.

The following items are considered in this cost study:

- A fixed hood having an elliptical cross section of 17 ft 6 in. on the major axis and 7 ft on the minor axis, and 9 ft 6 in. long. The plate is 3/8-in. carbon steel, with stiffeners of 7-in. channels. The fixed hood is bolted to the primary hood and to the smoke plenum of the secondary duct system.

Table 1. POSITIONS[a] OF MOVABLE HOODS

Type	Matte addition	Blowing or holding	Skimming	Rabbling	Collar pulling	Pouring
Movable	Retracted	Extended	Extended	Partially or fully extended	Retracted	Extended
Gate hood	Retracted	Extended	Extended	Partially extended	Retracted	Extended
Swing-away	Retracted	Retracted or in operating position	Operating position	Retracted	Retracted	Operating position

[a] Following are definitions of hood positions:

Retracted - hood is in its highest or extreme position away from the converter.

Extended - the hood is in its lowest or operating position.

Partially extended - hood is extended as far as practical to maximize secondary emissions control.

Table 2. PEDCo's ESTIMATED HOODING EFFICIENCIES[a]

(Values in percent)

Hood type	Matte or hot metal addition	Blister or hot metal pouring	Skimming or rabbling	Blowing
Fixed	30-50	30-50	30-50	60-70
Fixed and movable	30-50	40-70	40-70	70-90
Fixed and swing-away	30-50	80-90	50-70[c]	80-90
Fixed, movable, and swing-away	30-50	80-90	60-80[c]	80-90
Enclosed building	95[b]-100	95[b]-100	95[b]-100	95[b]-100

[a] Most system efficiencies would be higher if air motions (i.e., open doors, man-cooling fans, monitors, etc.) could be eliminated. Skimming is removal of slag from the converter by tilting of the converter. Rabbling is removal of slag from the converter by tilting of the converter and manual use of a rake to work the molten bath.

[b] Reduced efficiency due to air motions; if doors are left open this efficiency could drop to 50%.

[c] Efficiency during skimming would be similar to blister pouring.

Table 3. SUMMARY OF CURRENT FUGITIVE EMISSION CONTROL SYSTEMS

Type	Design and operation	Operational and maintenance problems	Efficiency
Monitor, Natural - U.S.	Simple design. Relies on outside air movement for removal of emissions from the area.	Haze in building during emissions; outside air movement affects time required to clear the area. Crane operator and maintenance personnel working above the converter may be required to wear face aspirators. Visible emissions in the monitor area. Maintenance in the converter area, electric overhead traveling crane and roof trusses for removal of the settled emissions other than gases.	Dependent on outside air currents and inside air motion.
Monitor, powered	Simple design. Large air movement required at the fans. Removal rate is constant.	Blind pockets or short-circuited flows could cause haze and emission buildup in the roof line area. Crane operator may require the use of a face aspirator at times. Maintenance of fans and drives. Visible emissions at each powered monitor. Maintenance in the converter area, electric overhead traveling crane and roof trusses for removal of settled emissions other than gases.	Dependent on number of monitors, fan size, building design above the converter proper; air motions.
Fixed hood with secondary emission ducting - U.S.	Clearance problems for crane hook and cables during collar pulling or matte additions. Retrofit difficulties for ducting, fans, breeching, and dust bins. Operational at all times that converters are on line. Good face and capture velocities required.	Operational damage to hood by swinging or uncontrolled electric overhead traveling crane action during matte addition or collar pulling. Maintenance is less in the converter area, electric overhead traveling crane and roof trusses due to particle buildup.	Dependent on the distance of the mouth of the fixed hood from the emission source; also on the capture and face velocity created by the fan at the mouth of the fixed hood.
Enclosed converter hood-swing away type with fixed hood	Clearance problems for crane hook and cables during collar pulling or matte additions (fixed hood). Clearance problems for floor space relationship to the fixed hood; rugged drive mechanism needed for swing-away converter hood.	Space occupied in the aisle by the swing-away converter hood when adding matte, rabbling, or skimming could hamper crane movements. Crane must deposit the ladles for pouring or skimming and at the completion of the operation must await retraction of the hood before engaging the ladle. Maintenance of the swing-away mechanism and minimal maintenance for removal of particulate buildup that occurs during matte additions and rabbling.	Dependent on operational cycle. When pouring, blowing or slagging the emission control would be good; when adding matte, or rabbling it would be similar to the fixed hood. Air motions influences efficiency in all operations.
Enclosed building	Requires careful consideration of all openings (personnel, truck, rail, materials) to minimize air motion. Roof monitor must handle all ventilation air for workers, emissions and in leakages. Increases building costs because of wind load design, tightness, and close-fitting openings.	All openings must be maintained constantly against excessive air infiltration. Tight siding and roofing required. Air circulation within the building for the workers and process must be carefully controlled. Intake and exhaust fans require preventive maintenance. Cleanup maintenance for settled particulates in the converter area is similar to that for a monitored system, either natural or powered.	Dependent on building tightness, air motion control, monitor exhaust capabilities.

- The movable hood in the retracted position is parked above the fixed hood. It has an independent track system with a five-speed, double-grooved hoist unit and slack cable limit switch. The movable head is 9 ft long and is elliptical, with a major axis of 18 ft 6 in. and minor axis of 7 ft 6 in. These dimensions provide a 3-1/3-in. clearance between the movable and fixed hoods. There are mating plates on the lower end of the fixed hood and on the top of the movable hood. The lower end of the movable hood is fitted with a 12-in. asbestos-type curtain that follows the elliptical perimeter to form a seal with the gate hood. The movable hood is constructed of 3/8-in. carbon steel with stiffeners of 7-in. channels.

- The gate hood is elliptical in cross section with a major axis of 16 ft 6 in. and a minor axis of 6 ft 6 in. It is 9 ft long. Clearance between the fixed hood and the gate is thus 3-1/2 in. The hood will be bolted to the gate. The plate is 3/8-in. carbon steel reinforced with 7-in. channels.

The dimensions listed above would be modified for each converter layout to provide the required clearances. Design considerations may dictate that the fixed hood is the largest unit, with the movable hood under it and the gate hood under the movable hood.

- If crane runway rail height is a problem, the smoke plenum of the secondary hooding duct system could be fitted in as shown. The plenum would span the primary uptake hood and would have a secondary hood dust bin affixed on each end. The dust bins would be equipped with pneumatic dust valves and discharge pipes. No provision is made in this study for removal of dust in the dust bins. The smoke plenum for this cost study is 4 ft by 4 ft 8 in. by 21 ft. It is constructed of 3/8-in. steel with 6-in. channel stiffeners.

- The secondary hooding duct system would have an uptake from each dust bin adjacent to the converter and then pass to its main ducting header for fugitive emissions. The damper valve shown would be adjacent to the main ducting header and would be closed when the converter is out of service. Existing facilities would determine the path of retrofit. The gases go to a dust bin ahead of the fans and from there to the breeching into the main converter gas duct and to the stack (Figure 10). For this study, the length of the main duct runs is 600 ft.

- The fans considered in this estimate are Buffalo Forge type 1320 BL, single inlet, arrangement 1, class 3 with 145 bhp, 785 rpm, 80,000 ft^3/min at 200°F. There would be one fan for each converter in the plant; as many fans as are required would be tied into the system.

- Support items for the system include piping, wiring, foundations, supports for ducting every 20 ft, expansion joints, miscellaneous platforms, and walkways. Valves, fans, dust bins, and similar items are flanged for ease of maintenance.

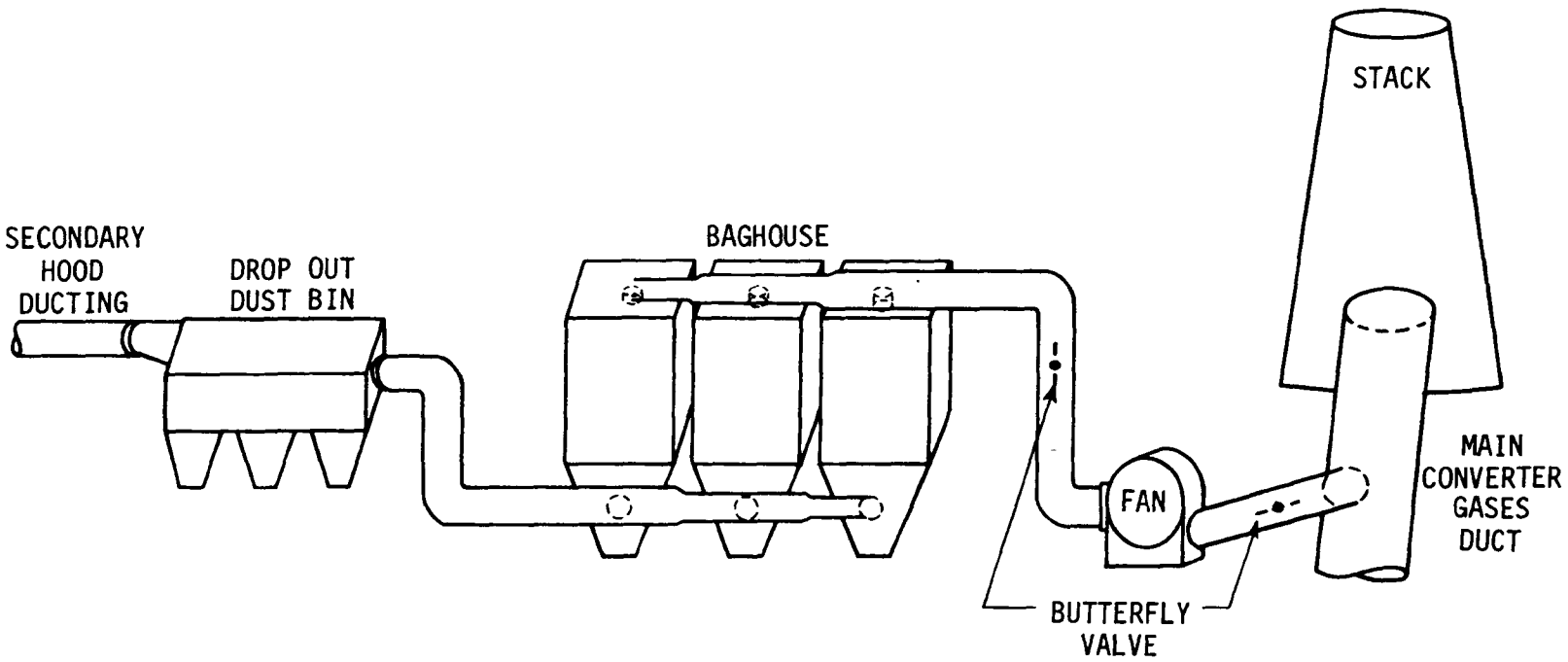


Figure 10. Fugitive dust ducting system (with baghouse) from converter building to stack.

Estimates of total installed costs are based on current (1978) costs for major components of specified sizes, as provided by equipment suppliers. Estimates of fabrication costs and installation charges are based on general accepted engineering practices.

A 5 percent contingency factor to allow for emissions testing, equipment changes, etc., is applied to the total of the direct and indirect costs. An escalation factor of 7-1/2 percent per year is used to account for increases in cost of equipment, labor, and services before and during construction. Direct capital costs include equipment, instrumentation, piping, electrical, structural, foundations, site work, insulation, and painting. Indirect capital costs include the following: engineering costs, contractor's fee and expenses, interest (accrued during construction on borrowed capital-estimated at 9 percent per year), freight, off-site expenditures, taxes (sales tax of 4 percent of equipment cost), start-up or shake-down, and spares.

Annualized costs include both operating costs and fixed capital charges and consist of: utilities; labor and fringe benefits, maintenance, plant overhead, and total fixed costs (amounting to 19.97 percent of total installed costs and consisting of depreciation over 15 years at 6.67 percent unless otherwise indicated, property insurance at 0.3 percent, property taxes at 4 percent, and interest on borrowed capital at 9 percent).

2. Capital Costs. Table 4 shows the direct cots, indirect costs including 1 year of contingency and escalation, and total capital costs for plants containing one to nine converters.

3. Operating Costs. Operating costs include the following: operating labor at $8 per man-hour, supervision at 15 percent of labor, maintenance for labor and supplies at 2 percent of total capital cost, maintenance materials at 15 percent of maintenance labor and supplies, electricity at 35 mills/kWh, plant overhead at 50 percent of operations, and payroll at 20 percent of the operating labor costs. The fixed costs include a straight-line 15 years depreciation, 0.3 percent for insurance, 4 percent for taxes, and 9 percent for capital costs. Table 5 lists the operating costs for a multiconverter plant.

Additional handling of slag and blister ladles may cause delays in operation of the movable and swing-away converter hoods. It is estimated that a delay of 5 to 15 seconds may occur with each ladle movement, equivalent to a total delay of 3 to 10 minutes per day or a 0.23 to 0.7 percent production slowdown. This loss is calculated on an annual basis as part of the operating cost since it is negligible in comparison with other delays that are encountered such as delay because of lack of matte or because the anode furnace cannot accept more blister copper or maintenance of converters or reverberatory or electric arc furnace, etc.

Each of these systems is connected to a main discharge duct and an exhauster type fan, which exhausts to a stack.

Relative capital costs of hooding for a five-converter smelter increase in the following order: fixed, fixed with movable, fixed with swing-away,

Table 4. ESTIMATED CAPITAL COSTS OF SECONDARY HOODING FOR MULTICONVERTER PLANT

(dollars)

No. of converters	Direct costs	Indirect costs	Total costs
1	760,000	532,000	1,292,000
2	1,216,000	785,000	2,001,000
3	1,532,000	963,000	2,495,000
4	1,771,000	1,255,000	3,026,000
5	2,211,000	1,463,000	3,674,000
6	3,219,000	2,122,000	5,341,000
7	3,601,000	2,383,000	5,984,000
8	3,880,000	2,545,000	6,425,000
9	4,402,000	2,850,000	7,252,000

Table 5. ESTIMATED ANNUAL OPERATING COSTS FOR SECONDARY HOODING IN A MULTICONVERTER PLANT

(dollars)

No. of converters	Labor supervision	Maintenance labor, supplies, and materials	Overhead plant and payroll	Utilities	Fixed costs	Total annual costs
1	10,000	30,000	22,000	1,000	258,000	321,000
2	21,000	46,000	38,000	3,000	400,000	508,000
3	31,000	57,000	50,000	4,000	498,000	640,000
4	41,000	70,000	64,000	5,000	604,000	784,000
5	51,000	85,000	78,000	6,000	734,000	954,000
6	62,000	124,000	105,000	8,000	1,077,000	1,376,000
7	72,000	138,000	119,000	9,000	1,195,000	1,533,000
8	82,000	148,000	131,000	10,000	1,283,000	1,654,000
9	93,000	167,000	149,000	11,000	1,448,000	1,868,000

fixed with movable and swing-away, and controlled building evacuation. Following are estimated costs to retrofit a smelter with five converters:

Fixed - $2.98 million

Fixed/swing-away - $3.04 million

Fixed/movable - $3.39 million

Fixed/movable/swing-away - $3.67 million

Enclosed building - $5.14 million

The enclosed building with baghouse, fans, and ducting is estimated at $11.78 million.

As shown in preceding tables, the costs of retrofitting fugitive emission control systems are high. The costs shown are for so-called "typical" smelters. Retrofit factors have been included in the costs and are estimated based on the "normal" problems encountered with retrofitting a converter shop that was not originally designed to have equipment installed. Some existing shops are so constricted that retrofitting ductwork that will not interfere with operations (i.e., crane, etc.) and is practically impossible. No smelter is really typical. They are all different and have different problems associated in retrofitting the equipment. Therefore, it must be pointed out that fugitive emission control costs must be analyzed on a site-specific basis and much thought given to all related operations near a hooding system before retrofit factors are assigned to a given plant.

F. CURRENT AND POSSIBLE FUTURE METHODS/PROCESSES FOR EMISSION CONTROL

The industry has remained the same in technology and design since 1909 with the reverberatory furnace and Peirce-Smith converter. Some methods have been utilized and many methods could possibly be adopted in order to control fugitive emissions in smelting and converting operations at a copper smelter. The ideas discussed here would involve design of equipment new to the copper industry, but not new to other metallurgical industries (i.e., steel industry).

- Charging floor technique
- Q-BOF type furnace
- Cascading system from calcined ore to matte to blister copper to anodes
- Covered ladles/stopper rods/autopour units
- Evacuated building with each furnace in its own enclosure

The following opinions have been formed with respect to control of fugitive emissions from personal observations of various copper smelters in the country:

Roasters - Nearly all of the multihearth roasters observed have observable particulate fugitive emissions and detectable sulfur dioxide emissions. As far as can be determined, these fugitive emissions have been greatly reduced by the use of fluid-bed roasters with cyclone collectors. These roasters appear to be operating with minimal problems. Besides nearly eliminating fugitives due to leakage, the concentrate is fed in a slurry form to the fluid-bed roaster, thus minimizing the escape of fugitives during concentrate transport. (Sulfur dioxide in the fluid-bed roaster gas stream, unlike multihearths, is rich enough to be sent to an acid plant. Thus, fluid-bed roasters, besides reducing fugitive emissions, also improve ambient air.)

Smelting furnaces - The reverberatory furnaces (reverbs) observed usually had leaks in the brickwork in which fugitive emissions could be seen. Sulfur dioxide emissions can usually be detected, particularly when standing at a location even with or above the reverb roof.

The electric furnaces observed have had a minimal amount of fugitives around them. The furnaces have had much tighter brickwork than the reverbs. At one smelter, no fugitive emissions could be seen coming from the electric furnace. At another smelter, only some emissions around the electrodes consisting of paste that was ignited at the furnace roof line could be seen.

The Noranda furnaces (reactors) observed showed no fugitive leakage during the smelting period.

Converting furnaces - The most common converter is the Peirce-Smith. All of these converters generate varying amounts of fugitive emissions, from minimal (barely visible) to considerable (dense cloud) depending on the mode of operations. Some Peirce-Smith converters emit minimal amounts of fugitives during blowing operations because the hooding system and the sliding gate form a close fit with the mouth of the converter. If wide gaps are present between the converter and the main hood and gate, or if leaks are present in the hooding system, then fugitives are heavy and are very noticeable in the converter aisle during blowing. The main point, however, is that fugitives can be minimal during slag or copper blowing if a converter has good hooding and good maintenance. The problem with all Peirce-Smith converters observed with respect to fugitive emissions occurs when the converters are "out of stack," meaning that the gate hood is up and the converter mouth is rotated out from under the hood (i.e., receiving a charge or is pouring slag or blister copper, etc.). When a converter containing matte is rolled out, air is blown through the tuyeres (which are approximately 8 in. below the bath level during blowing) for a short time to prevent plugging as the tuyeres emerge; also when the matte is added to the converter, air is blown through the tuyers for a short time to prevent plugging. Fugitive emissions are usually great during this period. Fugitives continue to escape the main hooding system even when the blow air has stopped, but in lesser amounts. Secondary hooding can help capture some of these fugitive emissions. In summary, fugitive emissions from Peirce-Smith converters can be controlled

effectively when they are in the blowing mode by means of tight hoods. When out of stack, however, only a portion of the fugitives from a Peirce-Smith can be controlled by secondary hooding. One smelter has a modified enclosed converter building in which 60 percent of the roof truss area is under control of a fugitive exhaust system to a baghouse. This system will not be commented on because it still is being modified and is not a totally enclosed and controlled evacuation type.

Escape of fugitive emissions could be minimal from the Peirce-Smith converter if it could be charged from a ground-operated machine (discussed later) rather than by an overhead-type crane. Secondary hooding would be effective if it could be kept in position directly over the fugitive sources. Retrofitted secondary hoods, now at some smelters, must be positioned to be clear of the crane operations for charging the converter, etc. This makes fugitive emission control at Peirce-Smith converters a difficult problem with the current building design.

Hoboken converters - It may not be fair to compare Hobokens and Peirce-Smith converters (with secondary hooding) on an equal basis. The Hoboken unit is designed to operate with a zero or slight negative pressure. The Hoboken-Overpelt unit in Belgium operates with little or no emissions during blowing operations. Some fugitive emissions are released during tapping, slagging, and charging operations at the ladle. A telescopic secondary hood operating in conjunction with a Hoboken converter would eliminate emissions during slagging and tapping blister copper and greatly reduce the small amounts of emissions during charging.

A Peirce-Smith converter with retrofitted secondary hooding cannot really be compared with new equipment designed to eliminate fugitive emissions. A better comparison would be the installation of a Peirce-Smith converter in a new shop which would be designed with hoods specifically to capture fugitive emissions.

The Hoboken converter can control fugitive emissions much more readily than the Peirce-Smith type.

Other observed equipment currently in use to control fugitives are as follows:

Hoods over matte and reverb slag tapping holes can be very effective. Hoods over a filling slag pot can also control emissions to the point where none can be observed. Swing-away hoods that can be lowered over ladles receiving matte can be effective. The hoods observed appear to pull most of the fugitives effectively, although some escape the outer periphery of the ladle or hood because of the large clearance between the hood and the top of the ladle.

Even newly designed shops installing secondary ductwork on a Peirce-Smith converter that would minimize fugitives and not be in the way of the overhead cranes etc., create many design problems.

A possible answer to the Peirce-Smith fugitive emission problem would be a charging floor technique. A charging machine(s), illustrated in Figure 11, could replace the overhead cranes that transport ladles of matte or slag. A charging machine, for example, could lift a ladle of matte, move back and pivot around 180 degrees, then move to the converter. The arms would lift the ladle to the desired height and tilt the ladle for discharging the matte into the converter. The converter would be contained or encapsulated with a hood to capture the fugitive emissions. A portion of the hood for the converter would be retractable during the times an overhead crane is used for maintenance. A Hoboken converter without a main hood would be better than a Peirce-Smith converter in this mode of operation.

With the elimination of overhead cranes for production in the converting aisle, the proper ducting could be installed above the ladle and mouth area. The ducting could be located in positions most suitable for when the converter would be in the rolled out position. A charging machine would perform all of the duties of overhead cranes, except for maintenance.

Whether using overhead cranes or charging machines, the transport of open ladles from a matte-producing furnace to a blister-producing furnace means escape of emissions from the ladles. Fugitive emissions during ladle transport are not a major source, but they do add to the fugitives problem. Because the ladles are often near the converter aisle floor, the fugitives that are generated are near the operations people. A possible method of reducing these "ladle" emissions would be to have covers on the ladles that could be placed on or taken off with a minimum amount of time and trouble. (This may be difficult in existing shops.) In conjunction with covers, specially designed bottom-pour ladles with stopper rods (Figure 12) could possibly be used at smelters for molten metal transport. Such a ladle with an autopour unit is used in the steel industry where it is attached to the operation of the stopper rod mechanism (Figure 12). This would allow the crane operator to control the flow of matte directly into a converter or blister copper into an anode furnace. A bottom-pour ladle could even be seated on a ladle support, and the ladle tapped to a specially designed opening in the end of a converter through a refractory-lined air tight joint/cover. (In this latter case, the mouth of the converter could be made smaller, if so desired, and would be used solely for pouring and not receiving hot metal charge.)

The electric overhead traveling crane (EOT) could be adapted to minimize fugitive emissions from ladles in transport, ladles being filled, or ladles pouring molten metal into a converter or anode furnace, by using a capture hood fixed to the spreader beam (Figure 13). The capture hood would be fixed to a sectionalized telescopic column (similar to that used in the steel industry for handling ingots going to and returning from a soaking pit). The telescopic column would be attached to the trolley, where a transfer duct would discharge to a fixed split rubber covered duct fixed to the EOT's girder. This duct would discharge to another split rubber covered duct adjacent to the building columns on the crane runway at the walkway level or at the roof truss line. This second duct would discharge to a baghouse. An I.D. fan, positioned on the trolley, walkway, or girder, would supply the required capture velocity. Another technique would be to discharge from the trolley to the roof truss and use the building monitor for discharging to a baghouse.

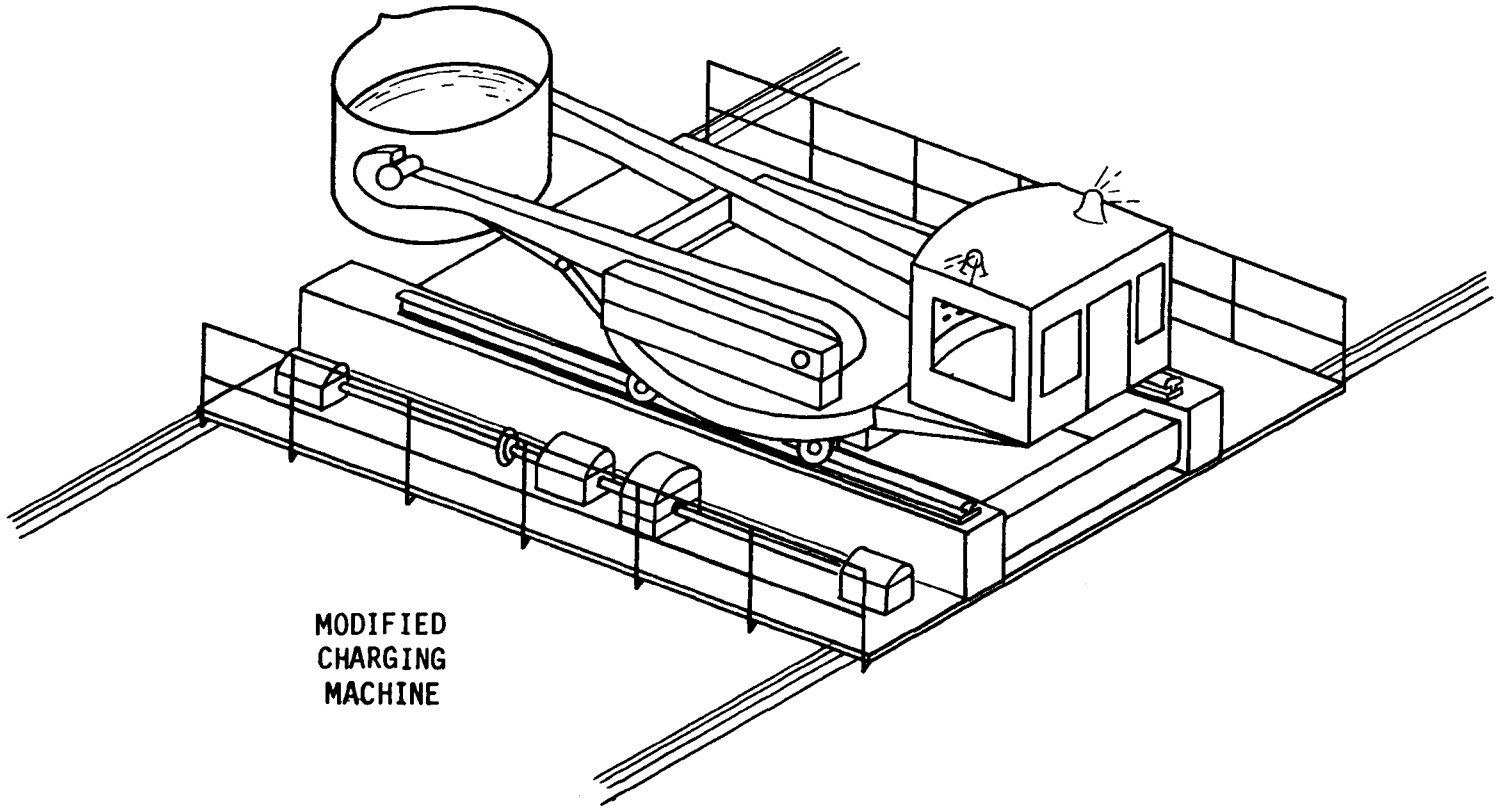


Figure 11. Modified charging machine.

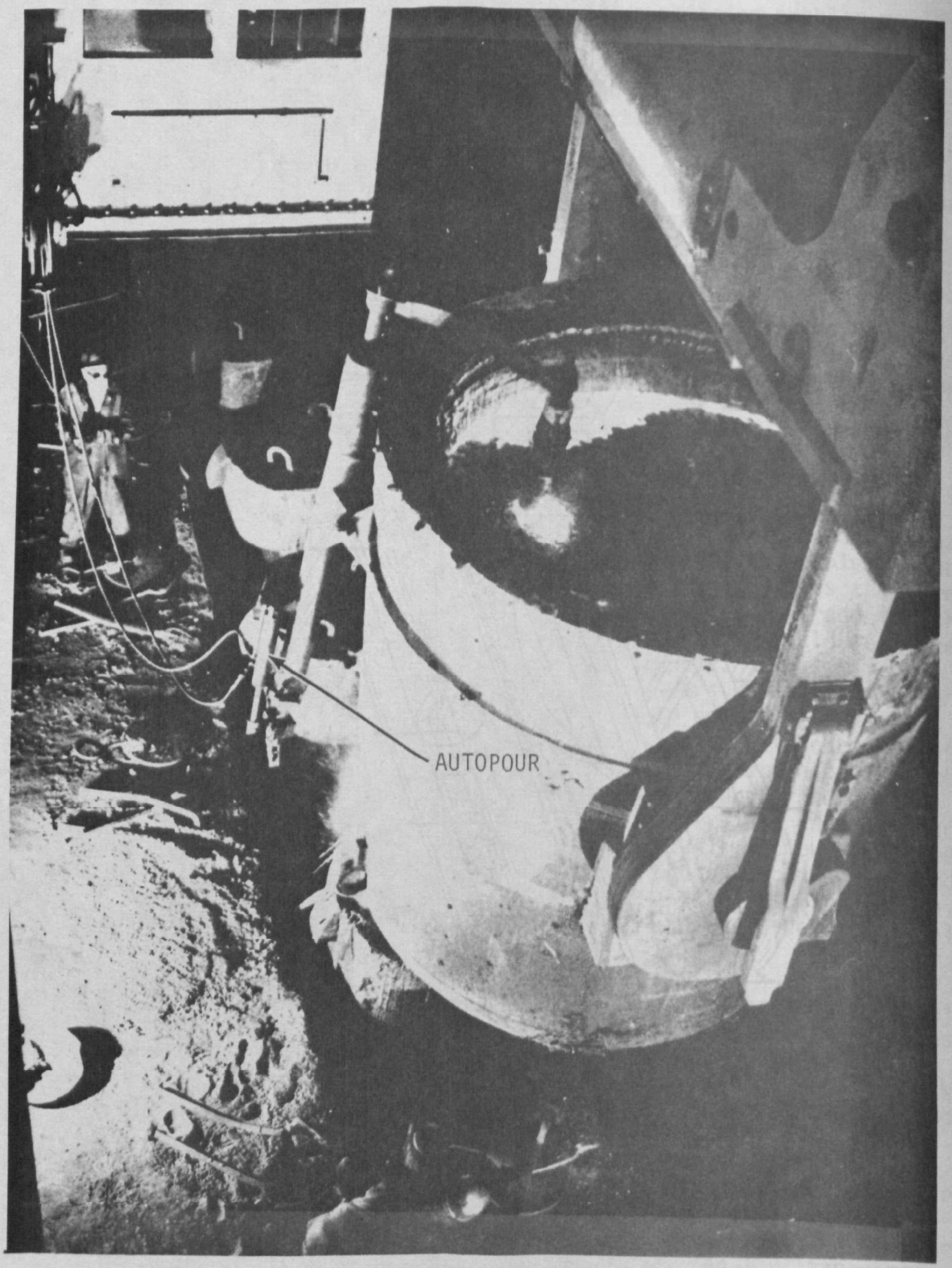


Figure 12. Hydraulic cylinder mounted on barrel of ladle rigging raises and lowers stopper rod to control flow of molten steel from ladle to ingot mold.

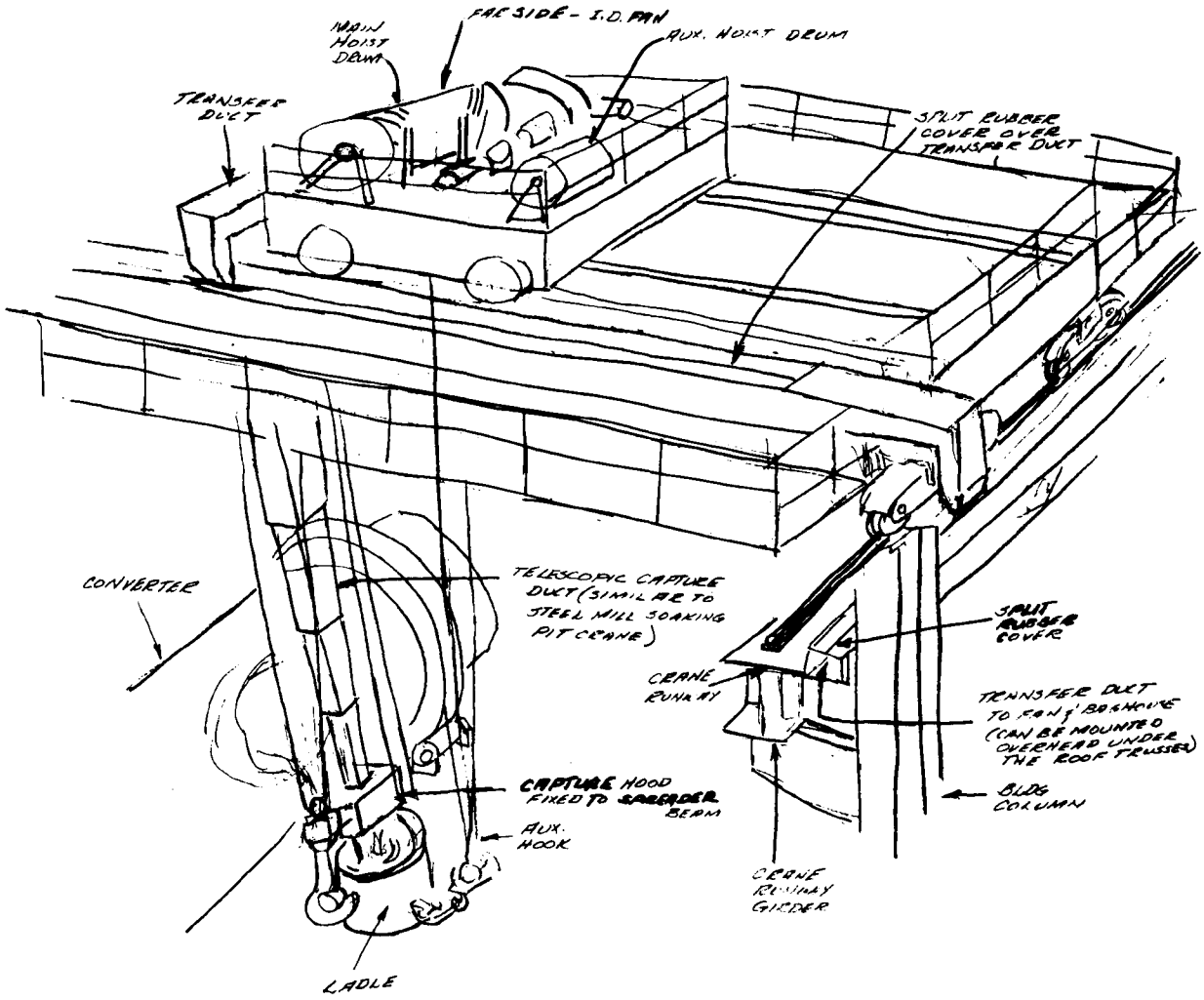


Figure 13. Sketch of E.O.T. crane with capture hood.

Another fugitive-emission control scheme could be an evacuated building with each furnace in its own enclosure with an independent pendant-operated crane for handling ladles, etc., within the enclosure. Ladles would be removed or brought into the enclosure through a movable rail-mounted car that would pass through a sliding door. Emissions would be exhausted through a duct system to a central point.

In summary, as far as fugitive emission control is concerned, there are methods currently used that are reducing fugitives, but many ideas need to be developed and good ideas tested so that fugitive emissions can be eliminated in a feasible manner. Elimination of fugitives would improve working conditions in a smelter. Besides that, new ideas and equipment that have been implemented in industry to solve a problem have often lead to a technical breakthrough that was not originally foreseen.

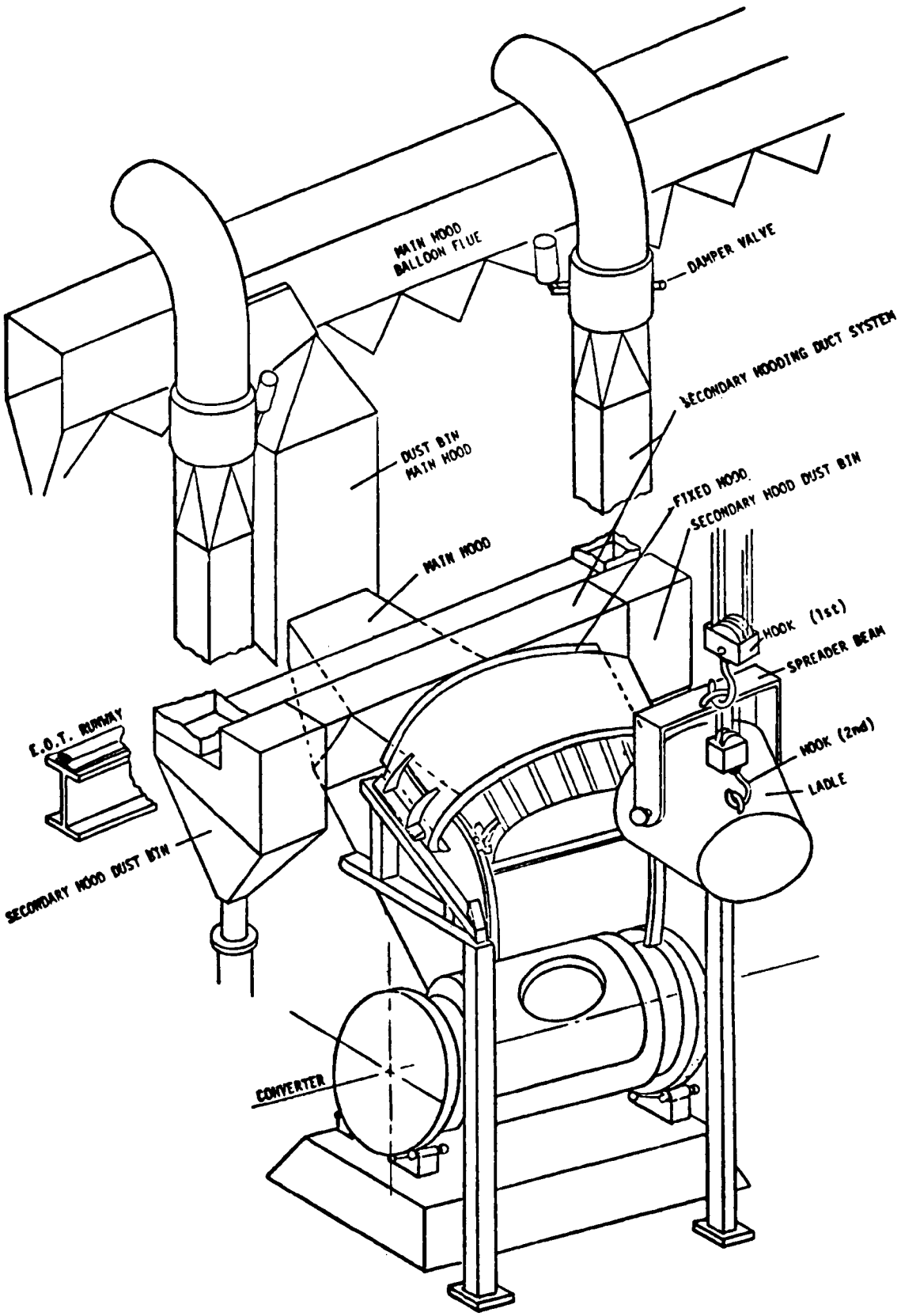


Figure A-1. Matte charging operation.

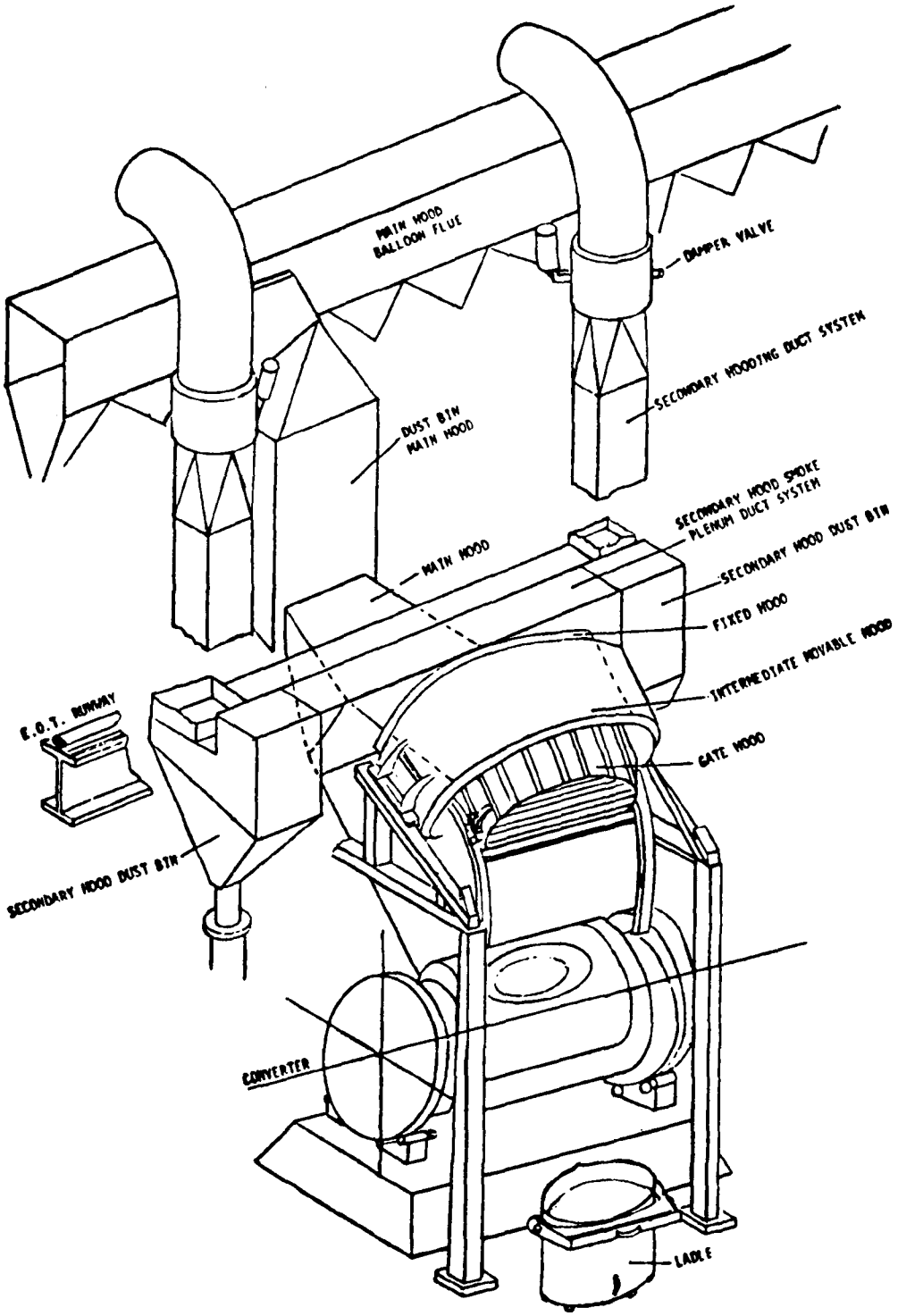


Figure A-2. Peirce-Smith converter - retracted hooding, pictorial view.

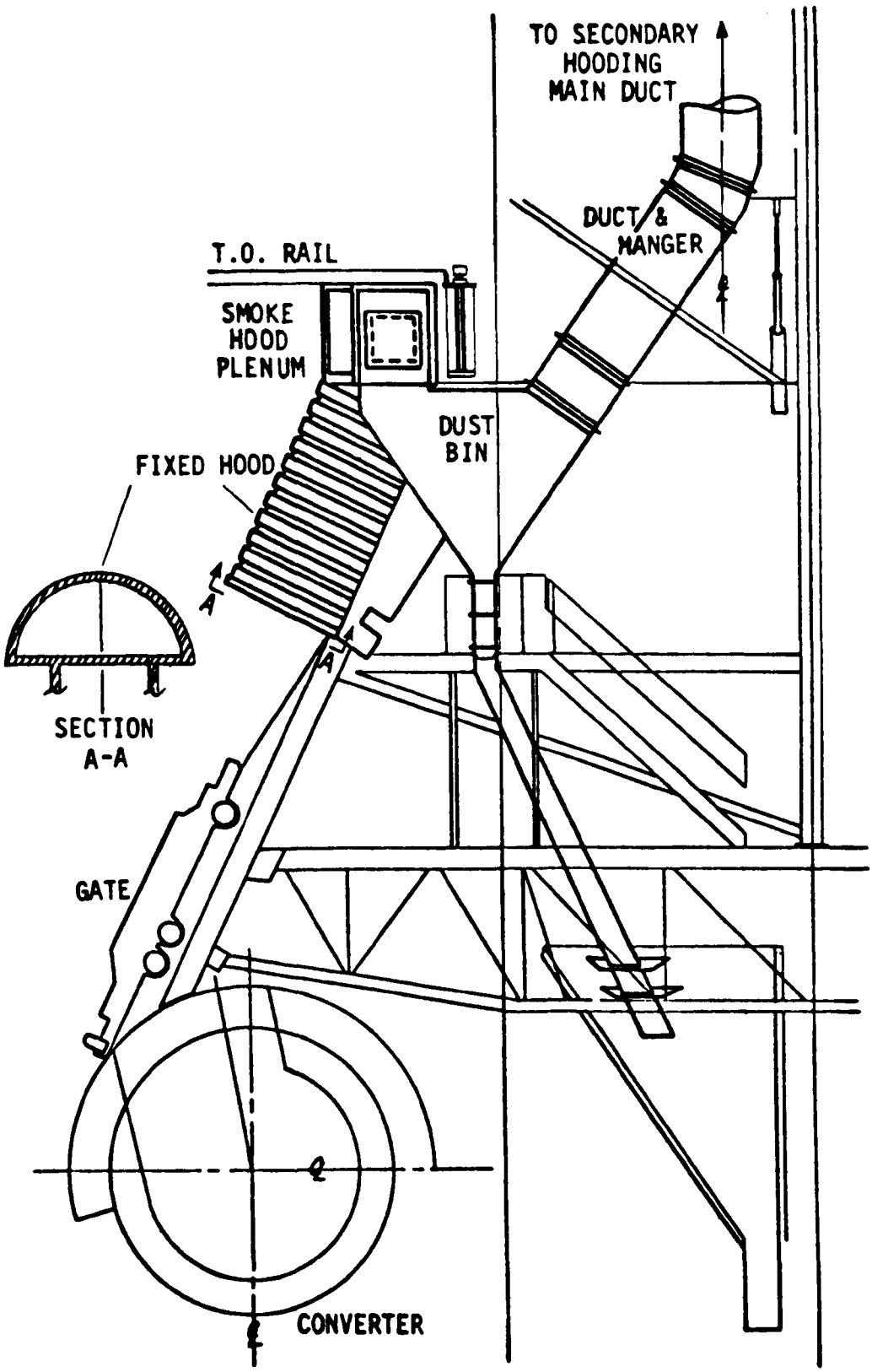


Figure A-3. Secondary converter hood configuration.

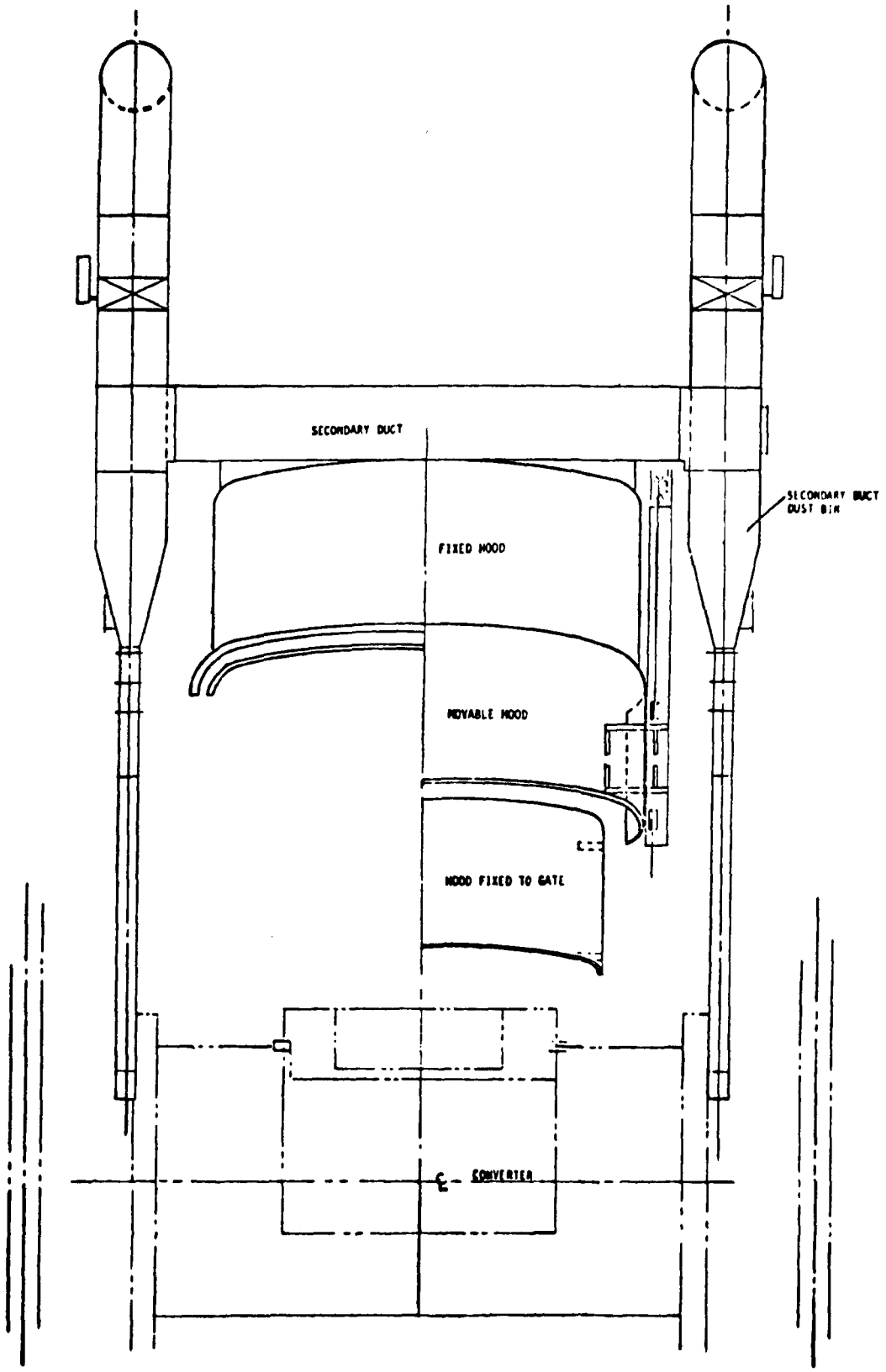


Figure A-4. Fixed, movable and gate hoods - front view.

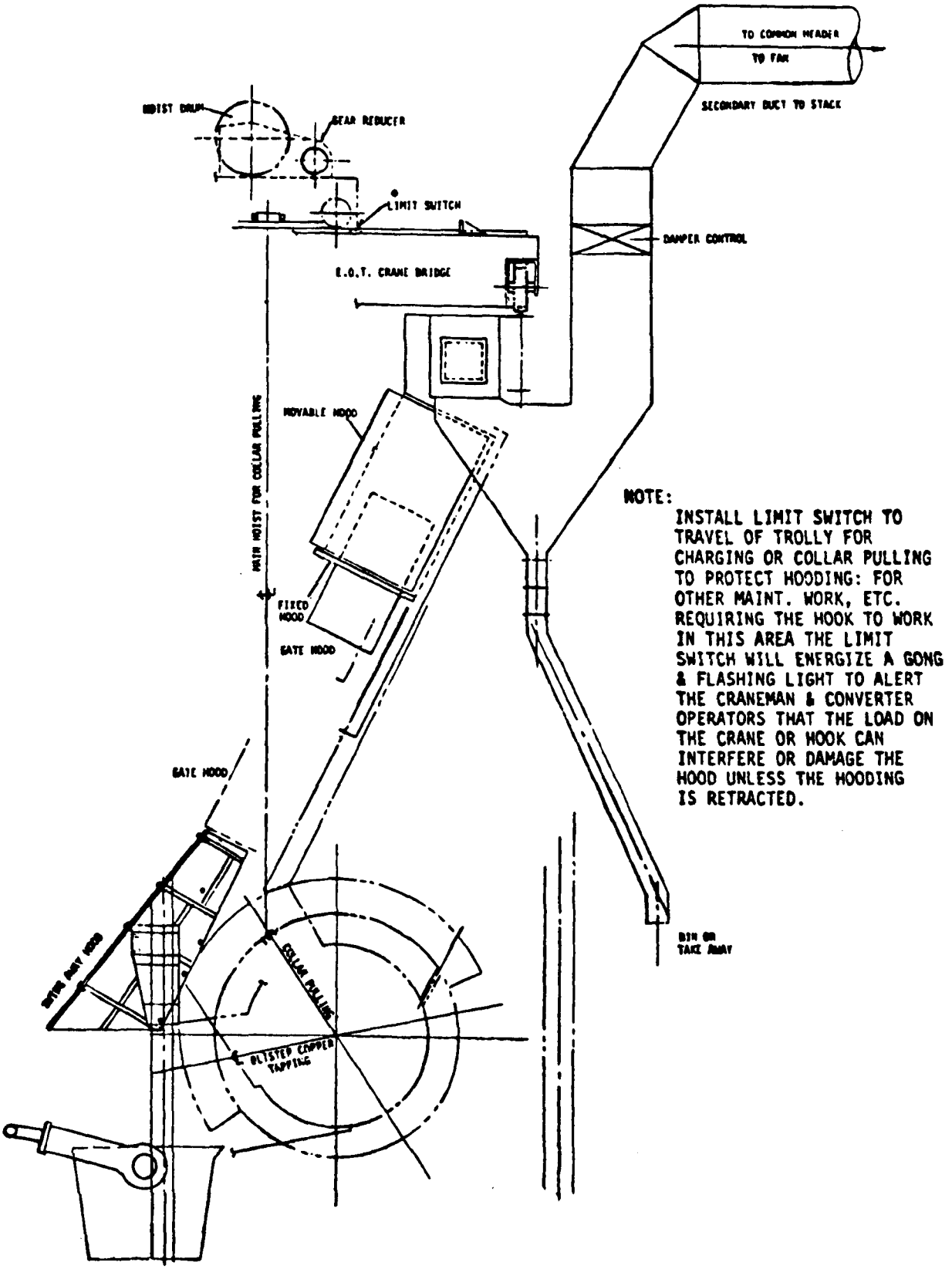


Figure A-5. Peirce-Smith converter - side view, blister pouring operation, hooding extended.

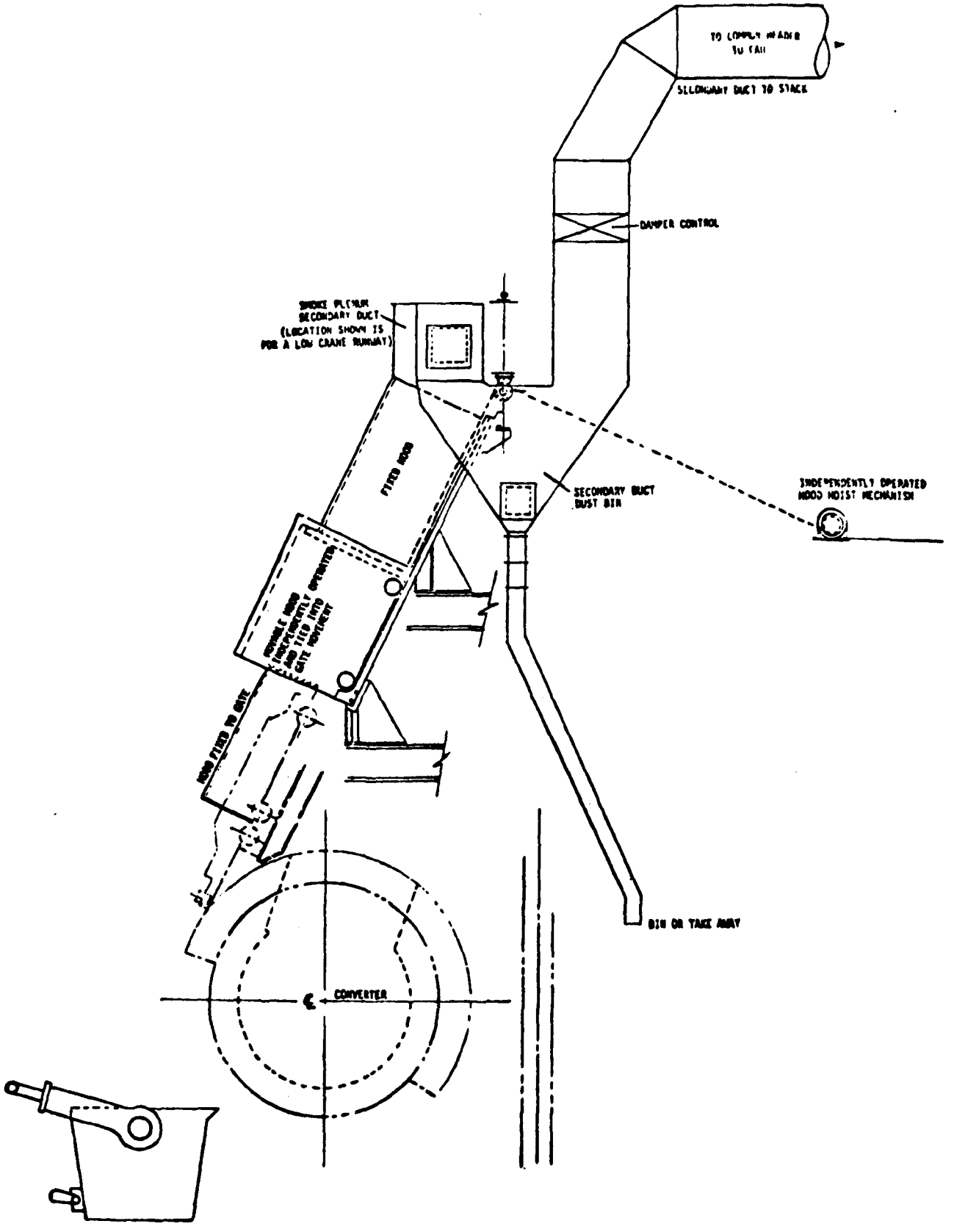


Figure A-6. Peirce-Smith converter - side view, hooding in position.

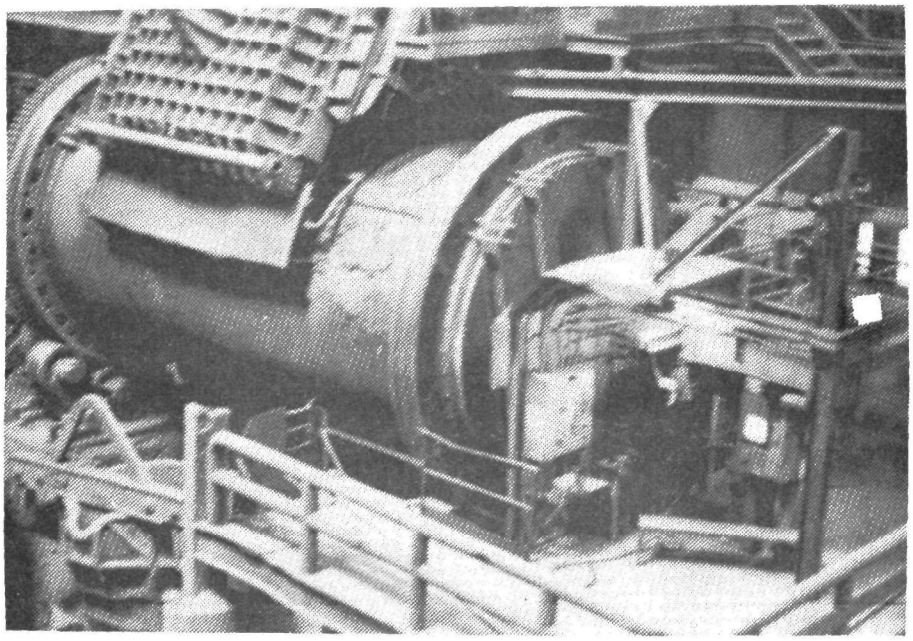


Photo A: Morenci - converter with gate (upper left) and apron (center left), converter operating platform (lower right), and typical ladle with lifting bale (lower left).

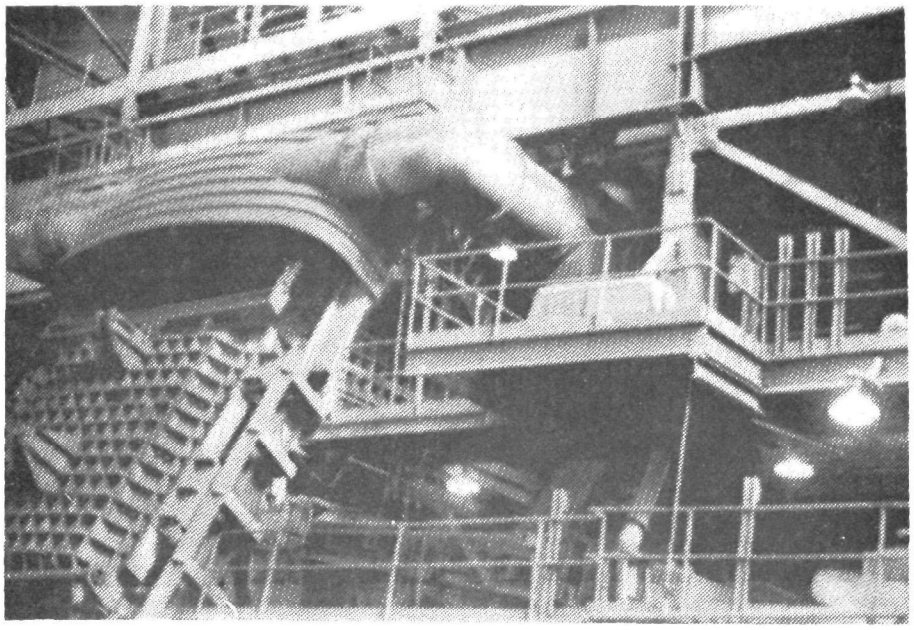


Photo B: Morenci - fixed hood, smoke plenum and duct (upper center and left), and gate (lower left).

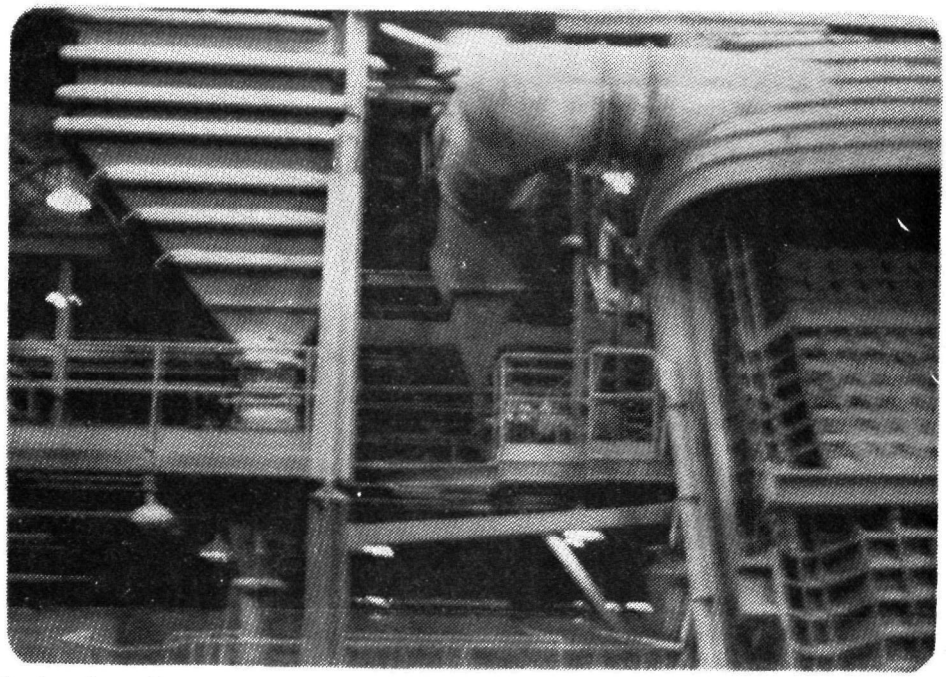


Photo C: Morenci - fixed hood, smoke plenum and duct to dust bin (upper right, upper and middle center), and gate (center and lower right).

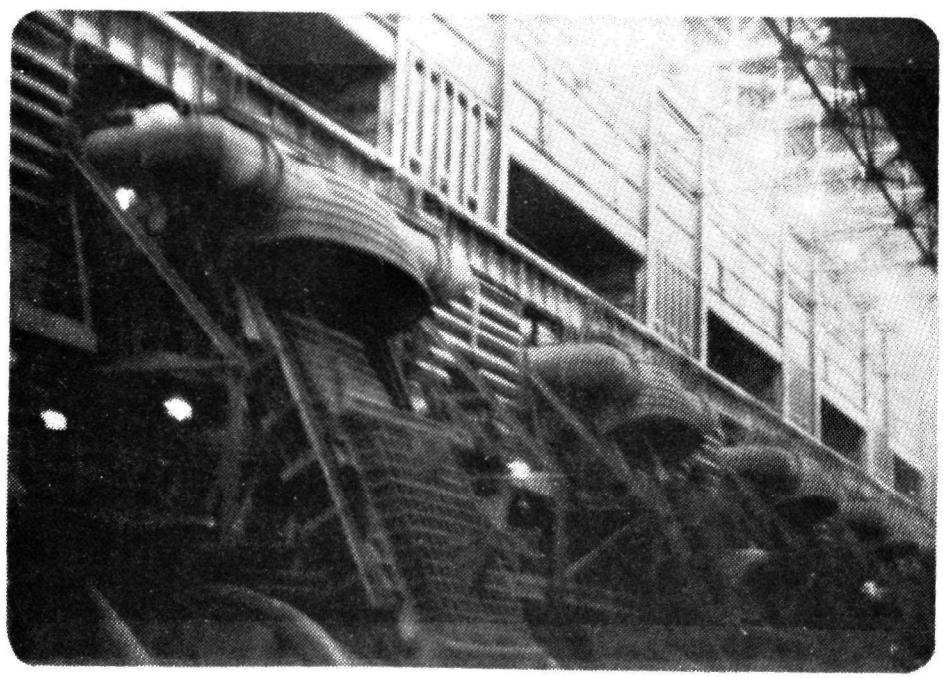


Photo D: Morenci - view of converter uptakes, fixed hood, smoke plenum, dust and gates.

Photo E: Morenci - view of converter aisle. Converters to the left and reverberatory furnaces are on the right.

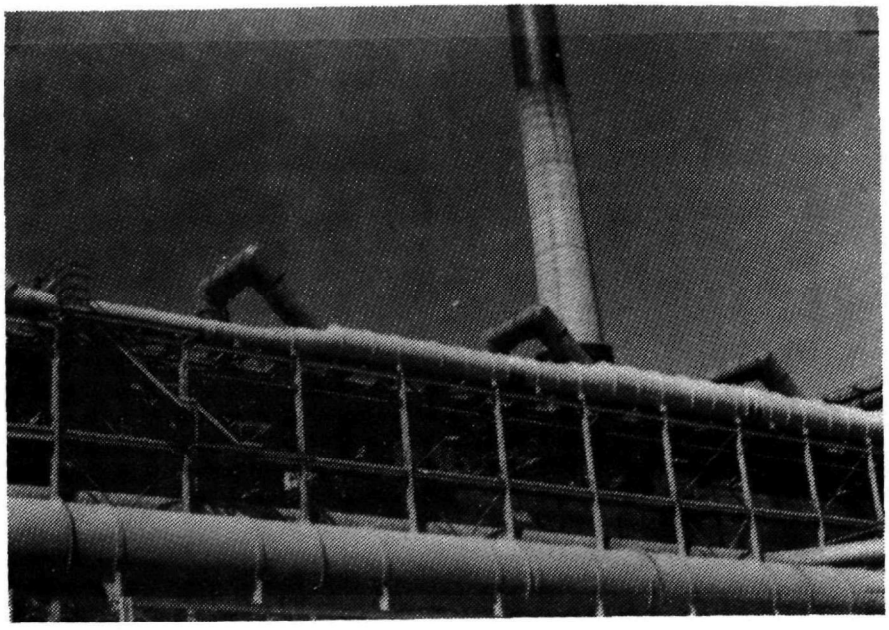


Photo F: Morenci - view of secondary hood ducting (center left to center right).

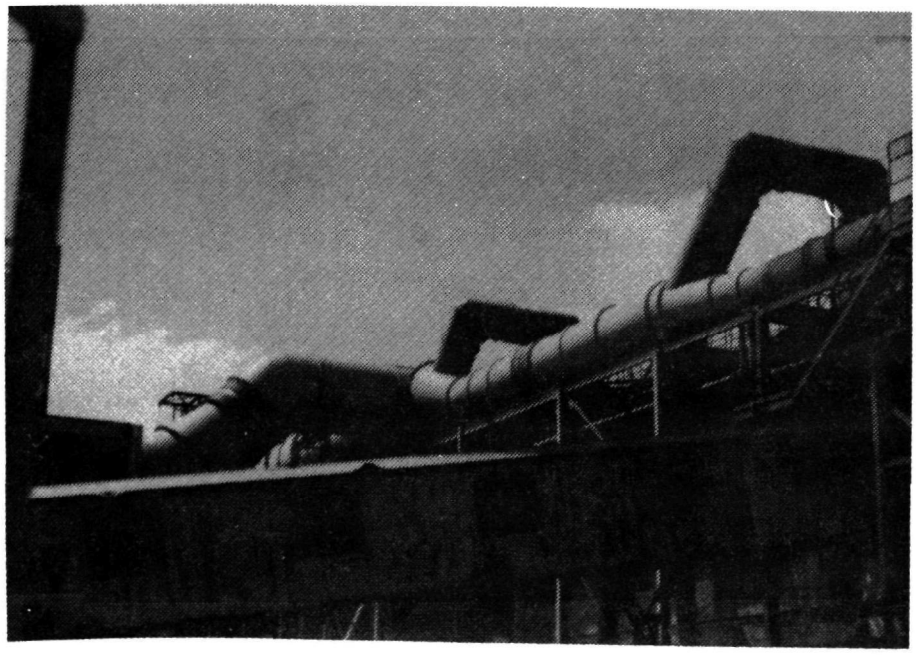


Photo G: Morenci - view of secondary hood ducting leading to the stack (center).

Photo H: Ajo - ladle (center) pouring matte into the converter by means of the overhead crane. Gate in raised position (upper center). Apron on converter is seen clearly.

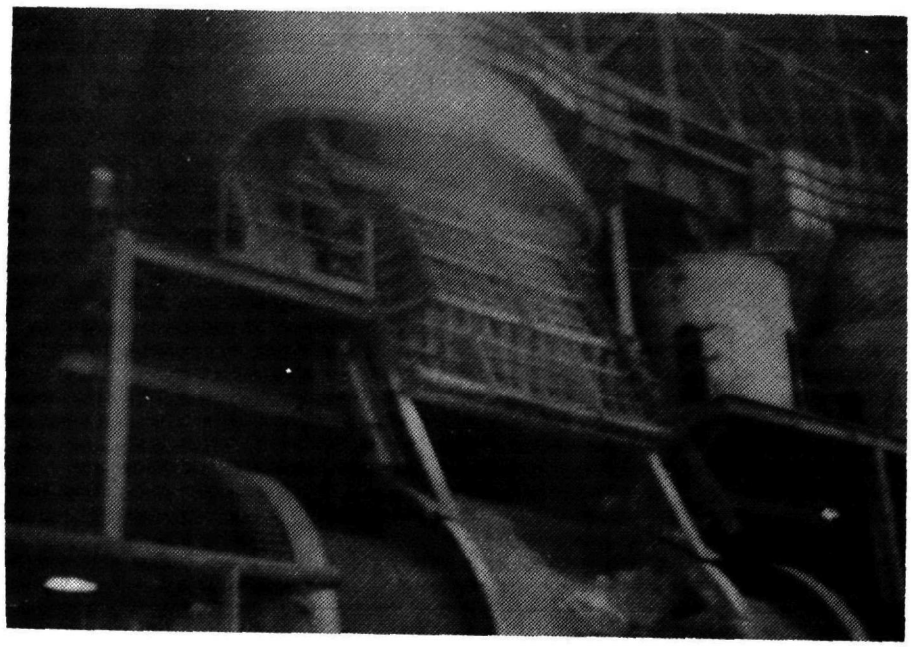


Photo I: Ajo - secondary emissions (at center) are visible with the gate in a raised position (center) and fixed hood (upper center).

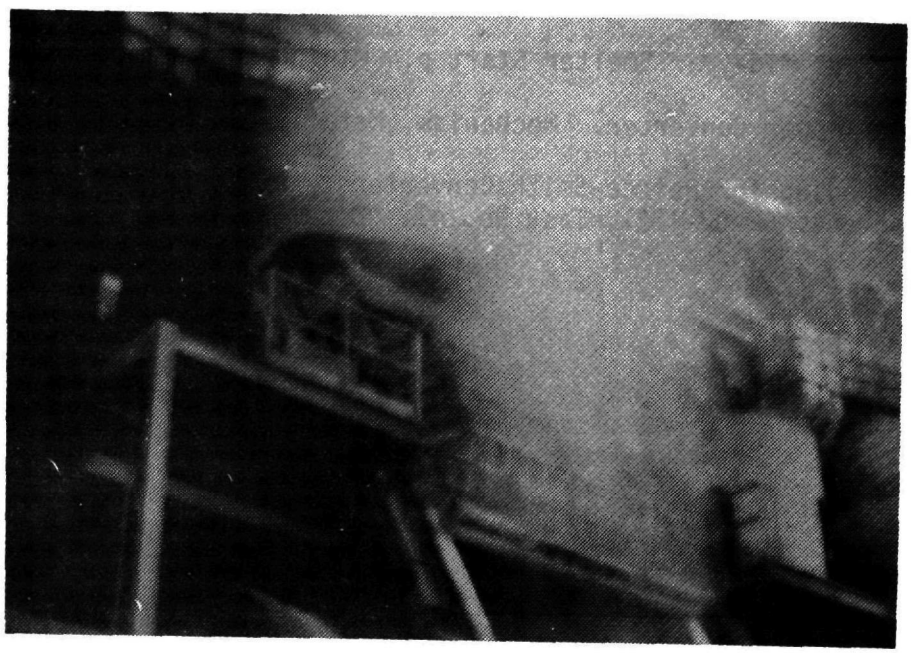


Photo J: Ajo - secondary emissions (at center) are visible with the gate in a raised position (upper center), the fixed hood, smoke plenum and dust bin (upper center and left).

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Core Room Emissions in Foundries

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ABSTRACT

Cores are the preformed sand inserts placed in foundry molds to produce the internal cavities in the castings used by our society. Traditionally the sand used for coremaking has been bonded with drying oils that were thermally set by the heat of electric, coke, oil or gas-fired ovens. Thirty years ago synthetic thermosetting resins, including furan and phenolic-based products, were formulated for coremaking. More recently other chemical binders, including some that do not require baking, have been developed that give faster cure times. For the most part, technical developments in foundry binders have been independent of environmental considerations.

It is estimated that the nation's foundries annually produce 3.3×10^6 pounds of emissions during coremaking. These emissions range from simple hydrocarbons such as methane through unsaturated aliphatics, various solvents, the carbon oxides, and others most noted for their odors. These emissions have two basic sources: The binder (and its subsequent polymerization reaction products) and the oven emissions associated with fuel combustion.

Recent research has developed a new binder system based on a starch product that satisfactorily bonds core sands which is inherently less prone to produce objectionable emissions. These low emissions levels have been demonstrated in the laboratory. The binder requires a drying step to develop maximum strength. Microwave energy has sucessfully been used with this system, effectively eliminating oven emissions at the foundry.

CORE ROOM EMISSIONS IN FOUNDRIES

The casting process is the basis of all manufacture involving metal. In the casting process, molten metal is poured into a mold which contains a cavity with the desired shape of the final product. The metal solidifies and cools in this configuration. Foundry molds are typically made from sand and have two mated parts: the cope (the upper half) and drag (the lower half). Sand molds are prepared by ramming or blowing sand around a pattern. Binders are added to the sand prior to molding so that the patterns may be withdrawn to leave the desired mold cavity. These binders may be as simple as the wet clay in green sand molds or as complex as organic resins.

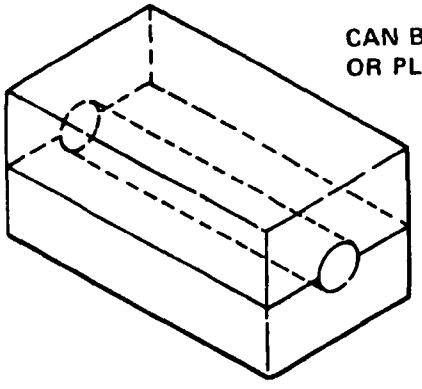
When the configuration of the casting requires internal voids or hidden cavities, it is most economical to cast them in place. Preformed sand inserts, called cores, are used for this purpose. A core must be rigid and self-supporting, and therefore the binding agent must give it more strength than is usually provided by mold binders. Traditionally, the sand is mixed with a drying oil which polymerizes or cures, to bond the sand when heated.

Cores are prepared in a core box, such as illustrated in Figure 1. Cores are often made with the aid of mechanical equipment such as squeeze molders, core blowers, or core shooters, which inject the sand mixture quickly and automatically into the core boxes. The boxes are stripped away from the sand and the core is laid on a flat or contoured support, called a core drier, which prevents deformation during the oven baking cycle.

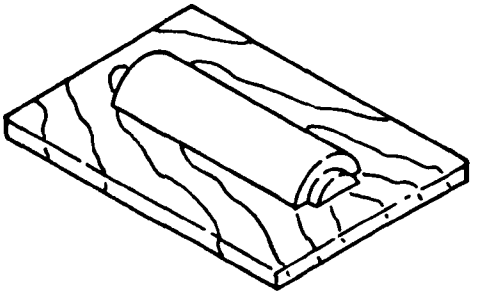
One of the most significant advances in foundry technology in the last thirty years has been the development of new binders for sand molds and cores. These binders make it practical to design more complicated cores and they also allow the foundryman to achieve better dimensional accuracy in the cast product. Table I lists some of these processes and the binders employed. The development and description of specific binders for these processes have recently been reviewed. (1,2)*

In general, binder development has been directed toward the use of complex organic resins, and the use of catalysts that allow curing without heat. Many of these "no-bake" binders were developed with no specific concern for their environmental impact. However, in current research and development of binders, more attention is being paid to reducing the level of emissions both in the mixing and curing, and during the pyrolysis of these binders. (3,4,5) Inorganic systems based on phosphates and silicates have been touted as solutions to environmental problems in the foundry.(2) Poor collapsibility of the cores has been reported, which makes core removal from the casting difficult and prevents their widespread use.(5,6) The addition of organic additives to these systems to improve collapsibility produces pyrolysis products of similar composition and magnitude to those from organic resins, (7,8), but the molding and mixing emissions are low.

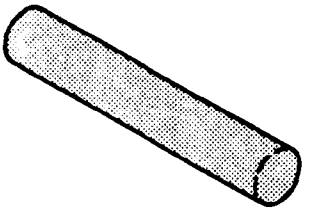
* Numbers in parentheses refer to items in "References."



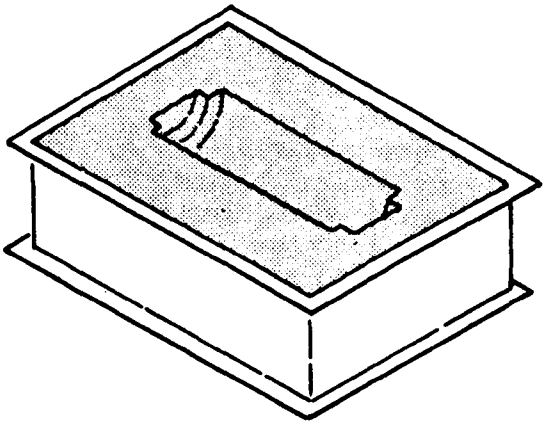
CORE BOX



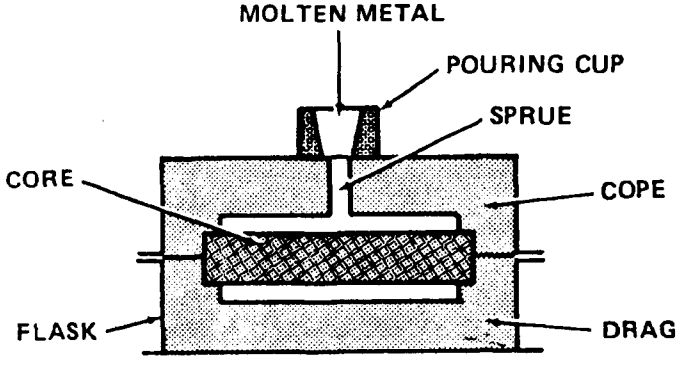
PATTERN

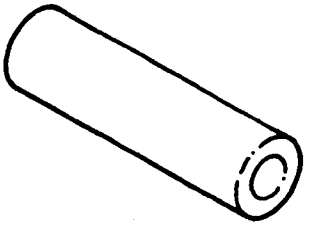


CORE



MOLD





FINISHED CASTING

Figure 1. CASTING NOMENCLATURE

TABLE I

Common Core Binders

Process	Binder
Oil	Drying oils
Shell	Phenolic Novalac Resins
Hot-Box	Phenolic or Furan Resins
No-Bake [Acid Cured]	Phenolic or Furan Resins
No-Bake	Urethane Resins
Cold-Box	Phenolic, Furan or Urethane Resins
Spirit	Carbohydrates (Starches)
Silicate	Sodium Silicate

The widely accepted organic binders that require heat for curing – specifically those for the shell, warm box, hot box and core oil processes – are declining in usage as shown in Figure 2. (9) Oil-sand mixtures must be heated in a core oven. These ovens may be heated by coal, oil, gas, or electricity. Oven designs have been proposed which heat cores with hot exhaust gases from metal melting furnaces. Whatever the fuel, core ovens require extensive floor space. Vertical ovens which allow sufficient time at temperature (dwell time) to cure the core binder are available which conserve floor space.

Recently, core ovens have been developed which employ microwave energy to cure the binder in a more controlled manner. Design requirements are radically different for this type of oven, in that wave reflection and shielding rather than insulation and burner design are critical to the oven's efficiency. (10,11,12) These ovens are particulary effective in drying core coatings.

Core Room Emissions

There are two sources of emissions in the core room: the emissions from the core ovens and those released during the curing of the sand binders.

The area where the cores are made is often separated from the remainder of the foundry, sometimes even detached from the foundry building. Core rooms are ventilated to remove the chemical emissions, and particularly objectionable odors, from the work area. However, these emissions are usually exhausted directly to the atmosphere outside the foundry, and may contribute significantly to foundry fugitive emissions.

Foundries using thermally-cured resins with core ovens can control in-plant emissions if the ovens are properly vented, but unless they are equipped with afterburners or scrubbers, pollutants will still be discharged to the atmosphere outside the plant. Such controls are rare. Table II lists the emissions expected during the mixing, molding, and curing of cores prepared with thermal setting binders.

Another approach has been to use "no-bake" binders, which require no heat to cure, but may still release objectionable emissions as indicated in Table III.

Some core binder systems inherently release objectionable emissions during curing. Hexamethylamine (Hexa) is added to "shell" sand to provide formaldehyde to cure the phenolic resole resin, but it also introduces ammonia into the atmosphere. (14,15) The urethane-base cold-box process uses odorous tertiary amines such as dimethylethylamine (DMEA) as a catylst, which must be controlled with a phosphoric acid scrubber. (16,17,18) Furan-phenol formaldehyde binders employing gaseous sulfur dioxide as a catalyst are also being marketed as cold-box systems. (19) For both of these processes, the catalytic gases must be contained and scrubbed to avoid contamination of the work area. (18,19) These controls also are effective in reducing fugitive emissions or these catalysts.

As shown in Tables II and III, hydrocarbons are emitted during the curing of the binders. The most common hydrocarbon emissions are solvents, which are added to the binders to lower the viscosity. Some of these are aromatic

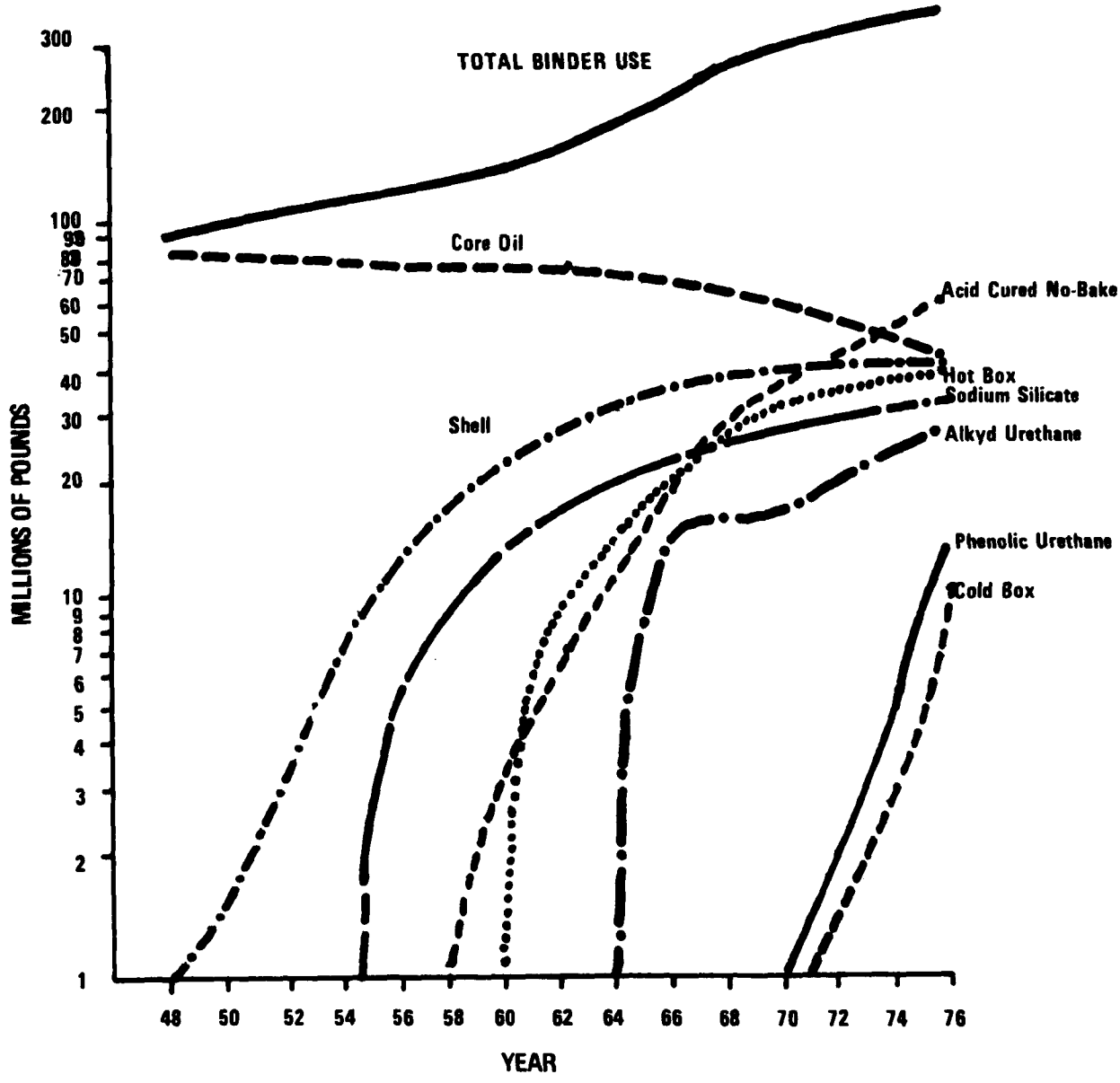


Figure 2. ANNUAL BINDER CONSUMPTION (US).

TABLE II

AIRBORNE EMISSIONS FROM CHEMICALLY BONDED THERMO-SETTING SYSTEMS DURING MIXING, MOLDING AND COREMAKING (13)

	CHEMICAL SPECIES											
	Carbon Monoxide	Formaldhyde	Hydrogen Cyanide	Ammonia	MID-Isocyanates	Sulfur Dioxide	Hydrogen Sulfide	Phenols	Benzene	Toluene	Furfuryl Alcohol	Methanol
CORE OIL	?	0	0	0	0	0	0	?	?	?	0	0
FURAN HOT-BOX	?	?	0	0	0	0	0	0	?	?	?	?
PHENOLIC HOT-BOX	?	X	0	0	0	0	0	?	?	?	?	?
SHELL	?	X	?	X	0	0	0	?	?	?	0	0
CARBOHYDRATE	?	?	0	0	0	0	0	0	0	0	0	?

ASUMES NORMAL VENTILATION, OPTIMIZED BINDER USAGE AND PROPER HANDLING OF BINDER SYSTEM.

0 NOT EXPECTED TO BE PRESENT IN SUFFICIENT QUANTITIES TO BE CONSIDERED A HEALTH HAZARD

? POSSIBLY PRESENT IN WORKING ENVIRONMENT DEPENDING UPON SPECIFIC FORMULATION AND SAND QUALITY

X PRESENT IN SUFFICIENT QUANTITIES TO BE CONSIDERED A POSSIBLE HEALTH HAZARD

TABLE III

AIRBORNE EMISSIONS FROM CHEMICALLY BONDED "NO-BAKE" SYSTEMS DURING MIXING, MOLDING & COREMAKING (13)

	CHEMICAL SPECIES												
	Carbon Monoxide	Formaldhyde	Hydrogen Cyanide	Ammonia	Aromatic Amines	MDI-Isocynates	Sulfur Dioxide	Hydrogen Sulfide	Phenols	Benzene	Toluene	Furfuryl Alcohol	Methanol
FURAN NO-BAKE (H_3PO_4)	0	?	0	0	0	0	0	0	?	0	0	?	0
FURAN NO-BAKE (TSA)	0	?	0	0	0	0	0	?	?	?	?	?	?
PHENOLIC NO-BAKE (TSA)	0	X	0	0	0	0	?	?	X	?	?	0	?
ALKYD URETHANE	0	0	0	0	?	0	0	0	0	0	0	0	0
PHENOLIC URETHANE	0	?	0	0	?	0	0	0	?	0	?	0	0
SODIUM SILICATE (CO_2 OR ESTER)	0	0	0	0	0	0	0	0	0	0	0	0	0

ASSUMES NORMAL VENTILATION, OPTIMIZED BINDER USAGE AND PROPER HANDLING OF BINDER SYSTEM

0 NOT EXPECTED TO BE PRESENT IN SUFFICIENT QUANTITIES TO BE CONSIDERED A HEALTH HAZARD

? POSSIBLY PRESENT IN WORKING ENVIRONMENT - DEPENDING UPON SPECIFIC FORMULATION AND SAND QUALITY

X PRESENT IN SUFFICIENT QUANTITIES TO BE CONSIDERED A POSSIBLE HEALTH HAZARD

compounds, which are readily released into the atmosphere. (1) Thermally cured binders are likely to give off methane and aliphatic hydrocarbons. In core rooms with inadequate ventilation, an oil mist is sometimes observed in the area. Some CO may be produced when curing thermal setting binders, especially if coated sand is exposed to higher temperatures. Traces of other organic compounds in the core room atmosphere, such as amines, can be objectionable because of their strong odor. (20)

The other source of core room emissions is the oven. Depending upon the oven type and configuration, there are combustion products from the fuel. Natural gas or propane is preferred for a number of applications, including the hot-box process and the shell process which was heated core boxes, because the combustion/temperature is readily controlled. Drying ovens with natural gas are also available, but with increasing industrial unavailability of this fuel, more conversions are seen to either oil or coke. In any case, conventional oven emissions such as methane, carbon monoxide, carbon dioxide, and water will be produced. More complex species may be formed, depending upon oven efficiency. Often all these combustion products are usually ventilated directly to the outside.

The use of electric heat essentially defers emission of these combustion products to a power plant, where control may be better effected. A comparison of oven emissions during core baking between an electric oven and a conventionally fired oven will define the proportions of the emissions due to the combustion of the fuel and that produced by binder decomposition.

During the work on organic binder decomposition emissions, a promising new system based on carbohydrates was identified. It is marketed as a core oil substitute and expected to produce fewer emissions than the conventionally used core oils since it is in a water medium. This binder is based upon the controlled formation of starch polymers. (21)

The Spirit† binder system is based on starch. Starch is predominantly α1-4 anhydro glucose polymer with some α1-6 linkages. This polymer is quite large in its natural form. In the Spirit system, starch is chemically treated give a controlled distribution of lower molecular weights that are dispersible in water. Addition of aldehyde groups with at least two aldehyde groups on each molecule completes the formulation. At the time of use, an inorganic salt catalyst is added. This gives a resin system that is basically carbohydrate. Crosslinking is presumed to proceed through a reaction of the aldehydes with alcohol groups present in the carbohydrate to form hemiacetal and then acetal configuration. Theoretically, low temperature pyrolysis, such as drying temperatures in the range of 120-150°C, should result in principally water and carbon with possibly some small quantities of formaldehyde.

† registered Trade mark of Krause Milling Co., Milwaukee, WI

SPIRIT BINDER EMISSIONS

Experiments were designed to evaluate the emissions during the baking of cores bonded with the carbohydrate-based Spirit binder. Cores were baked in a conventional electric resistance oven and also in a microwave oven. Table IV reports the results of these experiments. Analysis of samples of the gas taken in the oven cavity showed that the carbon monoxide content was less than 25 ppm. The total hydrocarbon concentration was approximately 25 ppm, but the aromatic portion, as reflected in the concentrations of benzene, toluene, xylene, and naphthalene, was extremely low. Trace amounts of formaldehyde were found. As the binder contains no nitrogen in its formulation, nitrogen-containing compounds were not expected nor analyzed for. These emissions are relatively low, approximately of the same order of magnitude as for the sodium silicate process.

It is anticipated that a foundry would be more satisfied with the shakeout characteristics of the Spirt binder than with sodium silicate. The general surface finish of a gray iron automotive casting produced with a Spirit bonded core (no mold wash) was equivalent to that of an oil-bonded core with a refractory core wash, as illustrated in Figure 3. (22) Both of these are commercially acceptable.

Shot blasting and grinding, which produce unhealthy dust emissions, are often necessary to remove adhering sand from castings produced with sodium silicate; and internal cores such as in Figure 3 might be difficult to produce with sodium silicate bonded sand because of the problems with core removal.

Microwave Ovens

As the starch-based binder must be dried to cure, it is particularly well adapted to microwave curing. The literature contains several references to this new application of microwave energy.(12,21,23,24) Essentially the microwave oven offers the foundryman a controlled, emission-free oven that readily dries sand. The sand aggregate is only indirectly heated in the oven cavity, and therefore energy is not wasted. Currently the largest technical problem in the use of microwave ovens to produce foundry cores lies in the development of suitable materials for core boxes and drying racks. Ideally, such materials should be readily formed, transparent to microwave energy, solvent-resistant, and resistant to temperatures up to approximately 300°C. (20,24) Many materials, including aluminum, transite, and plastics of several types, have been tried, but at this time the optimum material has not been developed. Once this problem has been overcome, it is anticipated that many foundries will consider conversion and subsequently may reduce their core room emissions.

Pouring Emissions

Finally, it is important to note that the emissions from the core material during the pouring are only a small part of the total pouring emissions. The core is normally surrounded by metal, such that the effective sand-to-metal ratio is very low and the pyrolysis of the core binder is nearly complete. Table V shows the results of the evaluation of organic core materials on

Table IV

Average Oven Emissions From
Baking of Starch Bonded Molds

	Microwave Oven		Electric Resistance Oven	
	µg/ℓ	(ppm)	µg/ℓ	(ppm)
Carbon Monoxide	-	<25	-	<25
Total Hydrocarbons*	-	35	-	18
Benzene	0.05	(0.02)	0.05	(0.02)
Toluene	0.01	(<0.01)	0.09	(0.02)
M-Xylene	0.08	(0.02)	0.09	(0.02)
O-Xylene	<0.01	(<0.01)	<0.01	(<0.01)
Naphthalene	<0.01	(<0.01)	<0.01	(<0.01)
Formaldehyde	1.04	(0.8)	0.60	(0.5)
Total Aldehydes#	<0.5	-	1.5	-
Propanol	0.05	(0.02)	0.11	(0.04)
Ethanol/Methanol+	0.31	(0.23)	0.18	(0.14)

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* As methane
# As acetaldehyde
+ As ethanol

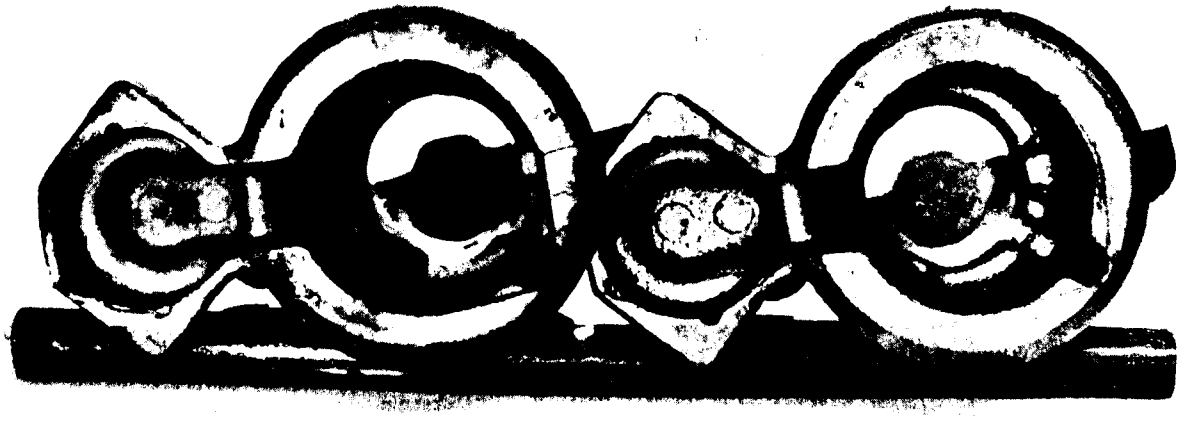

Figure 3. COMPARATIVE CASTINGS. Core for casting on left made with Spirit binder; Core on right made with core oil.

Table V

Results

Green Sand - Cored

Core Sand	Max. CO (ppm)	(Time)	Max. CO_2 (ppm)	(Time)	Max. CH_4 (ppm)	(Time)	Max. HC_T (ppm)	(Time) CH_4	NH_3 µg/l	HCN µg/l
Green sand	2400	(3 min)	18,100	(1 min)	680	(1 min)	980	(2 min)	1.0	0.63
Green sand	2400	(3 min)	11,400	(1 min)	540	(1 min)	1460	(6 min)	<0.20	0.91
Furan - TSA	1930	(1 min)	7,990	(1 min)	380	(1 min)	910	(3 min)	0.48	0.64
Furan - TSA	1770	(1 min)	6,750	(1 min)	430	(1 min)	840	(3 min)	<0.20	0.85
Alkyd isocyanate	1850	(1 min)	7,750	(1 min)	320	(1 min)	710	(10 min)	0.32	<0.21
Alkyd isocyanate	2070	(1 min)	5,540	(1 min)	460	(1 min)	860	(1 min)	0.45	2.70
Phenolic urethane	2220	(1 min)	11,500	(1 min)	240	(1 min)	510	(20 min)	0.21	<0.21
Phenolic urethane	2500	(1 min)	10,880	(1 min)	310	(4 min)	610	(2 min)	0.17	<0.21
Shell	2010	(5 min)	9,720	(1 min)	330	(5 min)	850	(2 min)	<0.20	-
Shell	1690	(3 min)	9,890	(1 min)	330	(4 min)	570	(7 min)	0.20	-
Phenol formaldehyde	1850	(5 min)	7,800	(1 min)	310	(5 min)	550	(5 min)	0.30	-

overall emissions during the pouring of a simple cylindrical casting, such as illustrated in Figure 1. These results clearly indicate that the molding sand decomposition (as for green sand) predominates emissions produced from thermal decomposition of the core. These emissions may also become fugitive emissions, (25) so it is important to bear in mind that whereas a new core binder, such as the carbohydrate-based Spirit binder, may reduce core room emissions, it may have little effect in the pouring area. The relative chemical simplicity of this binder indicates that a reduction in emissions in pouring and shakeout areas might be possible if the molds were bonded with it. Further development along these lines is expected. These efforts are expected to include quantification of the advantages which might be gained in reducing pouring room and shakeout emissions if the carbohydrate binder is used for molding.

CONCLUSIONS

Typical core room emissions include hydrocarbons ranging from methane to the light oils as well as aromatic compounds, including benzene. Other emissions may include carbon monoxide, carbon dioxide and other simple organic compounds such as formaldehyde. Some processes may release amines, ammonia, and sulfurous gases, depending upon the specific core binder.

There are essentially two sources of these emissions: the curing oven and the sand binder. In either case, these emissions are most often directly vented to the atmosphere. They present an undesired pollution and may have an irritating odor.

One engineering solution to the problem of controlling the fugitive emissions from the core room of the foundry may lie in binder substitution. A new type of binder based upon starch has been identified as a possible core oil substitute, and it promises to greatly reduce binder emissions in the core making and baking processes.

A new method of drying these carbohydrate-bonded cores employing microwave energy may also improve, or essentially eliminate, core room emissions in the foundry.

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OCTOBER 25, 1978

Wednesday Morning - SESSION V

Session Chairman: Henry J. Kolnsberg
Senior Project Manager
TRC - The Research Corporation
of New England

FUGITIVE EMISSIONS PROBLEMS AND CONTROL AT A SURFACE COAL MINE

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ABSTRACT

Fugitive dust control is not an exacting science and varies from region to region depending mainly upon climate, soils, and vegetative cover. Many new and innovative control techniques are infeasible due to their cost, effect upon equipment, or other forms of environmental degradation, i.e. water pollution. After studying various forms of fugitive dust control, I feel that implementation of conventional control techniques are usually the best.

Specifically, wind erosion of barren areas can be best controlled by application of hay mulch and/or seed depending upon the length of time an area is to be left unmined. When using electric drive trucks, i.e. Electro-Haul trucks, the most feasible control of pit haul road dust is through the use of water trucks. Chemical additives oftentimes can have a detrimental effect upon the electrical or mechanical systems of a haulage vehicle when used over a period of time. Many other forms of fugitive dust control have been recommended, i.e. irrigation sprinklers on roads, blasting mats, spray nozzles on shovels, water bags in blasting holes, etc. AMAX will continue to evaluate these types of control for application at their Wyoming mines. However, most of the tests so far have shown new techniques not feasible due to weather extremes, costs, effect on equipment and general unworkable situations.

Fugitive dust takes on many different meanings when defined in the eyes of the regulator, the environmentalist, the industrial representative, the agriculturalist and so on. Its definition as published in the "Prevention of Significant Deterioration Regulations," developed by the United States Environmental Protection Agency will ultimately be litigated. Regardless of the outcome of all of the interpretations or adaptations of the definition of fugitive dust, it will continue to cause problems for all concerned. My remarks today are limited to those problems associated with a large surface coal mine operating in the State of Wyoming, namely the AMAX Belle Ayr Mine.

The Belle Ayr Mine produced some 13.3 million tons of coal in 1977, giving it the distinction of being the largest producing coal mine in the United States. We will exceed this production level in 1978. Along with the prestige of being the largest mine comes the requests to do environmental impact studies on the mine, i.e., air, water, wildlife, etc., inspections from curious and serious regulators and environmental groups, salespersons, interested citizens and so on. Each of these groups have their own ideas on how we should control fugitive emissions. These range from doing nothing to enclosing the pit and putting on a baghouse to shutting down the entire operation. The latter two, however, would not be economically feasible for AMAX nor in the best interests of our Nation's energy goals. The AMAX Belle Ayr Mine is regulated by U.S.G.S., BLM, EPA, OSHA, MSHA, OSM, Wyoming Department of Environmental Quality which has an Air Quality, Land Quality, and Water Quality Division and the list appears to be broadening during each session of Congress. This serves only to further distort already cloudy issues.

Fugitive emissions sources are numerous in a surface coal mine. They include:

1. Haul road traffic - coal and overburden
2. Shovel-truck loading operations - coal and overburden
3. Topsoil removal
4. Blasting - coal and overburden
5. Truck dumping - coal and overburden
6. Open storage piles - not usually allowed for coal
7. Exposed areas including topsoil piles and grading and redistribution of topsoil and overburden
8. Haul road construction and maintenance
9. Train loading - coal

A few of the above sources are amenable to conventional treatment and are easily controlled. The other sources of fugitive dust are not so easily controlled both economically and technically. Control devices or methodologies must change with geographical locations due to climate, soil types and conditions and differing types of equipment used to mine coal.

Electric wheel motors, 100 degree temperatures, 10-20% relative humidities, wind chill factors of -50 degrees and strong winds, make control of fugitive dust seem impossible. Many natives of the State of Wyoming also feel it is impractical given existing dust levels.

There are several approaches to control of fugitive dust. AMAX has experimented with a number of these with limited success. When a new control is suggested, before any hands-on experimentation, one must go through a rigorous review of the control and potential problems. We review new control practices and techniques for their economical impact, i.e., cost per ton of coal produced, technical feasibility such as the applicability of water spray system operating during freezing conditions, safety considerations, effect on operating equipment, secondary environmental effects, area resource depletion and product availability. Many of the control techniques that I criticize may well be techniques or methods that others have found suitable. I am only reporting findings as they apply to our mine, and those that generally would apply to surface coal mines operating in the "Powder River Basin" of Wyoming.

Let us take each of the sources I have previously listed and discuss fugitive dust problems and control of them:

1. Haul Road Traffic. For those of you not directly familiar with our Belle Ayr Mine, we have two types of haul roads, i.e., overburden and coal. They sometimes cross each other and sometimes are one and the same. The life of a haul road can vary from several days to several years. As such, haul roads cannot be considered permanent as can the entrance roads leading into the mine. Most mines are being required by the Wyoming Department of Environmental Quality, Air Quality Division, to "hard surface" all entrance roads leading into the mine as part of their "permit-to'construct" application. AMAX is experimenting with soil binders and a double chip and seal surface or a two inch asphalt mat to meet these requirements. There are a number of different fugitive dust control strategies for haul road traffic. Most regulatory agencies recognize road watering, silt content of the soil, tire size and vehicle speed in calculating emissions from haul roads (U.S. EPA, 1973). The U.S. EPA publication AP-42 also establishes a climatic factor for rainfall using 0.01 inches of rain for 100% control of fugitive dust. In Wyoming the number of days per year receiving more than 0.01 inches of rain is 100 (U.S. EPA, 1975). Therefore, our climatic factor is $\frac{265}{365}$, or rainfall controls 100% of fugitive dust 28% of the time. Our problem is then controlling fugitive dust the other 72% of the time. This could be done with water trucks, water trucks with chemical additives, chemically treating the roads, hard surfacing the haul road or eliminating the haul road and replacing it with some other means of transporting the coal or overburden in a dust free manner.

My first choice would be to operate water trucks in a manner that simulates a rainfall event on the haul roads comparable to the rainfall event noted in AP-42. This control could also be altered by using something other than a water truck to apply the water, i.e., an irrigation system. This system, however, cannot be used to control fugitive dust during the freezing conditions unless an antifreeze is used in conjunction with the water. Unfortunately, the water and antifreeze make the road slippery and create a severe safety hazard. Also the height of the spray necessary to cover a 100' wide roadway, creates a mechanical nightmare with water being sprinkled almost as high as the truck cab.

Water trucks with chemical additives have proven to be very costly. If we use the chemical for 120 days per year we have spent a lot of money; potentially enough to buy another water truck that can be depreciated over several years. Other chemicals with which I am familiar cost large sums of money at their intital applications, but are reduced in cost on subsequent applications but require continuous maintenance. Some are corrosive, some have a potential impact on the electric wheel motors, and others cake the underside of the equipment prohibiting routine maintenance without high pressure steam or gasoline cleaning treatment. The main factor in making this analysis is that the water truck and operator are still essential to putting on the chemical. We must also be careful that any chemical used will not enter a water course and result in water pollution or a fish kill.

Chemical treatment and hard surfacing of haul roads can be treated as if they were the same because the constraints that limit their use within a surface coal mine are the same. Chemical treatment or hard surfacing of a haul road is very expensive due to the depth the application would have to be made to in order to withstand the weights of the haul trucks. An unloaded overburden truck weighs approximately 35 tons and a loaded truck will weigh in the 145 ton range. The chemically treated road would pot hole out as would a hard surfaced road. Spillage onto the road would eventually cover the chemically treated surface as well as a hard surface. The road is probably not in existence long enough to warrant this type of expenditure. Additionally, most mines use a motor grader to rip the road and then apply aggregate for traction during winter, i.e., snow and freezing conditions.

The movement of coal and overburden in other "dust free" manners is being explored by AMAX and other coal companies. I am talking about the use of enclosed conveyors for long haul situations. This alternative is not only being explored from a dust reduction standpoint, but also as a method to achieve greater economic efficiency. However, there are still some unresolved problems concerning conveyor system usage in AMAX.

Given the above brief explanation of money, technical shortcomings, mechanical hazards and unworkable situations, I believe that AMAX will

continue to utilize water trucks for fugitive dust control on haul roads. We will work with vehicle speeds and other readily available, proven techniques to control fugitive dust. We will also continue to explore the use of chemicals in water trucks, chemical treatment of roads and hard surfacing roads, but probably would not expend any large sums of money on implementation unless frequent violations of the ambient air quality standards for the State of Wyoming are noticed. We will continue to look at ways to increase water truck efficiency. This includes the installation of a surge tank at Belle Ayr for reduced cycle times and larger water truck tanks. In areas where water is not so abundant the use of chemicals may be quite advantageous in controlling fugitive dust. My discussion related only to our situations.

2. Shovel-Truck Loading Operations. Both overburden and coal are removed utilizing a truck-shovel operation at the AMAX Wyoming mines. Typical shovel bucket sizes are 24 cubic yards on the overburden shovels and 40 cubic yards on the coal shovels. Fugitive dust control on the shovels is a potential problem. A great deal of the time the material loaded is moist enough or frozen to the point that it is not a problem. Problems occur during the hot, dry, windy days of the summer months. I have worked with the idea of a spray system mounted on the shovel but feel that a potential safety hazard exists with water being sprayed around a shovel that utilizes a very high voltage line for power, i.e., 4160 volts. The spray system would not penetrate the coal or overburden surface enough to adequately control the dust. Other potential problems include alteration of the heating value of the coal by adding too much moisture, which is probably insignificant, coal sticking in the trucks and loadout silos, overburden sticking to the truck beds, limitation of the shovel operators' visibility, and overwatering of the loading area making it slick for truck and shovel operation. Presently, during dusty conditions, our water trucks will back up above the shovel and shoot water onto the bench of coal or overburden as a short-term mitigating measure.

3. Topsoil removal. Surface mine operations are required to save topsoil for later use. Topsoil removal is accomplished with pan scrapers. Dust eminates from working areas, haul roads and storage piles. To eliminate this fugitive dust problem and avoid potential soils contamination by using chemicals, we require all topsoil removal operations to be accompanied by a water truck. If the water truck breaks down during the topsoil removal operation and conditions are dusty, the operation will be shutdown until the water truck is back in service. All of AMAX topsoil removal operations at Belle Ayr are on a contract basis. This requires the water truck or trucks be included in the bid. We are presently experimenting with pre-wetting areas to be stripped of topsoil with an irrigation system and staged stripping of topsoil to take advantage of spring rains.

4. Blasting - Coal and Overburden. AMAX normally will blast 75% of the overburden and 100% of the coal removed. This is done to increase shovel

efficiency. I have helped with experiments incorporating the use of water sprayed over the area to be shot, water bags put down the hole on top of the explosive, differing types of energizing systems to actuate the shot (nonel, prima cord) with no appreciable reduction in dust levels from the shots. Some people have suggested the use of blasting mats to control dust. Blasting mats were developed to control flyrock, not dust. With all of the safety (MSHA) regulations, OSM regulations and other common sense controls, I am of the opinion that there is very little or nothing we can do to control fugitive dust from blasting operations. Maybe blasting operations should not be considered as a problem as it is short-term, has limited control possibilities, and most of the dust settles in a short distance from the shot.

5. Truck Dumping - Coal and Overburden. Truck dumping of coal into a hopper can be easily controlled. There are two approaches: a baghouse system drawing a negative air pressure on the hopper, or using a spray bar system creating a fine water mist over and in the truck dump. Such systems are commercially available and as such do not present an unsolveable problem. Overburden truck dumping is a source of fugitive dust and I am not familiar with any types of control other than reducing drop heights from the trucks, limiting the heights of piles within the pit to keep it out of the wind, orienting dump areas at right angles to prevailing winds, and watering turn around and backup areas next to the overburden dump.

6. Open Storage Piles. Open coal storage piles are usually not allowed by the State of Wyoming Air Quality Division. Open storage of topsoil is controlled in the same way that we control exposed areas as discussed below. There are a number of chemical binders, mechanical methods and other control strategies that can be implemented to control fugitive dust from coal storage piles. As I do not have any actual experience with these, I will not discuss their application.

7. Exposed Areas Including Topsoil Piles and Grading and Redistribution of Topsoil and Overburden. I have developed a wind and water erosion control strategy for the AMAX Belle Ayr Mine as per the OSM requirements. Briefly, AMAX relies upon SCS established practices for controlling wind erosion of exposed areas. Included within controlling fugitive dust from exposed areas is limiting vehicular traffic to established roads or staking off non-travel areas. The area of soil exposed and length of time that it is exposed should be kept to a minimum. There are several practices that can be utilized to control fugitive dust from exposed areas. Usually, the lowest cost practices are the most effective in controlling erosion. Straw mulch or hay mulch at an application rate of two tons per acre can control up to 90% of the wind erosion. The mulch should be anchored to be effective in controlling erosion.

Practices to reduce fugitive dust:

Practice	Condition Where Applicable
1. Disturbed Area Stabilization (with mulching only)	On areas to be bare less than 6 months or where seedings cannot be made.
2. Disturbed Area Stabilization (temporary seeding potential fertilization)	Areas which would remain bare for one year or less before permanent grading and seeding.
3. Disturbed Area Stabilization (permanent seeding potential fertilization)	On bare areas where permanent vegetation is needed.

Fugitive dust from exposed areas can be effectively controlled if mulching, seeding and fertilization operations are conducted as soon as possible after the disturbance. Of course, final and permanent reclamation is the ultimate answer to controlling fugitive dust from exposed areas.

8. Haul Road Construction and Maintenance. Haul road construction is usually done by backfill operations utilizing dozers and front end loaders while maintenance is done by motor graders. The only realistic control again dictates the use of water trucks to control fugitive dust. Please note discussion of chemical usage in Part I, Haul Road Traffic.

9. Train Loading - Coal. Actual monitoring tests have shown this source to be of little significance when left uncontrolled. Coal is loaded by gravity flow from a 4 silo arrangement into a train driven underneath. The moisture content of the stored coal probably reduces emissions more than anything. There are spray bar systems with or without chemical additives or binders available for use. Our experiments have shown to date that these systems are not necessary. Future tests may require further scrutinization of these controls.

Other sources of fugitive dust contained within the pit of a surface coal mine produce insignificant emissions when compared to those discussed already. I have limited the scope of this paper to those fugitive dust problem areas and potential control techniques that I am familiar with. I am sure that there are a number of other control techniques that merit consideration and experimentation by the AMAX Coal Company. AMAX is dedicated to continued evaluation of these types of new and innovative techniques for application at the Belle Ayr Mine. We have found by actual monitoring that county gravel roads probably produce as much if not more fugitive dust than the mines. That is why we are hard surfacing light duty access road utilized

by our employees going to and from work.

A lot of the issues concerning "fugitive emissions problems and control" will eventually be resolved by the courts. We must not lose sight of the Nation's energy goals, and the fact that the cost of controlling fugitive emissions will ultimately be borne by the consumer in the form of increased electrical rates and product values. Fugitive emissions control should not neglect public health considerations as related to particle size distribution. It is my contention that a large percentage of fugitive dust falls out of the air before it ever hits the pit boundaries, let alone before it reaches property lines. Property line concentrations should be regulated levels, not pit concentrations. Pit concentrations are already being monitored by the use of dust pumps as required by MSHA and compliance with these standards is ongoing at Belle Ayr. These tests relate to the respirable fraction of the dust.

We need to take a realistic approach to fugitive emissions control. New technology must be given time to be proven both technically and economically. Thank you for the opportunity to speak at this symposium. I hope you find my remarks enlightening and challenging for without challenge there would be no change.

AIRCRAFT TURBINE ENGINE PARTICULATE EMISSION CHARACTERIZATION

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ABSTRACT

A particulate matter sampler is described along with operational experience and the data obtained using the sampler on the JT8D and JT9D aircraft turbine engines. The sampler acquires a sample of the exhaust particles emitted during the ground operation of the engine and permits the characterization of the mass concentration, elemental composition, particle size distribution, and particle morphology. Mass concentration is obtained gravimetrically. Elemental composition is obtained by energy dispersive X-ray and particle size and shape by electron microscopy and image analysis. Particle size data is also obtained with the Electrical Aerosol Analyzer.

The sample is extracted from the exit plane of the engine. The sampling probe geometry corresponds to the FAA's recommended methodology for gaseous emission sampling. The extracted sample is conditioned through dilution to minimize local condensation, particle deposition, and sample bias before collection on substrates suitable for analysis.

THE PHYSICAL AND CHEMICAL CHARACTERIZATION of particles emitted by aircraft turbine engines has not received comprehensive investigation. Therefore, a sampling system was designed and constructed to collect particles from aircraft turbine engines (1, 2).* The sampler is specifically designed to collect representative samples of particulate matter from the exhaust at the exit plane of low bypass ratio, mixed flow, and high bypass ratio aircraft turbofan engines operating in an engine test cell. These engine types are represented in the current commercial fleet by the JT3D, JT8D, and JT9D series of engines. The sampler uses a mixing type sampling rake, a specialized aerosol transport and conditioning system, and sample collection near the engine to minimize particle deposition, re-entrainment, and agglomeration during transport. A feature of the sampler is the bifurcation of the extracted exhaust sample - a large portion for bulk analysis and a smaller portion for detailed particle characterization.

SAMPLER DESCRIPTION

A flow diagram of the sampler is shown in Figure 1. A "diamond" shaped sampling rake mounts directly behind the engine exhaust plane. An extraction nozzle is located at the midpoint of each leg of the diamond. The four nozzles are evenly spaced on an arc whose radius is 62% of the engine's exit plane radius. The leading edge of the nozzle extends about ten diameters upstream of the support structure. The nozzle diameters are 0.170 ± 0.008 cm. The geometry of the rake corresponds to that recommended by the Federal Aviation Administration (FAA) for gaseous emission sampling and has been shown to give comparable results to a 24 sampling nozzle rake system (3). The four sampling nozzles are manifolded to provide a composite sample. The manifold is connected to the primary diluter via a flexible coupling that accommodates movement of the engine during testing.

The function of the primary diluter is to condition the extracted particulate matter sample. The sample is cooled without passing through a dew point and diluted to reduce particle agglomeration effects. The unique design of the diluter reduces wall losses and aids in the preservation of particle mass and particle size distribution (4, 5). The diluter consists of a porous tube housed within a larger tube. Clean, dry, metered, compressed air is delivered to the annular space at four locations and flows through the porous inner tube providing a clean boundary layer that reduces particle deposition and subsequently mixes with the sample to provide dilution and temperature control.

*Numbers in parentheses designate References at end of paper.

After conditioning, the sample is bifurcated. The majority of the sample is transported to the mass sampling turret where the particulate matter is removed by filtration onto tared Gelman, type AE, glass fiber filters, 142 mm in diameter. A smaller portion of the sample is adjusted in concentration by a secondary diluter similar in design to the primary diluter and transported either to the Electrical Aerosol Analyzer (EAA) for real-time particle size distribution measurement or for collection onto membrane filters and electron microscope grids for subsequent analysis of particle size, shape, and elemental composition. The membrane filters used are 0.03 μm pore size Nuclepore filters, 47 mm diameter.

With the exception of the EAA, the sampler is designed for operation in close proximity to an operating turbine engine. All functions of the sampler, therefore, are remotely controlled from a centralized control panel that can be located up to 25 feet from the filtration units in a safe location such as the test cell control room. The EAA continuously malfunctioned in the test cell environment despite efforts to provide a suitable sound protection enclosure. As a result, the EAA is operated in the test cell control room.

Flow control valves for the sampler are located downstream of the particle collection sites to eliminate particle deposition in the valve assembly. This requirement imposed operational constraints in the system--each sample has to be acquired sequentially and flow switching procedures must be carefully performed to prevent rupture of the filters. Also, because the extracted sample flow from the engine is not measured directly, the primary diluter flow and the total sample flow have to be accurately determined. The dilution flow rates are measured by precision drilled orifices; total flow is measured with a hot-wire anemometer. The uncertainty in total flow measurement is ±0.09 std. m^3/hr. while the dilution flow is repeatable to approximately ±0.1 std. m^3/hr. The maximum error in the extracted sample flow measurement occurs at idle power and is on the order of 10%.

The turret assemblies for sample collection provide positions for six filters. Samples, thus, can be acquired at five engine power settings -- idle, approach, cruise, climb-out, and take-off -- before the engine needs to be shut down and the filters recovered. The sixth turret position is a dummy used during start-up, shut-down, and while power settings are being changed and engine operation stabilized.

The sampler is designed for isokinetic sampling of the exhaust from the JT8D and JT9D engines at power settings from idle through cruise. At climb-out and take-off power, sonic conditions are approached at the engine's exit plane and sampling at this rate would cause severe flow complications resulting in modification of the extracted exhaust particles. The flow through the extraction nozzles was, therefore, limited to a local Mach number of 0.8. Extracted sample flow ranged from 0.9 std. m^3/hr. to 4.8 std. m^3/hr. for engine power settings from idle to take-off (6).

TEST SITE AND DATA ANALYSIS PROCEDURES

The particulate matter sampler was operated at United Airlines' San Francisco, Ca., Maintenance Facility on JT8D and JT9D aircraft turbine engines. Figure 2 shows the sampler set-up in a JT9D test cell. Due to United's fleet needs, sampling time was at a premium and several different engines were used during the test series. The engines tested are given in Tables 1 and 2 along with data on the mass emission rate and the particle size distribution of the exhaust particles as determined by electron microscopy and image analysis. In addition, particle sized distribution measurements were made with the EAA and particle shape was determined by image analysis. Two fuels were used during the test series, pearl kerosene (PK), a high hydrogen, low aromatic content fuel, and JET A, a standard commercial aircraft turbine engine fuel with a lower hydrogen and higher aromatic content. The use of two fuels permits the determination of the effect of fuel composition on emissions. A single batch of PK fuel was purchased and stored at United's facility. Its composition, therefore, was constant throughout the tests and repeatability measurements were possible. JET A fuel composition varied during the tests. All fuels were sampled and analyzed; significant data is given in Table 3.

Mass emission rates were determined by differential gravimetric analysis. A Gelman, type AE, glass fiber filter was used to collect the particulate matter for mass determination. The filters were weighed on an analytical balance before and after sample collection and corrected for shifts in weight due to hygroscopic effects with control filters.

Particle size data was obtained by two methods. One method used, the Electrical Aerosol Analyzer (EAA), provided a real-time measure of the particle size distribution. The EAA used was Model 3030, Thermo-Systems, Inc., St. Paul, Mn. The instrument was operated in accordance with the operating and service manual provided with the instrument.

The second procedure used for particle size analysis was electron microscopy and image analysis. The samples, collected on Nuclepore membrane filters or carbon coated electron microscope grids attached to the membrane filter, were used in the analysis. The electron microscope used was the JEOL-100C analytical scanning/transmission microscope equipped with an energy dispersive X-ray system for elemental analysis. After applying a thin film of gold to the filter surface with a Hummer II plasma specimen coater, the samples were observed and photographed in the scanning microscope at several magnifications ranging from 10,000 to 100,000X. A magnification of 30,000X was found to be optimal for particle size and shape analysis. Particle size and shape analysis were performed by interrogating the electronphotomicrographs with an Imanco Quantimet 720 Image Analyzer. Particle size was computed from the area measurement of each particle. The particle size reported, thus, represents the diameter of a circle of area equal to that of the particle.

Particle shape was determined using the image analyzer and interrogating the particles for their area and perimeter. A two-dimensional shape factor was then calculated from the ratio A/P^2. This ratio was plotted as a function of the equivalent area particle diameter.

Elemental composition of the particles was determined while the particles were under observation in the electron microscope. A Kevex energy dispersive X-ray spectrometer coupled with the Tracor/Northern 880 X-ray analysis system was used.

RESULTS AND DISCUSSION

MASS EMISSIONS -- Mass emission data are reported in Tables 1 and 2 and plotted for the JT8D engine as a function of primary air flow in Figure 3. The data show an increasing emission rate with power. The emission rate ranged from 0.15 kg/hr at idle to a range of 0.25 to 4.62 kg/hr at climb-out and 0.76 to 1.79 kg/hr at take-off. Data for the JT9D engine show a similar trend and the same order of magnitude. However, primary air flows for the JT9D engine are significantly greater. Primary air flows as a function of engine power is given in Table 4. With the restricted sampling time available, 12-30 minutes depending on the operating mode, and the low emission rates for the JT8D and JT9D engines, the gain in filter weights were only a few milligrams. Additional testing with longer sampling times are needed for a more accurate determination of mass emission rates. Earlier data (2) collected from a TF-30 engine suggest the sampler can reproduce mass concentration data within ±2% at the cruise power setting for this engine. Effects of engine and fuel variability are discernible, however. The JT8D-7 engine, 648735, gave lower emission rates than the other engines and the JT8D-15 engine tested with both JET A and PK fuel gave a lower emission rate with the PK fuel.

PARTICLE SIZE DATA -- Particle size data obtained by electron microscopy and image analysis are reported in Tables 1 and 2 and summarized in Tables 5 and 6. For the JT8D engines, operating on JET A fuel, the average geometric mean particle size, $\bar{d}g$ increased with engine power. At idle, $\bar{d}g$ was 0.045 μm, at approach $\bar{d}g$ was 0.068 μm, at cruise and climb-out $\bar{d}g$ was 0.078 μm, and at take-off $\bar{d}g$ was 0.084 μm. Particle size is influenced by fuel type; PK fuel gave smaller particle sizes. This is consistent with the lower mass emission rate found for the engine when operated on PK fuel. Also consistent is the performance of JT8D engine 648735. This engine, which produced the lowest emission rates, also produced the smallest particle sizes. The geometric standard deviations for the particle size distributions ranged from 1.6 to 2.3. Engine 648735 gave the widest distribution of particle sizes. The JT9D engines gave similar trends to the JT8D engines, but indications are that the particles are smaller.

Particle size distribution data were also obtained with the EAA. Typical data for the JT8D and JT9D engines are plotted in Figures 4 and 5. These data support the results obtained by the electron microscope and image analysis measurements. Higher power settings produce a greater concentration of particles in all sizes ranges above about 0.02 μm. Below 0.02 μm, data is conflicting but the performance of the EAA is known to be erratic in this range (7). Particle concentration for the JT8D engine is about one order of magnitude greater than for the JT9D engine. The influence of fuel composition on particle size is illustrated in Figures 6 and 7 for the JT9D engine at approach and take-off power. At all power settings, PK fuel produced few particles.

PARTICLE SHAPE -- Particle shape data shows the particle structure is more complex at climb-out and take-off than at the lower power settings. Figures 8 through 11 illustrate this effect. The effect is evident from the photographs shown in Figures 12 and 13. As an aid in interpreting the shape factor, A/P^2, Table 7 is presented. The shape factor for a disc, the two-dimensional representation of a sphere, is $1/4\ \pi$ or 0.0795. A 5-chain agglomerate has a shape factor of 0.0159; a 10-chain, 0.0078; and a 50-chain, 0.00016. Thus, as the shape factor decreases the particles are less spherical and more highly agglomerated. Generally, particles with shape factors above about 0.03 can be considered single particles trending toward a spherical shape. As shown in Figures 9 and 11, the very large exhaust particles have small shape factors and are, thus, highly agglomerated. Idle power produces relatively small particles of simple shape while climb-out and take-off power produces a wider range of particle sizes of more complex shape.

ELEMENTAL COMPOSITION -- The elemental composition of the individual exhaust particles was determined by energy dispersive X-ray techniques. No spectrum was obtained indicating the particle composition is essentially carbon.

CONCLUSIONS

The particulate matter emitted during the test cell operation of the JT8D and JT9D aircraft turbofan engines was characterized using a sampler designed expressly for this purpose. The particles are principally carbon and less than 0.1 μm in size. Particle size increases with engine power. At idle, the average geometric mean particle size was 0.045 μm and at take-off, 0.097 μm for the JT8D engine operating on JET A fuel. Particle size is influenced by the fuel composition. PK fuel with its higher hydrogen, lower aromatic, and higher smoke point content gave smaller exhaust particles. Particle shapes are more uniform and trend toward sphericity at idle; at climb-out and take-off the particle structures are more varied and complex. The mass emission rate for the JT8D engine was 0.15 kg/hr at idle and ranged from 0.76 to 1.79 kg/hr at take-off. PK fuel produced lower emissions than JET A fuel. Variability among the engines tested was noted. The limited data available indicate the JT9D produces less particulate matter than the JT8D. Particle concentration data show approximately an order of magnitude difference between the JT8D and JT9D engines.

The results indicate that the sampler provides a valid sample of the particles present at the exhaust plane of aircraft turbine engines.

ACKNOWLEDGEMENT

The work reported was sponsored by the United States Department of Transportation -Federal Aviation Administration under contract DOT-FA75-WA-3722. The assistance of Messrs. Paul Campbell and Ralph Johnson of United Airlines and Dr. Donald Fenton of New Mexico State University is gratefully acknowledged.

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Table 1 -- Summary of Turbine Engine Sampling Data - JT8D

Sample Date	Engine Type	Engine Number	Fuel	Operating Mode	Primary Air (kg/sec)	Particle Emission (kg/hr)	Particle Size Distribution[1]	
							(dg, μm)[2]	(σg)[3]
1/10/78	JT8D-7	654957	JET A	Idle	16	0.15	0.052	1.7
				Approach	33	1.00	0.086	1.7
				Cruise	50	1.78	0.081	1.7
				Climb-out	61	2.97	0.096	1.6
				Take-off	68	0.95	0.099	1.7
4/24/78	JT8D-15	648796	JET A	Idle	17	--	0.042	1.7
				Approach	33	0.30	0.067	1.8
				Cruise	50	1.03	0.092	1.8
				Climb-out	62	4.62	0.088	1.9
				Take-off	69	--	0.097	2.0
4/25/78	JT8D-7	648735	JET A	Idle	17	--	0.043	1.6
				Approach	33	0.34	0.054	2.1
				Cruise	49	0.25	0.069	2.0
				Climb-out	61	0.25	0.049	2.3
				Take-off	68	0.76	0.060	2.2
6/20/78	JT8D-15	696572	JET A	Idle	17	--	0.042	1.7
				Approach	33	0.24	0.066	1.7
				Cruise	50	1.12	0.071	1.9
				Climb-out	62	0.74	0.079	1.6
				Take-off	69	1.38	0.080	1.6
4/24/78	JT8D-15	648796	PK	Idle	17	--	0.032	1.7
				Approach	33	0.08	0.043	1.8
				Cruise	50	0.24	0.079	1.9
				Climb-out	62	1.27	0.075	1.8
				Take-off	69	1.79	0.077	1.9

[1]Obtained by electron microscopy and image analysis
[2]Geometric mean particle diameter
[3]Geometric standard deviation

Table 2 -- Summary of Turbine Engine Sampling Data - JT9D-3A

Sample Date	Engine Number	Fuel	Operating Mode	Primary Air (kg/sec)	Particle Emission (kg/hr)	Particle Size Distribution[1]	
						(dg, μm)[2]	(σg)[3]
1/9/78	662734	JET A	Climb-out	99	--	0.084	1.6
			Take-off	111	--	0.117	1.6
4/21/78	663031	JET A	Approach	62	0.60	0.054	1.8
			Take-off	111	2.87	0.077	1.8
6/21/78	663082	JET A	Idle	30	--	0.041	1.5
			Approach	62	--	0.047	1.7
			Cruise	82	0.32	---	-
			Climb-out	99	--	0.040	1.8
6/22/78	662794	JET A	Cruise	82	--	0.041	1.7
			Climb-out	99	0.22	0.045	1.6
4/21/78	663031	PK	Approach	62	1.33	0.054	1.6
			Take-off	111	--	0.050	1.9

[1]Obtained by electron microscopy and image analysis
[2]Geometric mean particle diameter
[3]Geometric standard deviation

Table 3 -- Significant Fuel Analytical Data

Analysis	JET A	PK
Hydrogen Content, %	13.05-13.65	14.25
Aromatic Content, %	17-20	1
Naphthalene Content, %	1.6-2.5	0.1
Olefins Content, %	1-2	1
Smoke Point, mm.	20-21	35

Table 4 -- Primary Air Flow as a Function of Engine Power

	Primary Air Flow (kg/sec)	
Power Setting	JT8D	JT9D
Idle	16	30
Approach	33	62
Cruise	50	82
Climb-out	62	99
Take-off	69	111

Table 5 -- Particle Size of JT8D Emission as Obtained by Electron Microscopy

Operating Mode	Geometric Mean Particle Size, μm[1]		Effect of Fuel on Geometric Mean Particle Size, μm	
	Range	Aver.	JET A	PK
Idle	0.042-0.052	0.045	0.042	0.032
Approach	0.054-0.086	0.068	0.067	0.043
Cruise	0.069-0.092	0.078	0.092	0.079
Climb-out	0.049-0.096	0.078	0.088	0.075
Take-off	0.060-0.099	0.084	0.097	0.077

[1]JET A fuel

Table 6 -- Particle Size of JT9D Emission as Obtained by Electron Microscopy

Operating Mode	Geometric Mean Particle Size, μm[1]		Effect of Fuel on Geometric Mean Particle Size, μm	
	Range	Aver.	JET A	PK
Idle	---	0.041	---	---
Approach	0.047-0.054	0.051	0.054	0.054
Cruise	---	0.041	---	---
Climb-out	0.040-0.084	0.056	---	---
Take-out	0.077-0.117	0.097	0.077	0.050

[1]JET A fuel

Table 7 -- Particle Shape Characterization

No. of Particles in Agglomerate Chain	Aspect Ratio 1/d	P/d Ratio	Shape Factor A/P^2
1	1:1	3.14	0.0795
3	3:1	9.42	0.0265
5	5:1	15.7	0.0159
10	10:1	31.4	0.0078
50	50:1	157	0.00016

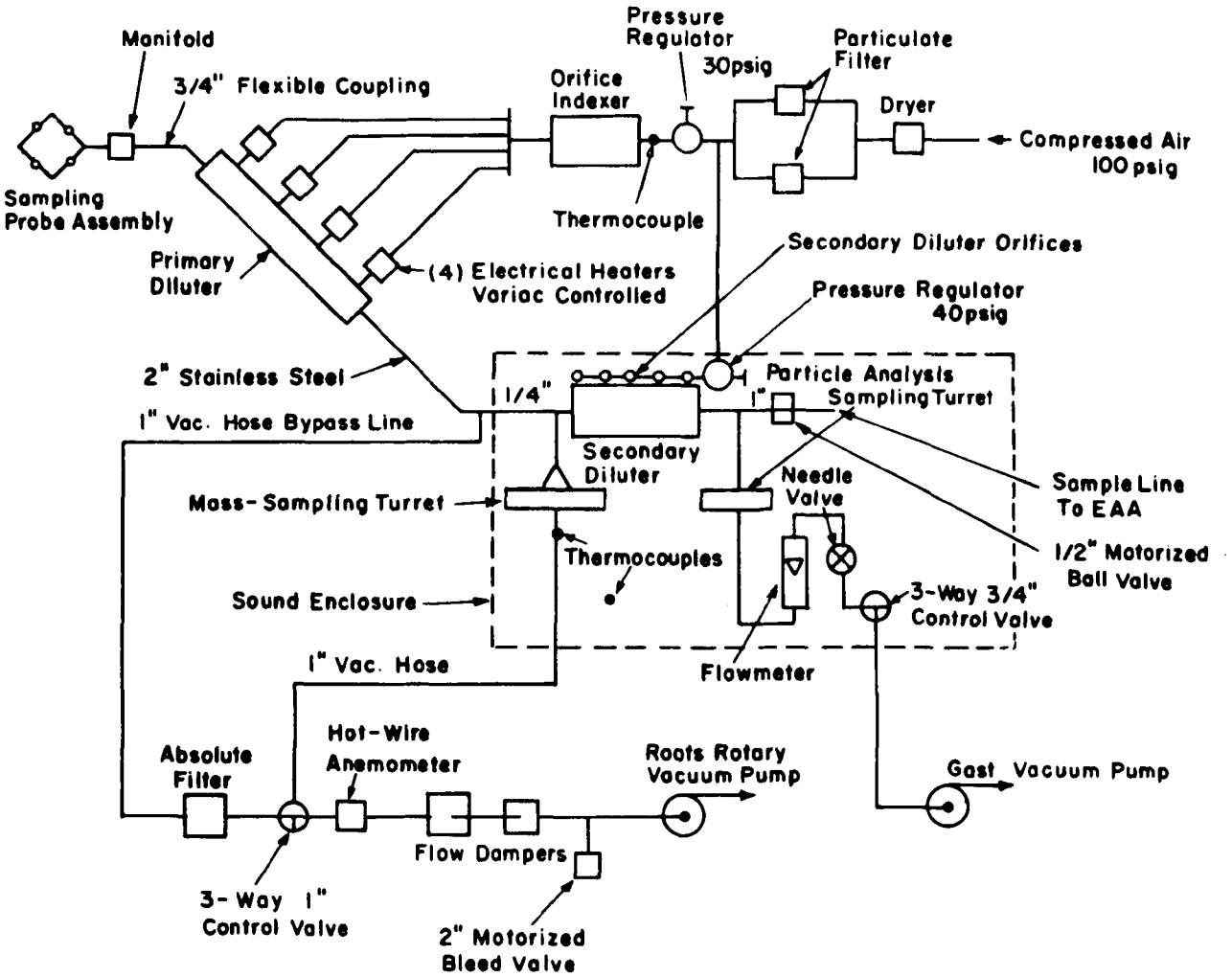


Fig. 1 – Schematic diagram of the engine exhaust particulate sampler

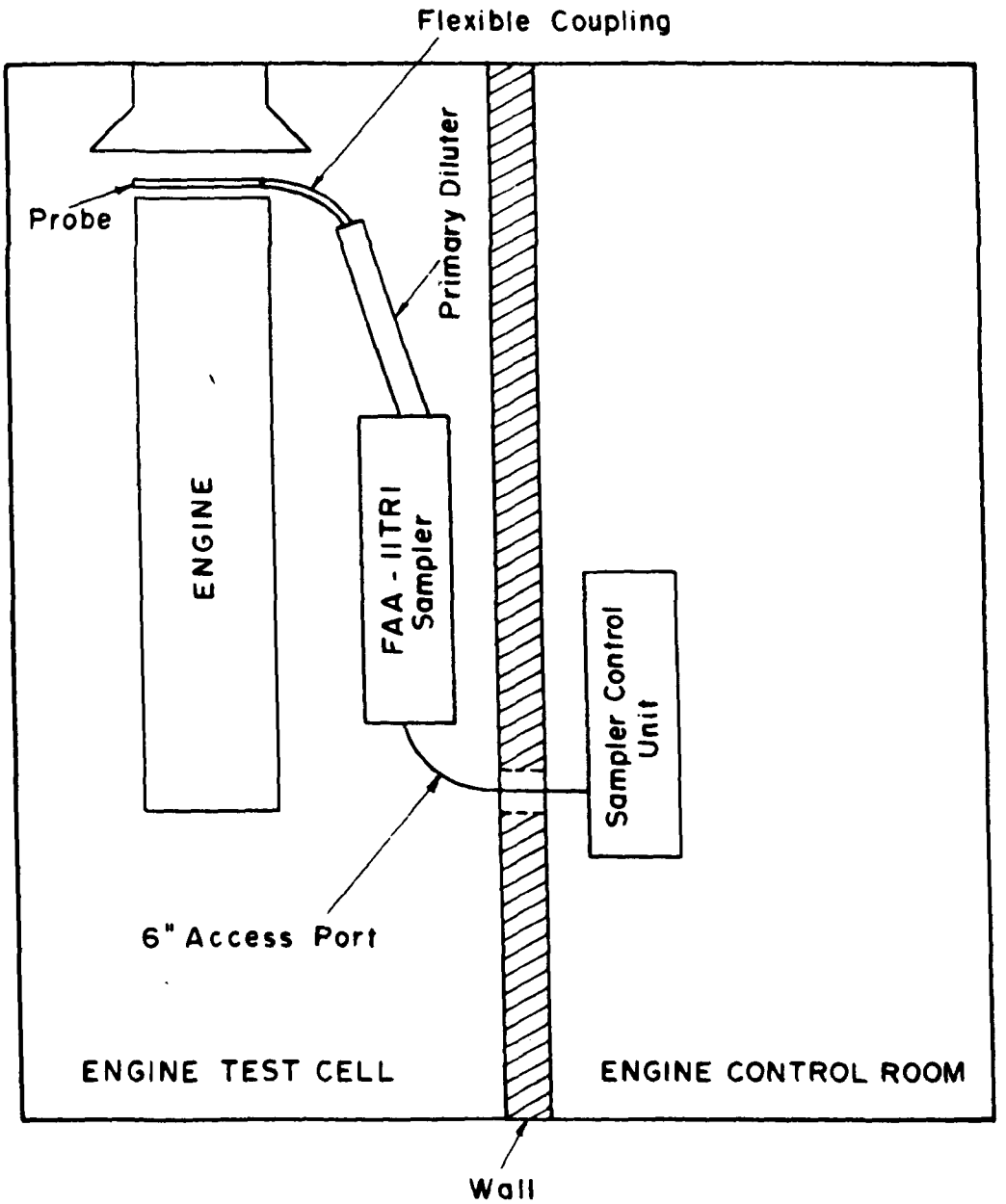


Fig. 2 – Typical layout for FAA - IITRI sampler in an engine test cell

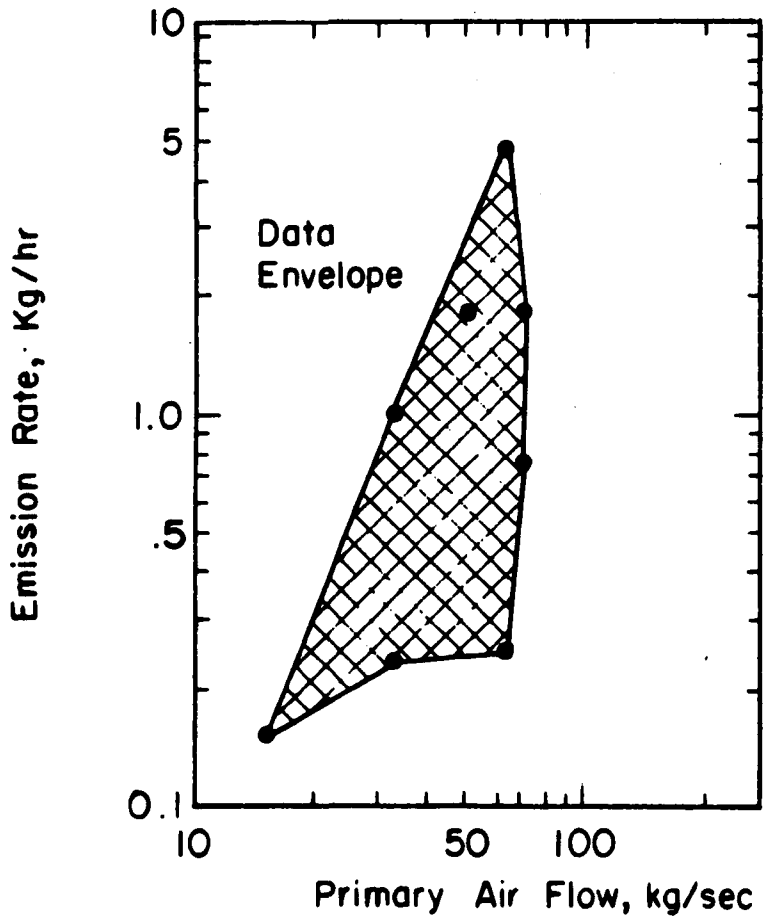


Fig. 3 – Mass emissions for JT8D JET A fuel

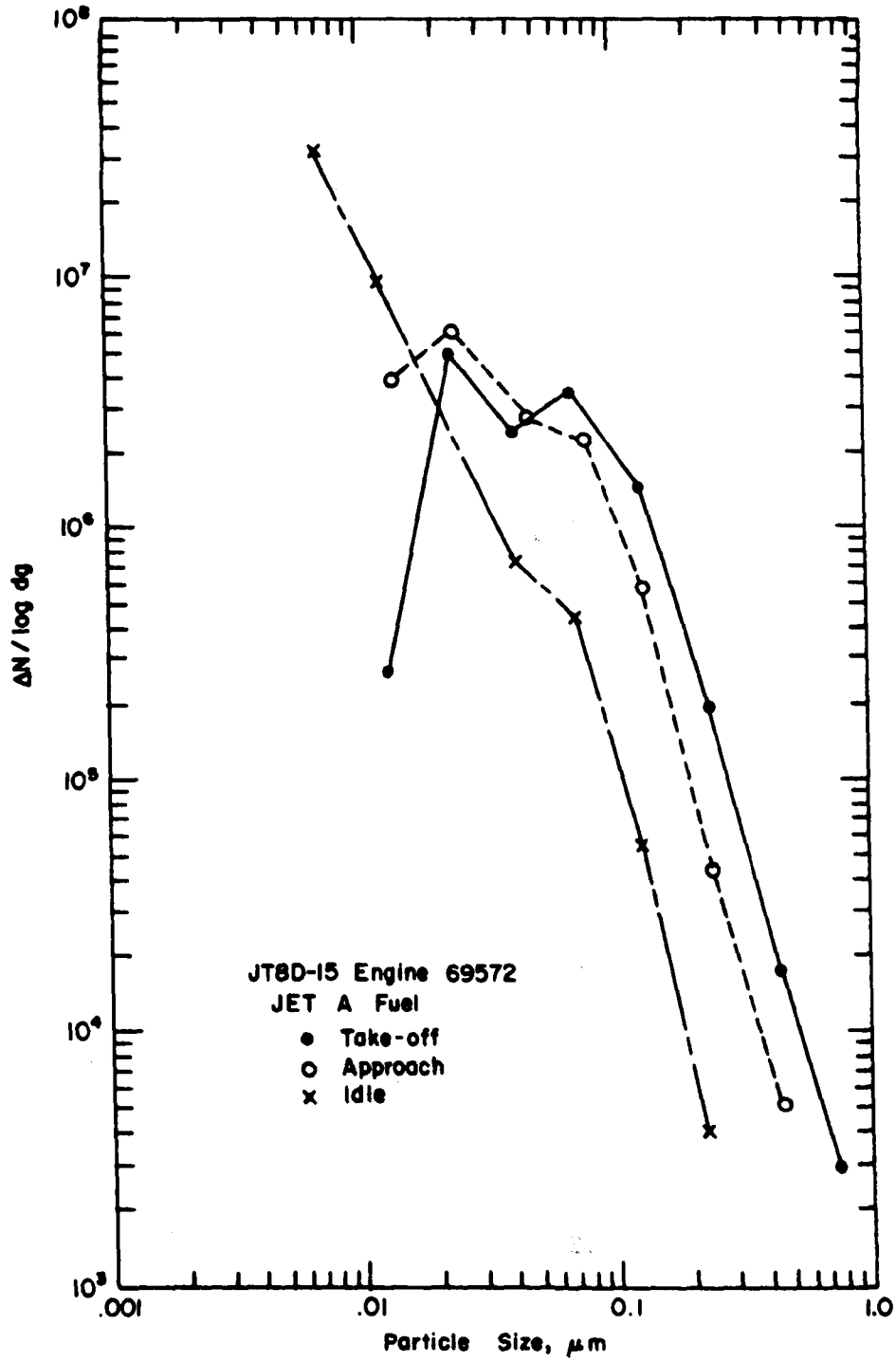


Fig. 4 - EAA particle size data for the JT8D engine

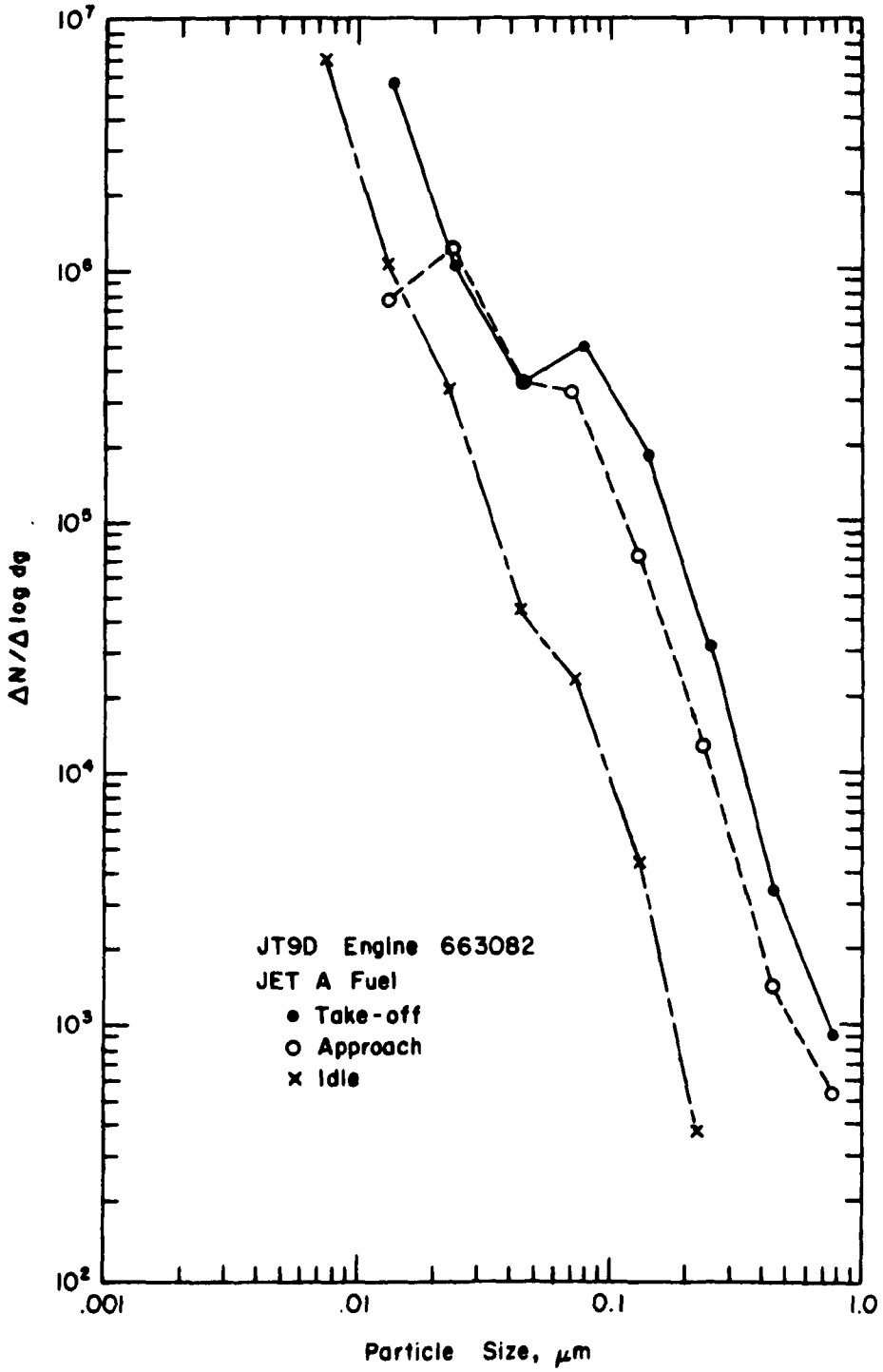


Fig. 5 – EAA particle size data for the JT9D engine

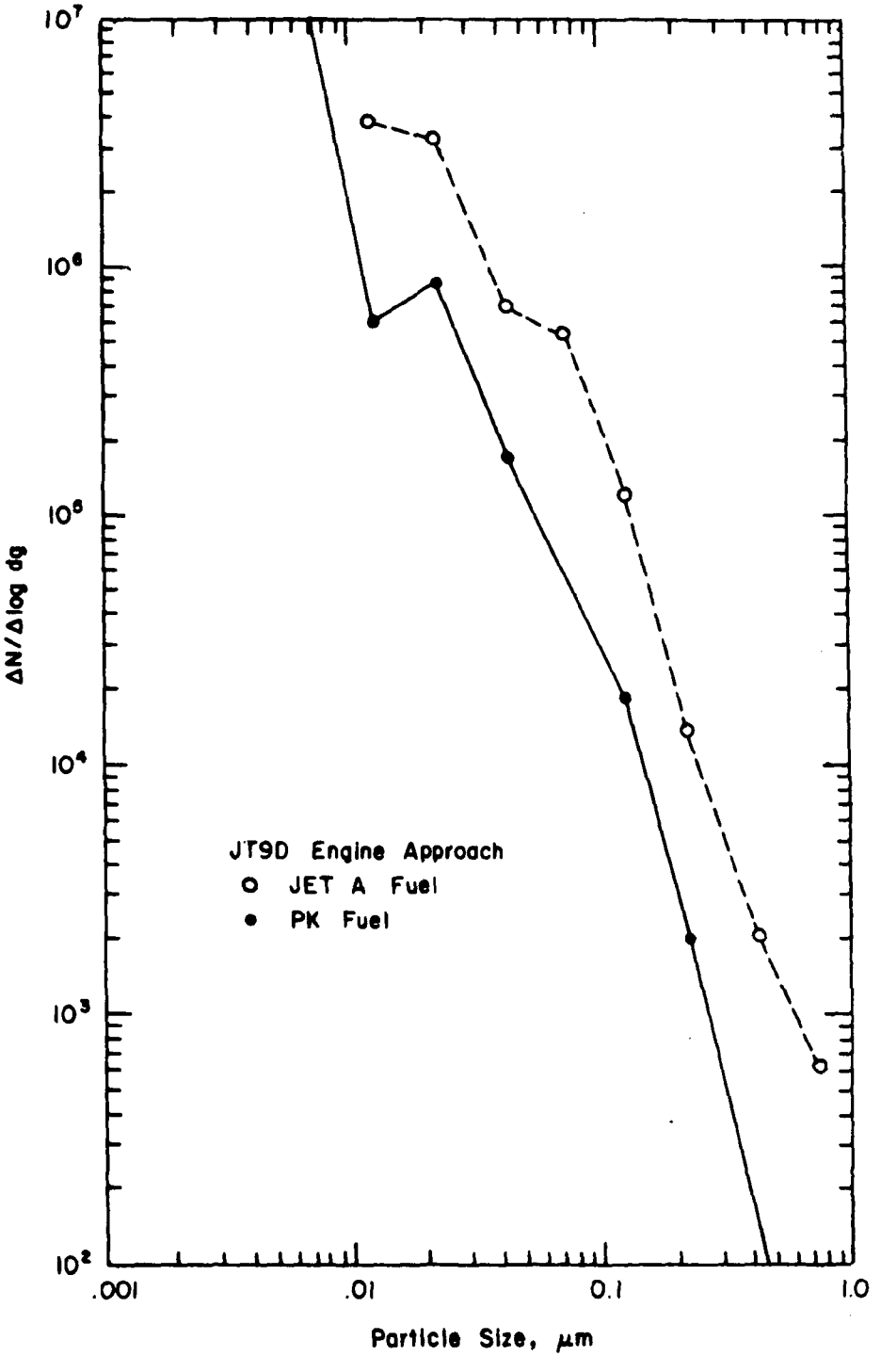


Fig. 6 – Effect of fuel on particle size of emissions

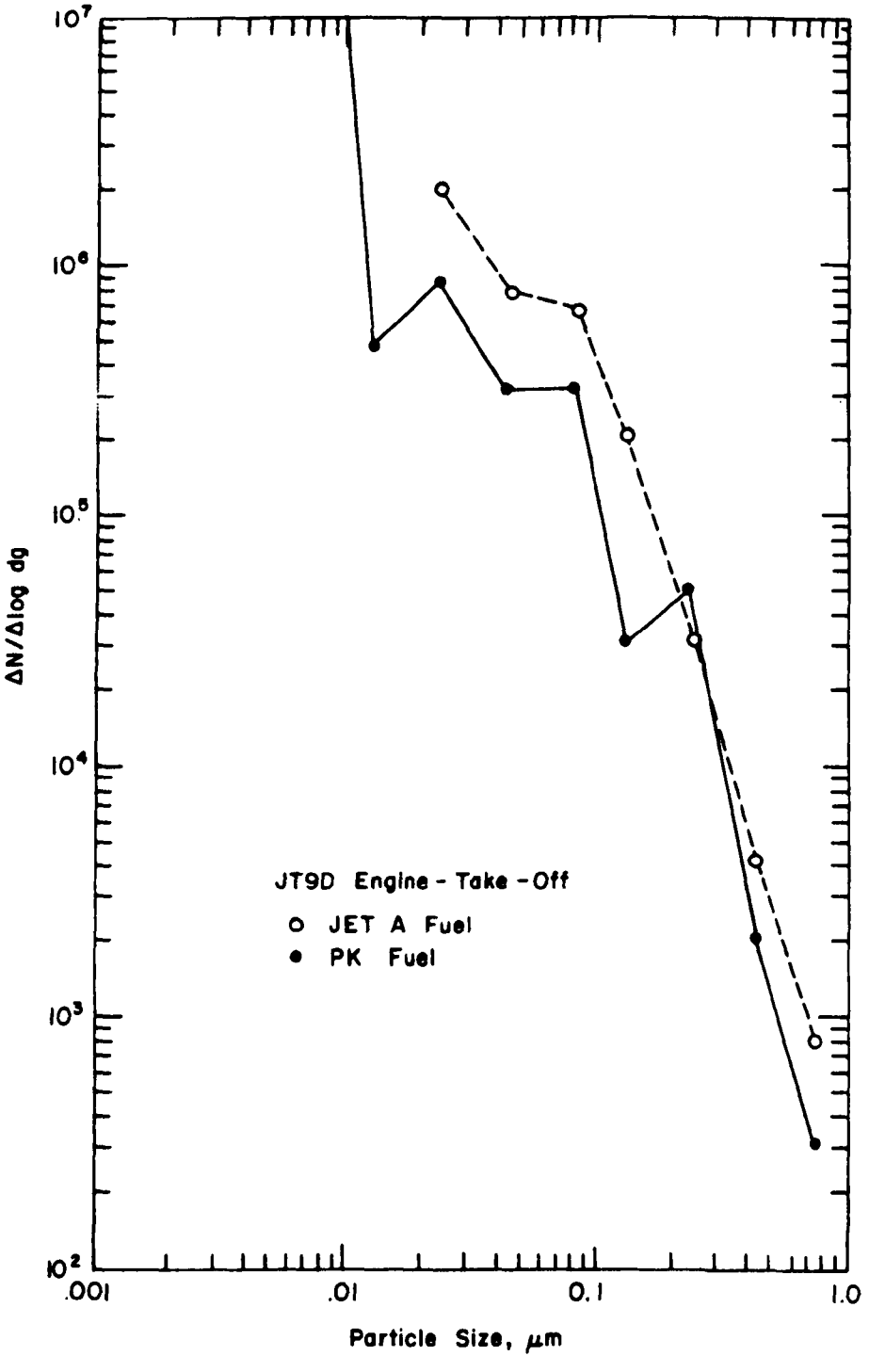


Fig. 7 – Effects of fuel on particle size of emissions

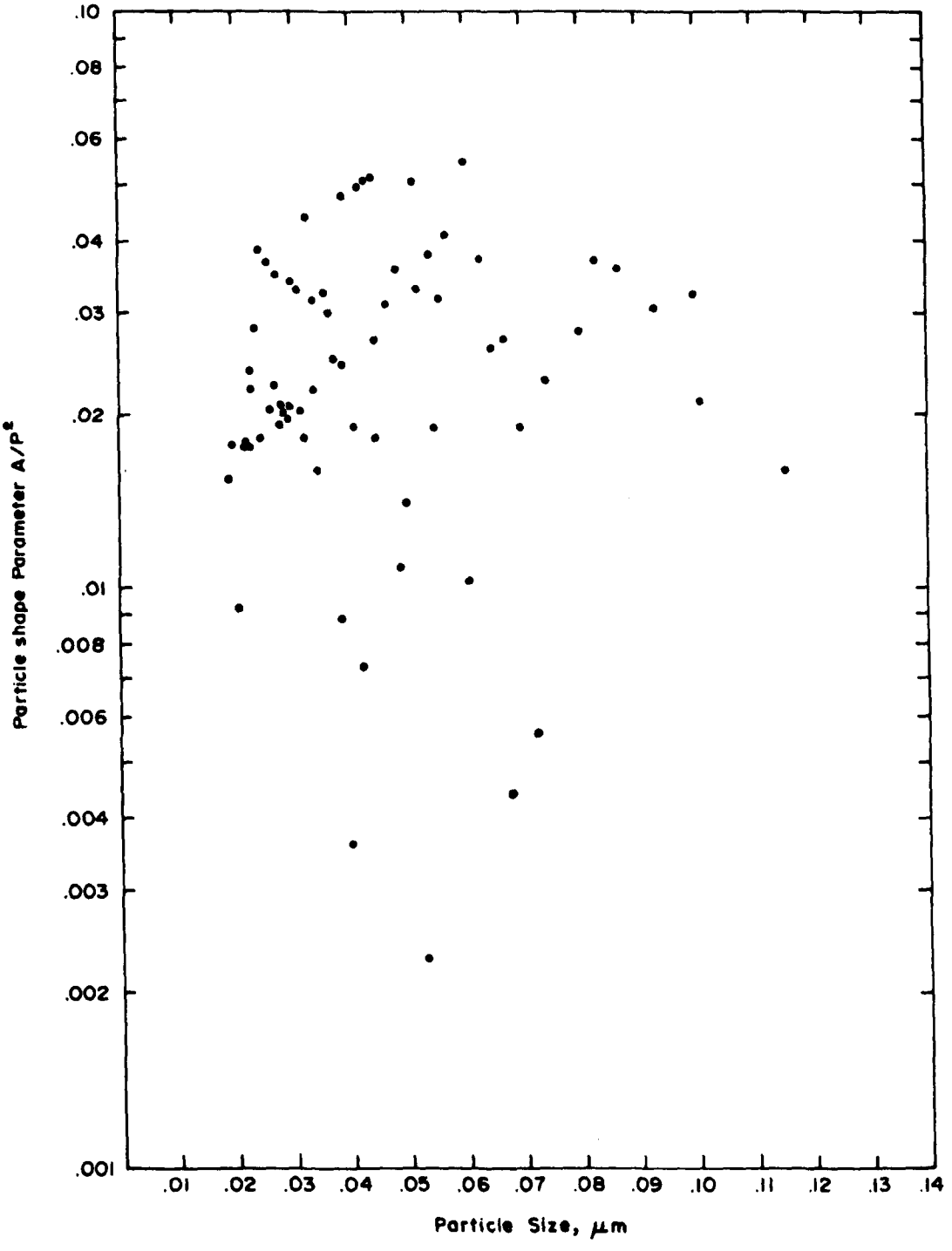


Fig. 8 - Particle shape factor, JT8D engine, idle, JET A fuel.

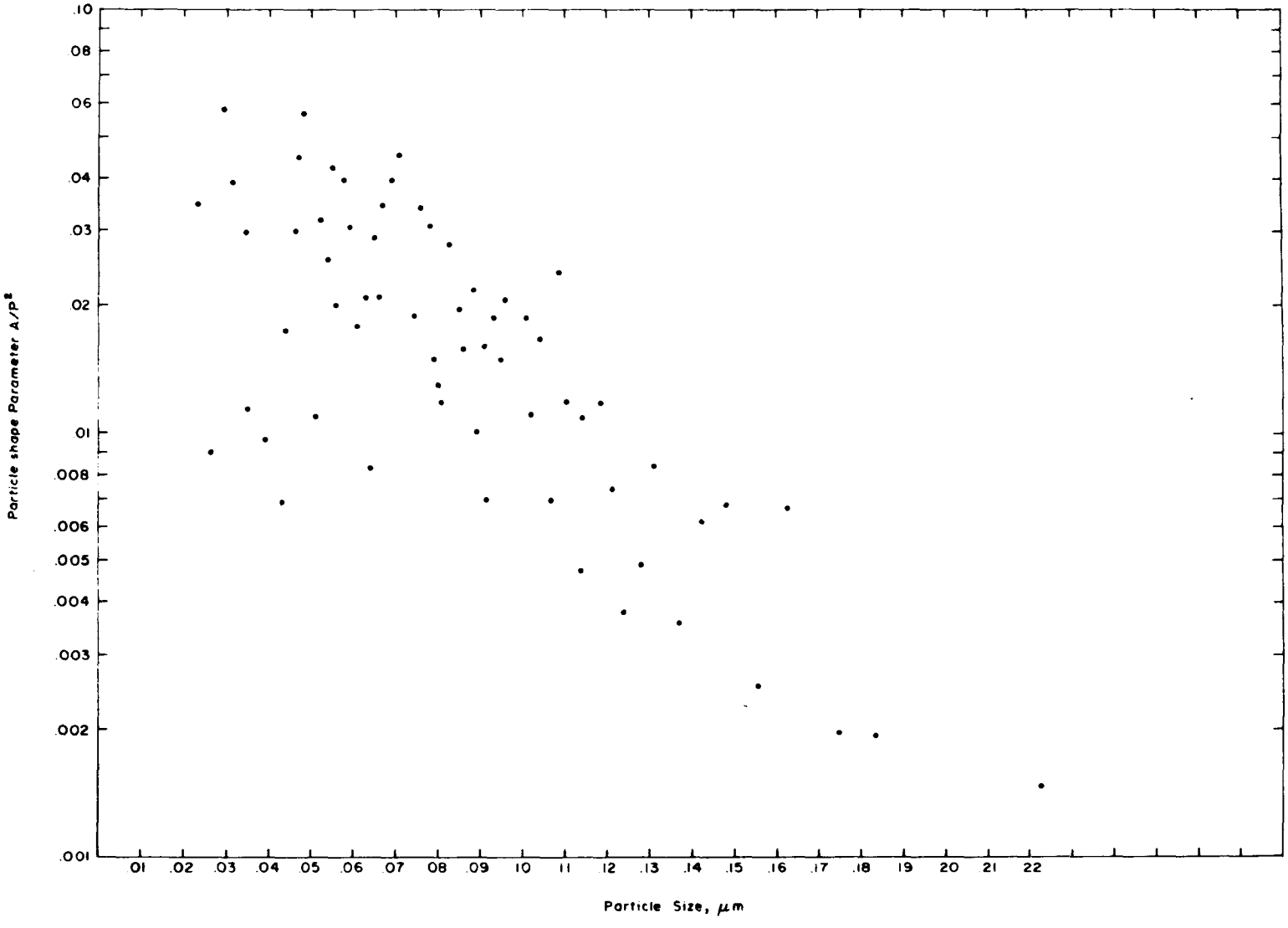


Fig. 9 - Particle shape factor, JT8D engine, take off, JET A fuel

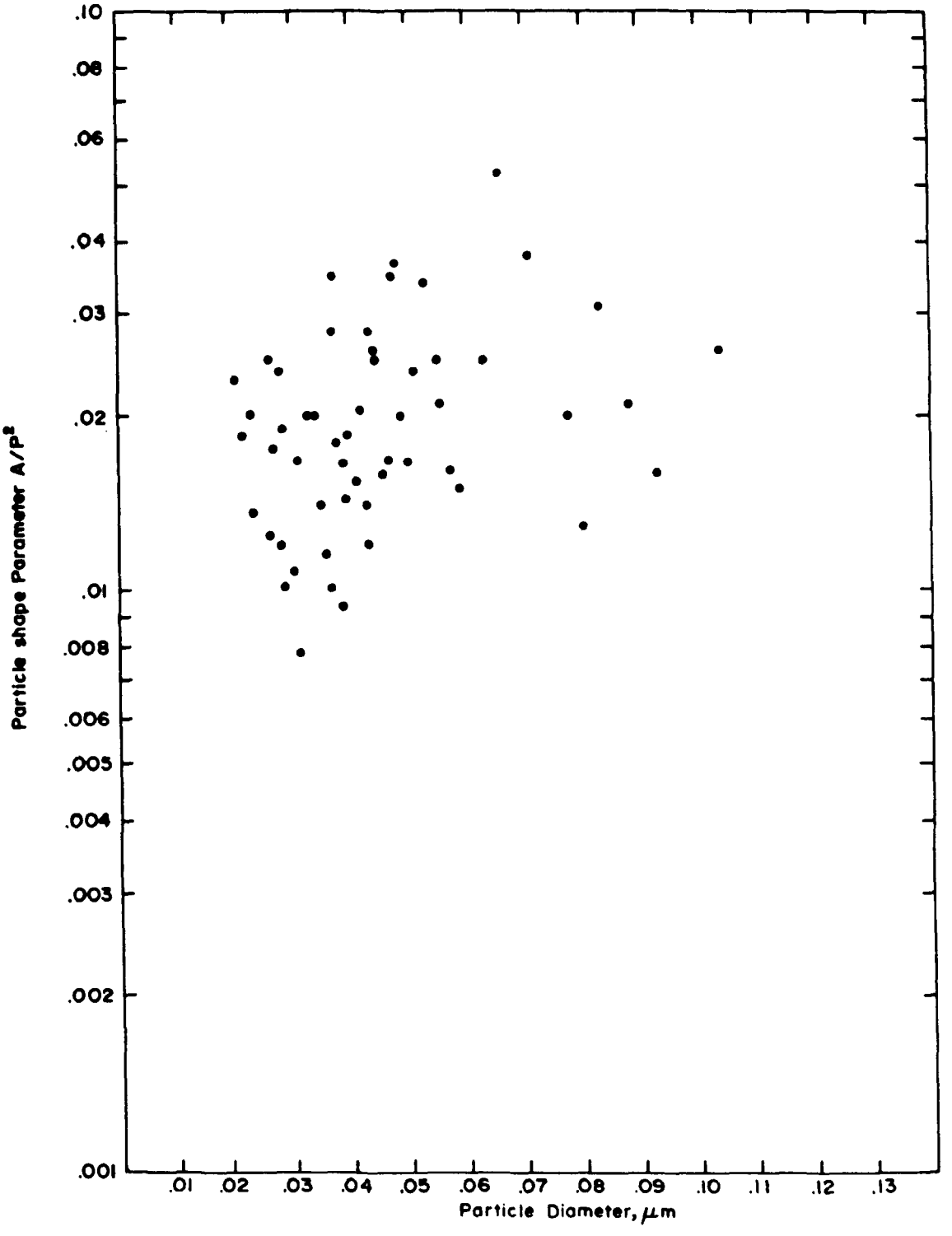


Fig. 10 - Particle shape factor, JT9D engine, idle, JET A fuel

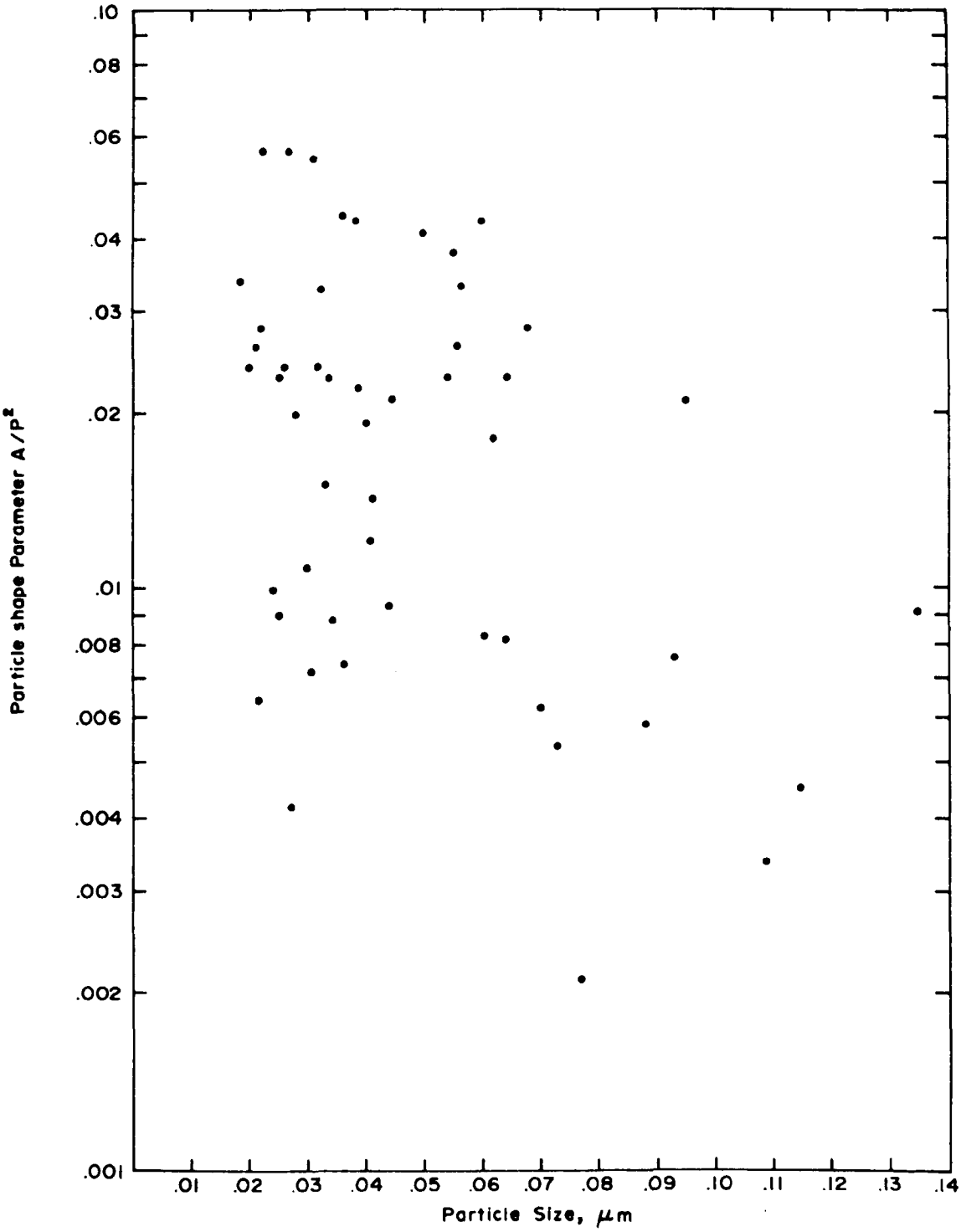
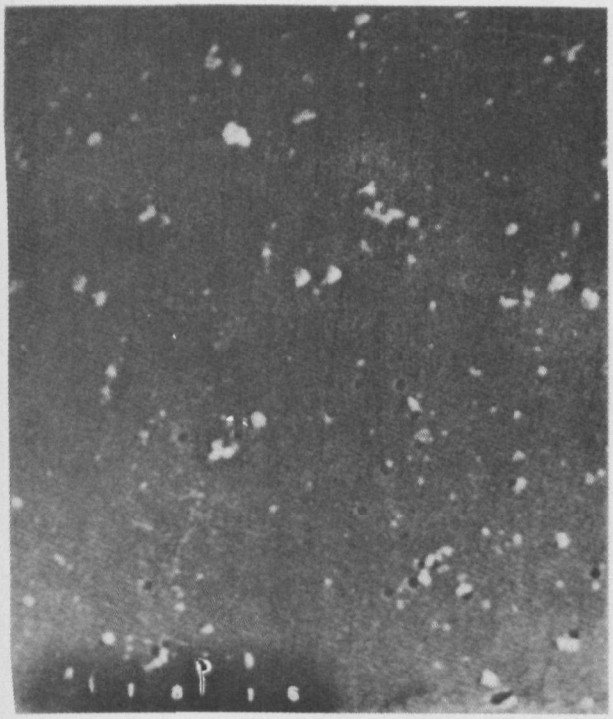
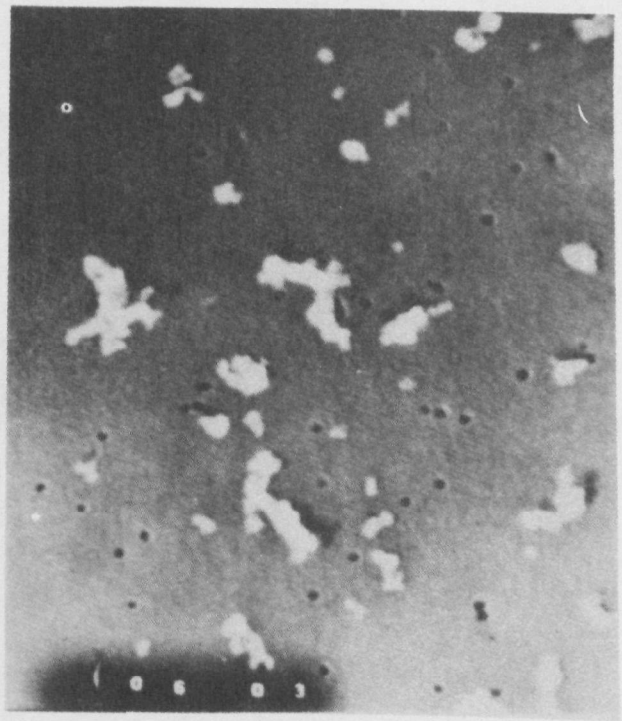


Fig. 11 - Particle shape factor, JT9D engine, climb out, JET A

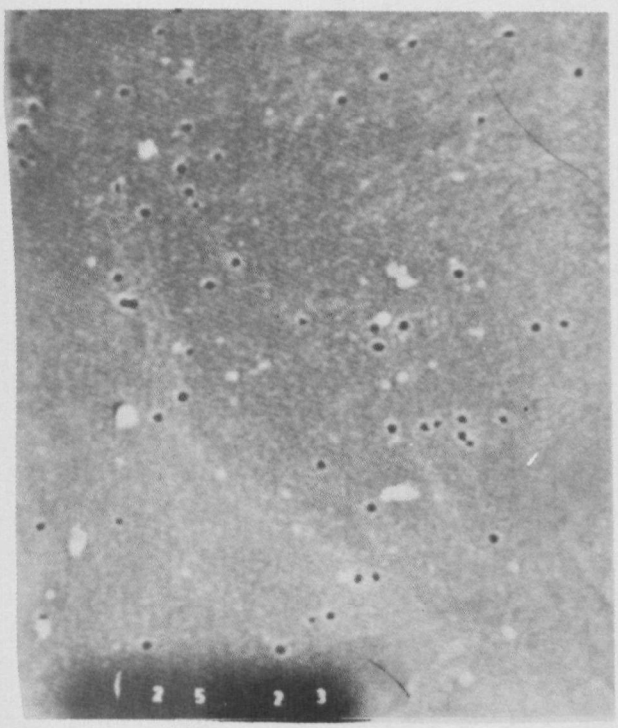


Idle

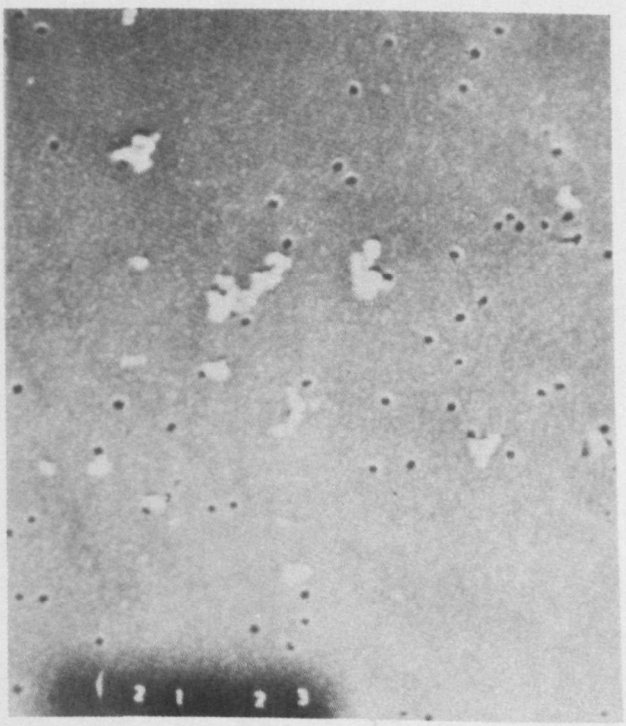


Cruise

Fig. 12 Particulate Emissions, JT8D engine, JET A fuel



Idle



Climb-out

Fig. 13 Particulate Emissions, JT9D engine, JET A fuel

MEASUREMENT OF FUGITIVE DUST EMISSIONS FROM HAUL ROADS

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ABSTRACT

This paper presents the results of a field testing program used to develop emission factors for dust generated by heavy-duty truck traffic on unpaved roads. Emission tests were performed on roads traveled by haul trucks within an open pit taconite mine and within two integrated iron and steel plants. Both chemically treated and untreated roads were tested. The basic field measurements consisted of fugitive dust exposure profiles and particle size distributions using vertically oriented arrays of isokinetic samplers such that the total passage of airborne dust downwind of the test road segment could be determined.

Test results are used to extend the applicability of the emission factor equation previously developed for light- and medium-duty truck traffic on unpaved roads associated with agriculture and industry. Correction terms which appear in the equation account for the dependence of emissions on vehicle weight, vehicle speed, and the fraction of suspendable fines (silt) in the road surface material. Based on an expanded data set of 24 tests, including vehicle weights up to 157 tons, the unmodified equation predicts measured emission factors with a relative standard deviation of predictive error equal to 26.1%. Test results suggest that inclusion of an additional correction term involving number of wheels per vehicle would lower the predictive error. Particle size data indicate that the fine particle ($< 5\ \mu m$) mass fraction of the suspended particulate ($< 30\ \mu m$) emissions is approximately 35%, independent of vehicle weight and road surface composition. Finally, limited testing of chemical dust suppressants shows high initial control efficiency (exceeding 90%) which decays with road usage.

INTRODUCTION

Until recently, the national effort to control industrial sources of air pollution has focused on emissions discharged from stacks, ducts, or flues and carried to the point of discharge in confined flow streams. Control strategies have been based on the assumption that the primary air quality impact of industrial operations results from the discharge of air pollution from conventional ducted sources.

However, failure to achieve the air quality improvements anticipated from the control of ducted emissions has spurred a detailed reexamination of the industrial air pollution problem. Evidence is mounting which indicates that fugitive (nonducted) emissions contribute substantially to the air quality impact of industrial operations and, in certain industries, may exceed the effects of stack emissions.

Industrial sources of fugitive particulate emissions may be divided into two classes--process sources and open dust sources. Process sources are fully or partially enclosed operations from which emissions escape into the workplace environment and/or the ambient air. Examples of process sources are crushers, sintering machines, and metallurgical furnaces. Open dust sources include those sources, such as raw material storage piles, from which emissions are generated by the forces of wind and machinery acting on exposed aggregate materials.

The travel of heavy vehicles on unpaved roads is the category of open dust sources to which this paper is addressed. Truck traffic on haul roads is a major source of fugitive particulate emissions associated with the surface mining and processing of both metallic and nonmetallic minerals. Quantification and characterization of dust emissions from haul roads are necessary in order to assess control needs and to develop cost-effective control measures, as necessary.

QUANTIFICATION OF OPEN DUST SOURCES

Fugitive emissions from open dust sources are especially difficult to characterize for the following reasons:

1. Emission rates have a high degree of temporal variability.

2. Emissions are discharged from a wide variety of source configurations.

3. Emissions are comprised of a wide range of particle sizes including coarse particles which deposit immediately adjacent to the source.

The scheme for quantification of emission factors for open dust sources must effectively deal with these complications.

In 1972, Midwest Research Institute (MRI) (1) initiated a field testing program to develop emission factors for four major categories of fugitive dust sources: unpaved roads, agricultural tilling, aggregate storage piles, and heavy construction operations. Because the emission factors were to be applicable on a national basis, an analysis of the physical principles of fugitive dust generation was performed to ascertain the parameters which would cause emissions to vary from one location to another. These parameters were found to be grouped into three categories:

1. Measures of source activity or energy expended (for example, the speed and weight of a vehicle traveling on an unpaved road).

2. Properties of the material being disturbed (for example, the content of silt in the surface material on an unpaved road).

3. Climatic parameters (for example, number of precipitation-free days per year on which emissions tend to be at a maximum).

By constructing the emission factors as mathematical equations with multiplicative correction terms, the factors became applicable to a range of source conditions limited only by the extent of the program of experimental verification.

The use of the silt content as a measure of the dust generation potential of a material acted on by the forces of wind and/or machinery was an important step in extending the applicability of the emission factor equations to the wide variety of aggregate materials of industrial importance. The upper size limit of silt particles (75 μm in diameter) is the smallest particle size for which size analysis by dry sieving is practical; and this particle size is also a reasonable upper limit for particulates which can become airborne. Analysis of atmospheric samples of fugitive dust indicates a consistency in size distribution so that particles in specific size ranges exhibit fairly constant mass ratios (1,2).

In order to quantify source-specific emission factors, MRI developed the "exposure profiling" technique (3), utilizing the isokinetic profiling concept which is the basis for conventional source testing. Exposure profiling consists of the direct measurement of the passage of airborne pollutant immediately downwind of the source by means of simultaneous multipoint sampling over the effective cross section of the fugitive emissions plume. This technique uses a mass-balance calculation scheme similar to Environmental Protection

Agency (EPA) Method 5 stack testing rather than requiring indirect calculation through the application of a generalized atmospheric dispersion model.

Specifically, the exposure profiling method was used to develop emission factors for: (a) light-duty vehicular traffic on unpaved (dirt and gravel) roads (1); (b) agricultural tilling utilizing a one-way disk plow and a sweep-type plow (1); (c) load-out of crushed limestone utilizing a 1.75 cu yard loader (1); and (d) vehicular traffic on paved urban roadways (2). In order to extend the applicability of the emissions factor equations to sources involving larger scale materials handling equipment, the following sources were tested at two integrated iron and steel plants (Test Series A and E) (4): (a) light-duty vehicular traffic on industrial unpaved roads; (b) heavy-duty vehicular traffic on unpaved roads; (c) mixed vehicular traffic on industrial paved roads; (d) mobile stacking of lump iron ore; (e) mobile stacking of pelletized iron ore; and (f) loadout of processed slag into a truck with a front-end loader. All sources were tested under dry road conditions (i.e., daytime periods at least 3 days subsequent to a precipitation occurrence) so that worst case emissions could be determined and used as a starting point for projecting annual emissions.

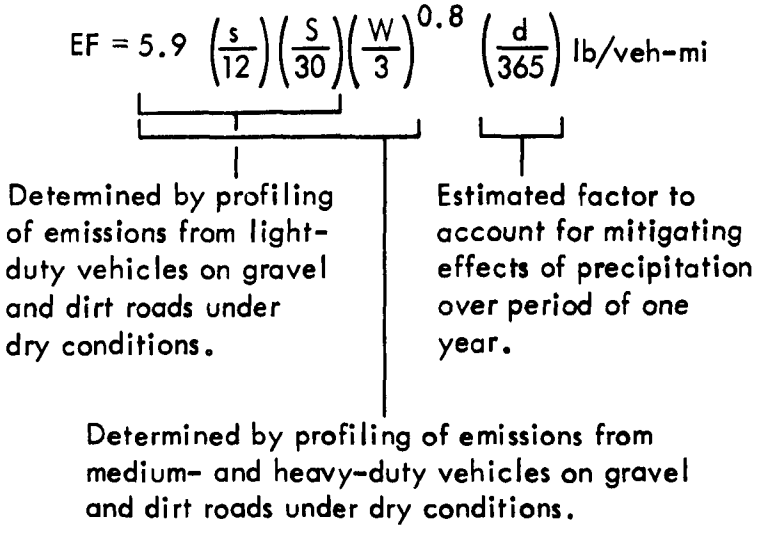
EXPANDED TESTING OF UNPAVED ROAD DUST EMISSIONS

Figure 1 presents the emission factor equation previously developed for dust entrainment by vehicles traveling on unpaved roads. Although this equation was found to have high predictive reliability for dry road conditions (relative standard deviation of prediction error equal to 11.2% of the mean measured value), the data base was limited to 11 tests. Moreover, only two tests had been conducted for vehicle weights representative of haul trucks used in mining and processing of minerals.

Therefore, in order to strengthen the data base for the predictive emission factor equation, 16 additional tests of uncontrolled emissions from unpaved roads were performed during the summer of 1978. Seven of these tests (Series I) were conducted at an open pit taconite mine and the remaining tests (Series F and G) at two integrated iron and steel plants. In addition, five tests of traffic on roads treated with chemical dusts were performed.

As in the previous tests, the primary tool for measuring fugitive dust generated from vehicular traffic on unpaved roads was the MRI exposure profiler. A vertical line grid of samplers (see Figure 2) was used for measurement of dust emissions. At all times the MRI exposure profiler was positioned within 5 m of the downwind edge of the test road with air samplers covering the effective cross section of the fugitive dust plume.

Other equipment utilized in the testing included: (a) cascade impactors with cyclone preseparators for particle sizing, (b) high-volume air samplers for determining upwind particulate concentrations, and (c) recording wind



where: EF = suspended particulate emissions (lb/veh-mi)
s = silt content of road surface material (%)
S = average vehicle speed (mph)
W = average vehicle weight (tons)
d = dry days per year

Figure 1. Predictive emission factor equation for vehicular traffic on unpaved roads.

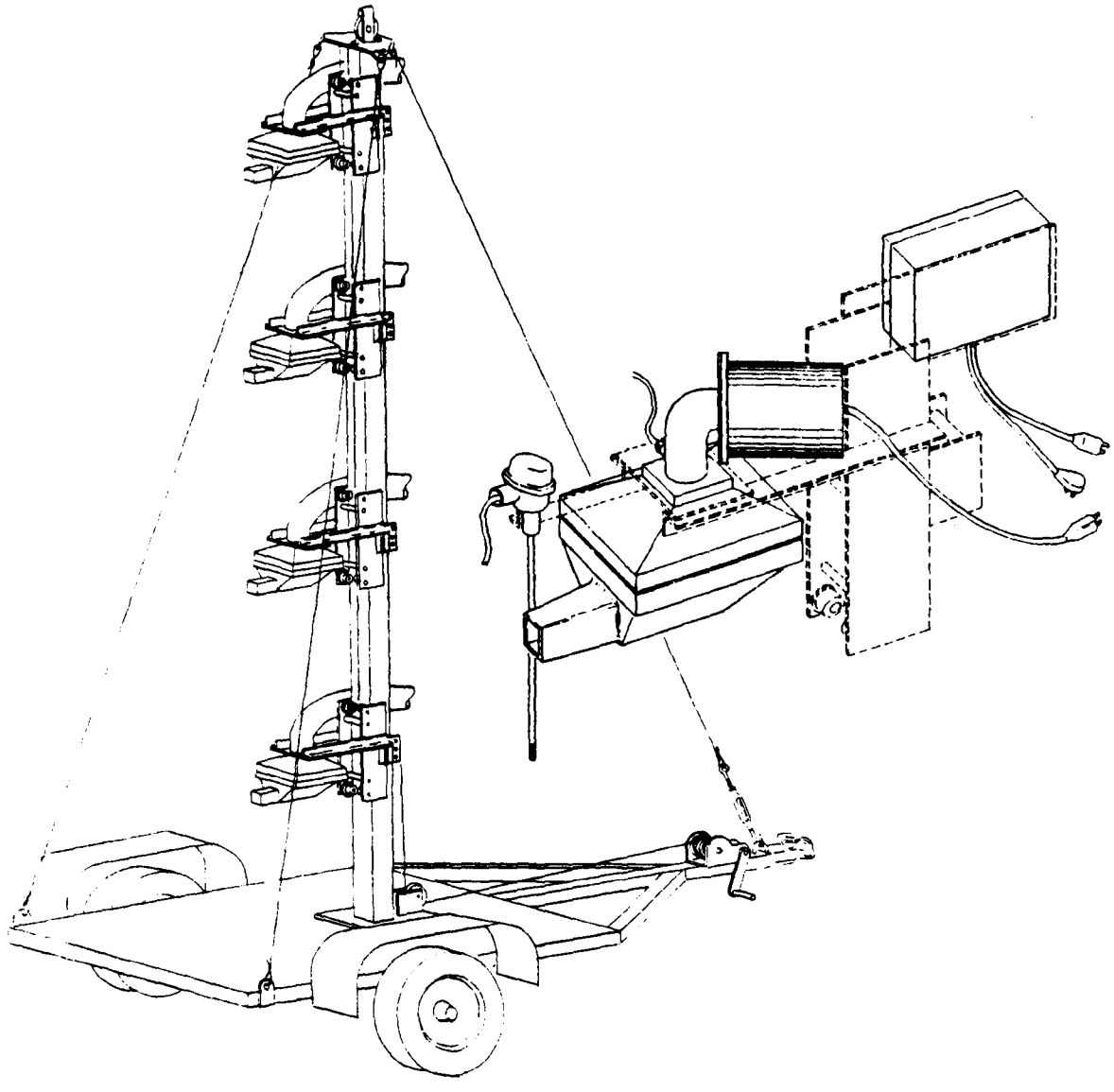


Figure 2. MRI exposure profiler.

instruments utilized to determine mean wind speed and direction for adjusting the MRI exposure profiler to isokinetic sampling conditions. A detailed description of the testing methodology is provided elsewhere (4).

In order to determine the properties of road surface aggregate being disturbed by the action of moving vehicles, representative samples of the surface materials were obtained for analysis in the laboratory. Unpaved road test segments were sampled by removing loose material (by means of broom sweeping) from lateral strips of road surface extending across the traveled portion. Moisture contents of samples were determined in the laboratory by weight loss after oven drying at 110°C, and texture was determined by standard dry sieving techniques.

In addition to the silt content of the road surface material, the emission factor equation (Figure 1) requires data on vehicle speed and weight, averaged over the vehicle passes (approximately 50) accumulated during a test. During each test, the speeds of vehicles passing the sampling station were estimated by timing over a known travel distance. Estimates of vehicle weights were obtained from plant personnel. In some tests, the vehicle passes sampled were dominated by controlled test vehicles traveling at preselected speeds.

TEST RESULTS

The cumulative results of the field testing of vehicular traffic on unpaved roads are provided in Table 1, which presents the emission factors for suspended particulate (smaller than 30 μm in Stokes diameter) and for fine particulate (smaller than 5 μm in Stokes diameter), along with surface material characteristics. The upper size limit of 30 μm for suspended particulate is the approximate effective cutoff diameter for capture of fugitive dust by a standard high-volume particulate sampler (based on a typical particle density of 2 to 2.5 g/cu cm) (1). Both Table 1 and Figure 3 compare actual emission factors with predicted values.

Excluding run Nos. I-1, I-7, and I-8 for the reasons given in the footnotes to Table 1, the relative standard deviation of prediction errors is 26.1% of the mean measured emission factor. For Test Series E and G, the emission factor equation consistently underpredicts the measured factors. This appears to be due to the effect of 10- and 18-wheel trucks, which comprised a substantial number of the passes in those tests. In all other test series, the vehicle mix was dominated by four- and/or six-wheel vehicles.

As indicated in Figure 4, there is no apparent relation between the fraction of the emissions consisting of fine particles and the average vehicle weight or the road surface composition. The average value is approximately 35% by weight.

TABLE 1. PREDICTED VERSUS ACTUAL EMISSIONS--UNPAVED ROADS

Test No.	Road surface: Type	Road surface: Silt (%)	Mean vehicle speed (mph)	Mean vehicle weight (tons)	Emission factor[a] (lb/vehicle mile): Predicted[b]	Emission factor[a] (lb/vehicle mile): Actual	Percent difference[c]	Predicted / Actual
R-1	Crushed limestone	12	30	3	5.9	6.0	-2	0.98
R-2		13	30	3	6.4	6.8	-6	0.94
R-3		13	40	3	8.5	7.9	8	1.08
R-8	Dirt	20	30	3	9.8	8.1	21	1.21
R-10		5	40	3	3.3	3.9	-15	0.85
R-13		68	30	3	33	32	3	1.03
A-14	Crushed slag	4.8	30	70	29	27	7	1.07
A-15		4.8	30	70	29	29	0	1.00
E-1	Dirt	8.7	14	34	14	17	-17	0.82
E-2		8.7	16	34	16	16	0	1.00
E-3		8.7	16	23	12	19	-37	0.63
F-21	Dirt/crushed slag	9.0	15	3	2.2	3.2	-31	0.69
F-22		9.0	15	3	2.2	1.7	29	1.29
F-23		9.0	15	4	2.8	2.4	17	1.17
F-24	Dirt/slag (Coherex®)	0.03	15	3	[g]	0.073	-	-
F-25		0.02	15	3	[g]	0.39	-	-
G-27	Crushed slag	5.3	22	17	7.7	13.7	-44	0.56
G-28		5.3	23	12	6.1	9.0	-32	0.68
G-29		5.3	24	9	5.0	6.8	-26	0.74
G-30		4.3	25	14	6.0	11.6	-48	0.52
G-31		4.3	29	8	4.5	6.6	-32	0.68
G-32		4.3	22	30	9.8	21.8	-55	0.45
I-1	Crushed rock and glacial till	4.7	15	67	13.9	6.4[d]	117	2.17
I-2		4.7	15	67	13.9	9.9	40	1.40
I-3		4.7	15	67	13.9	20.3	-32	0.68
I-4		4.7	15	157	27.4	24.8	10	1.10
I-5		4.7	15	157	27.4	30.7	-11	0.89
I-7	Crushed rock (taconite/waste)	6.1	13.5	118	25.5	16.1[e]	58	1.58
I-8		6.1	13.5	117	25.3	14.6[e]	73	1.73
I-9	Crushed rock (TREX)	1.3	13	110	[g]	1.3	-	-
I-10		1.5[f]	13	112	[g]	2.6	-	-
I-11		1.8	14	127	[g]	3.3	-	-

a/ Particles smaller than 30 μm in diameter.
b/ Based on MRI emission factor.
c/ 100 x (predicted-actual)/actual.
d/ Test Series I-1 through I-5 performed on previously inactive road.
e/ Tests performed on day following 2 days of rain totaling 1.13 in.
f/ Assumed value.
g/ Equation not applicable.

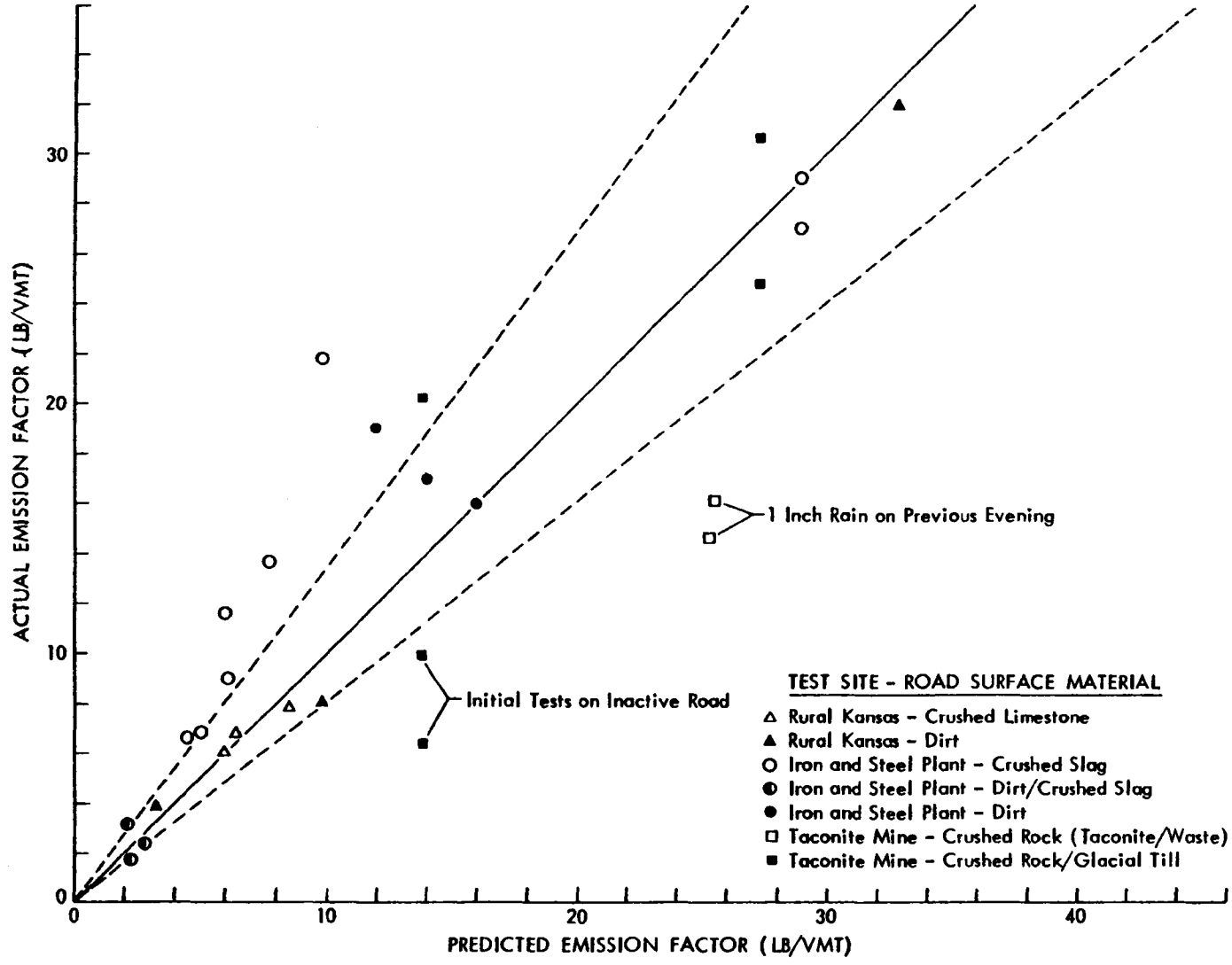


Figure 3. Comparison of predicted and actual emissions--untreated roads.

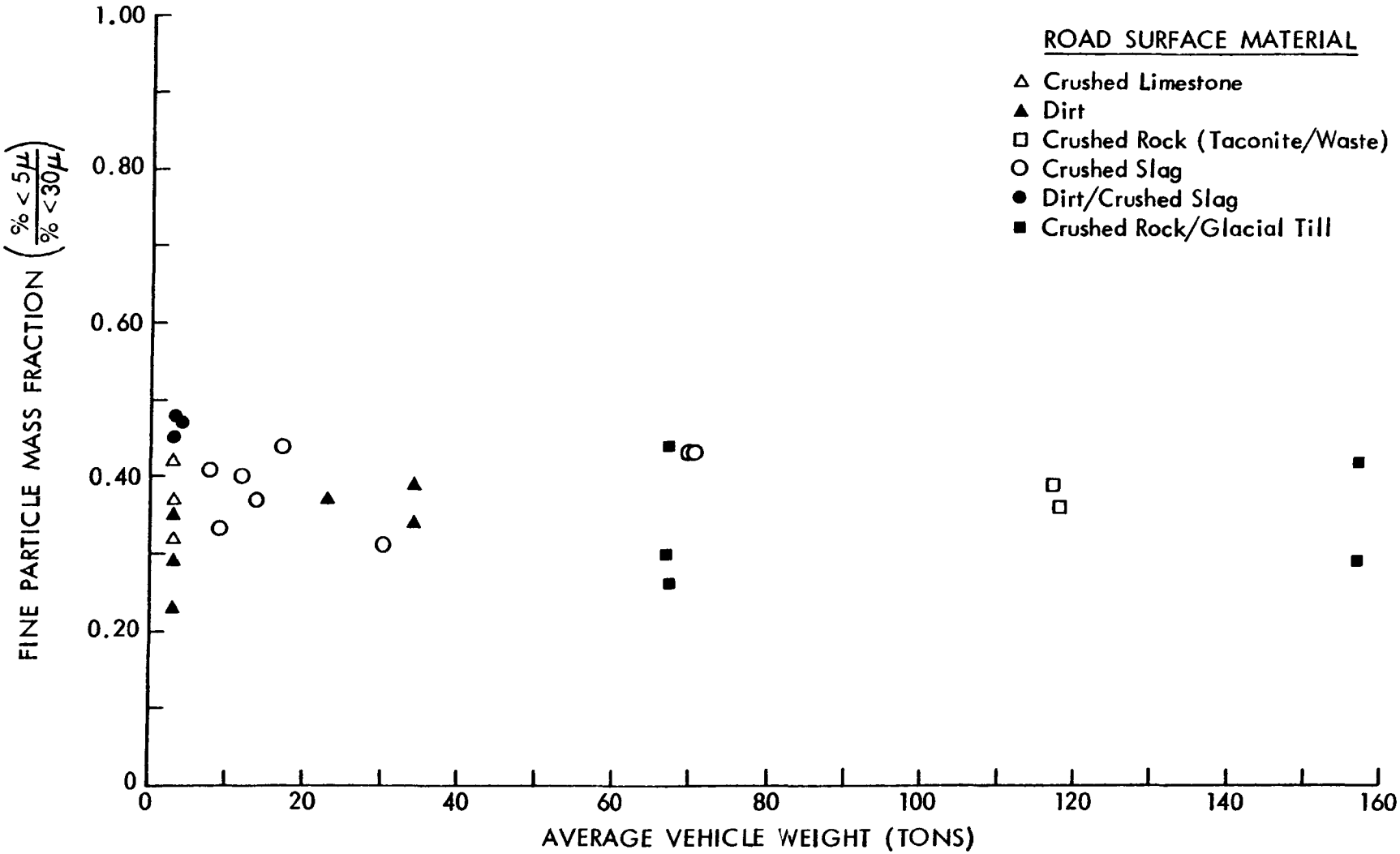


Figure 4. Fine particle fractions of TSP emissions.

Analysis of parameters affecting the atmospheric transport of fugitive dust indicates that only the portion smaller than about 5 μm in diameter will be transported over distances greater than a few kilometers from the source (5).

As stated above, limited testing of the effects of chemical dust suppressants was also conducted. At the taconite mine site, TREX (ammonium lignin sulfonate--a water soluble by-product of papermaking) was applied to the waste rock aggregate comprising the surface of a haul road. A 20 to 25% solution of TREX in water was sprayed on the road at a rate of 0.08 gal/sq yard of road surface. The other chemical dust suppressant tested was Coherex® (a petroleum-based emulsion), which was used to treat a dirt/slag surfaced service road traveled by light- and medium-duty vehicles at an integrated iron and steel plant. Coherex® was applied at 10% strength in water.

Figure 5 shows a plot of measured dust control efficiency as a function of the number of vehicle passes following application of the road dust suppressant. Control efficiency was calculated by comparing controlled emissions with uncontrolled emissions measured prior to road surface treatment. As indicated, the effectiveness of road dust suppressants is initially high but begins to decay with road usage. According to taconite mine personnel, the binding effect of TREX can be partially restored by the addition of water to the road surface. It should also be noted that the apparent performance of Coherex® was negatively affected by tracking of material from the untreated road surface connected to the 200-ft treated segment.

With regard to the effects of natural mitigation of road dust emissions, the final term in the emission factor equation for traffic on unpaved roads (Figure 1) is used to reduce emissions from dry conditions to annual average conditions. The simple assumption is made that emissions are negligible on days with measurable precipitation and are at a maximum on the rest of the days. Obviously, neither assumption is defendable alone; but there is a reasonable balancing effect. On the one hand, 0.01 in. of rain would have a negligible effect in reducing emissions on an otherwise dry, sunny day. On the other hand, even on dry days, emissions during early morning hours are reduced because of overnight condensation and upward migration of subsurface moisture; and on cloudy, humid days, road surface material tends to retain moisture. Further natural mitigation occurs because of snow cover and frozen surface conditions. In any case, further experimentation is needed to verify and/or refine this factor.

CONCLUSIONS

Based on an expanded data set of 24 tests, the MRI emission factor equation for traffic-entrained dust from unpaved roads predicts measured emission factors with a relative standard deviation of the prediction error equal to 26.1% of the mean measured value. It appears that an additional adjustment term related to the average number of wheels per vehicle would

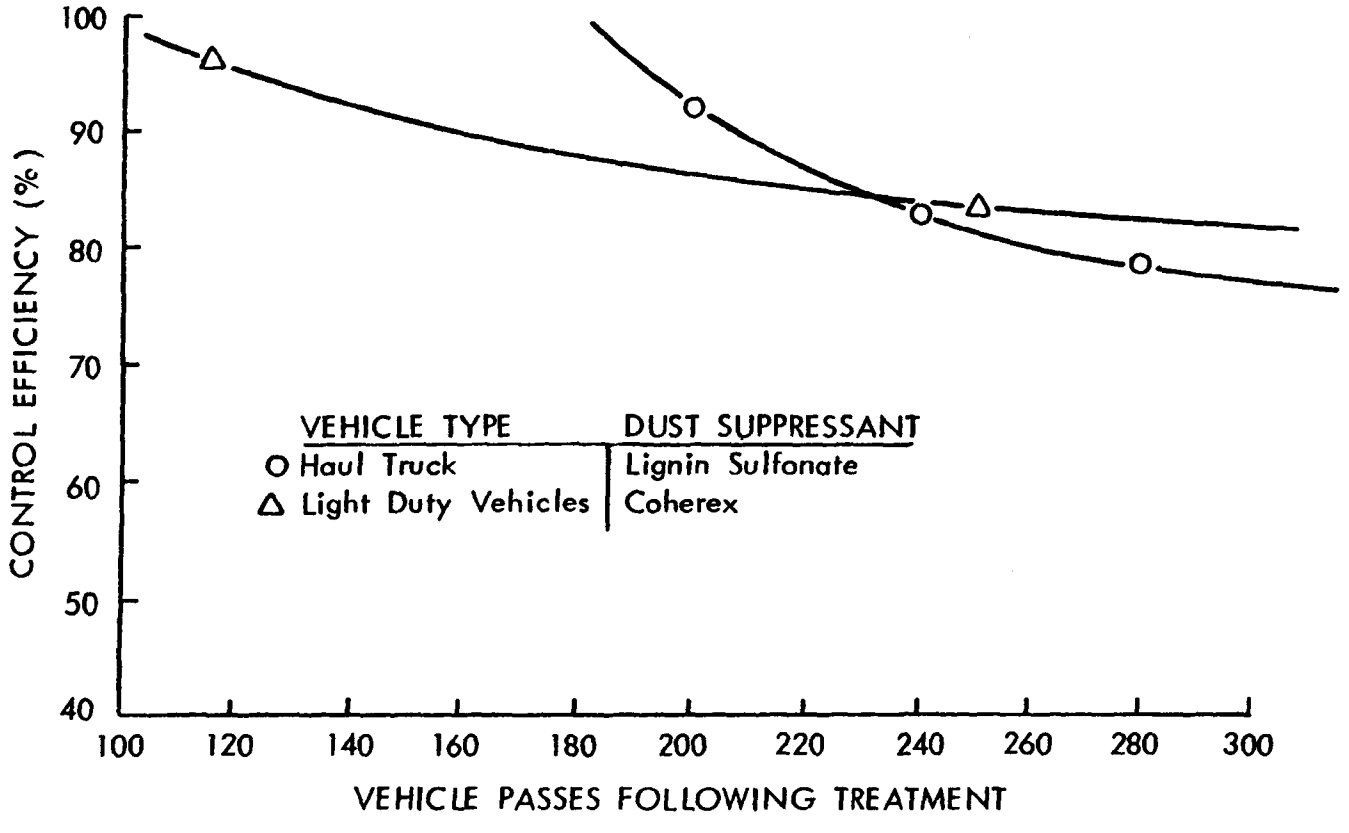


Figure 5. Effectiveness of road dust suppressants.

reduce the mean prediction error because there is a clear tendency to underpredict measured emission factors when the road is traveled by a substantial portion of 10- and/or 18-wheel vehicles rather than 4- and 6-wheel vehicles.

Approximately 35% of measured road dust emissions in the suspended particulate size range (particles smaller than 30 μm in diameter) consist of fine particles (particles smaller than 5 μm in diameter), which have the potential for transport over distances greater than a few kilometers from the source. This fraction appears to be independent of average vehicle weight and road surface composition.

Limited testing of chemical dust suppressants for industrial unpaved roads indicates a high initial control efficiency (exceeding 90%), which decays by more than 10% with the passage of 200 to 300 vehicles. Consistent with the emission factor equation, the lowering of emissions is reflected by the reduced silt content of the road surface material after the application of chemical dust suppressants. Additional testing is needed to better quantify the performance of road dust suppressants. Testing is also needed to verify and/or refine the emission factor adjustment term which accounts for climatic mitigation.

ACKNOWLEDGEMENT

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FACTORS FOR CONVERSION TO METRIC UNITS

1 kilogram = 2.2 lb

1 kilometer = 0.62 miles

1 metric ton = 1.1 short tons

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DEVELOPMENT OF MEASUREMENT METHODOLOGY FOR EVALUATING FUGITIVE PARTICULATE EMISSIONS

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ABSTRACT

A research project is under way to develop and demonstrate a method for evaluating fugitive particulate emissions from an industrial plant, by measuring the flow of total particulate pollution in the atmosphere downwind of it. Using both surface sampling and remote-sensing techniques for sampling above the surface level, the method is to be capable of measuring the spatial distribution, absolute concentration, and velocity of the particulate cloud issuing from the plant.

The objective of the current study is to demonstrate the proposed method under field conditions, but in circumstances that simplify and control the variables of the general problem. A field test was recently completed that used an aerosol generator to emit a controlled aerosol stream into the atmosphere from a point source with known particulate feed rates. A mobile lidar system was used to make a series of remote observations of the cross-plume particulate distribution 300 to 500 m downwind of the source.

Data presented show that the cross-plume integrated backscatter is responsive to the particulate feed rate. The problem of deriving an absolute mass emission rate from such backscatter traverses of a plume is discussed.

I INTRODUCTION

The total source strength of pollution emitted by industrial plants is the aggregate of all diffuse and minor specific emissions as well as major identifiable point sources. Therefore, for many plants, the measurement of individual emissions from the multiplicity of sources is neither economical nor practical.

The only feasible approach is to measure, as accurately as possible, the concentration throughout a cross section of the downwind "plume" of the combined fugitive emissions, to integrate these, and, from a measurement of the integrated wind velocity through the plane of the cross section, to calculate the pollutant mass flow. For particulate fugitive emissions, any deposition or transformation in the intervening distance must also be considered, to derive an estimate of the total source strength.

The problems of accomplishing such measurements with existing technology are many. Specifically, with in-situ samplers it is virtually impossible to characterize adequately the concentration of particles throughout the total cross section of the plume, or to relate any measurements made to the total envelope of the plume, or to determine its extent. This is especially the case above the surface, because of the extended and variable nature of the multiple sources of fugitive emissions.

Lidar (laser radar) observation fulfills, as no other method does, the requirement for delineating the spatial distribution of elevated particulate pollution plumes, and also for readily distinguishing between background pollution and pollution from the plant being studied. While there are limitations and difficulties in using lidar backscatter measurements for determining absolute particulate concentrations, it is possible to evaluate, with useful accuracy, the near-instantaneous mass distribution of particulate material within a selected cross section or envelope. It is thus possible to derive series of such cross sections in time, and to relate these to a measurement of mean wind velocity, to derive an estimate of mass flow and hence of source strength. (Several lidar techniques have been demonstrated for wind measurement, but these are not currently at an operational stage and simpler, more conventional methods of wind measurement are still indicated.) Finally, eye safety from laser systems used over industrial sites must be considered. In principle, however, it is apparent that lidar offers a unique potential for eventually providing quantitative measurement of fugitive particulate emissions.

This paper presents results from an initial attempt to demonstrate, using presently available research equipment, the capabilities of lidar for measuring particulate fugitive emissions, and recommends several research areas requiring further consideration to fully exploit the lidar technique for this purpose.

II THE LIDAR TECHNIQUE

Lidar (Light Detection and Ranging) uses laser energy in radar fashion to effectively observe remote atmospheric constituents. The lidar system being used to develop methodologies for fugitive dust measurement is SRI International's Mark IX mobile system, designed for use with atmospheric research programs. Figure 1 presents views of the lidar system, a block diagram, and a list of specifications. The lidar is installed within an 18-ft van complete with real-time data processing and display capabilities as well as with power-generating equipment, enabling operation at any site.

Figure 2 is an example of an intensity-modulated TV display depicting the cross-plume aerosol structure observed by scanning the lidar in elevation. The brightness of this display is proportional to the logarithm of the backscattered light detected by the lidar system. Clearly, lidar can be used to map relative aerosol density distributions over remote distances. Computer-generated vertical-concentration profiles are plotted on the cross section for locations indicated by the cursor marks drawn above the plume return. Similarly, the backscattered data can be spatially integrated to determine a relative cross-plume density. Light that is elastically backscattered from suspended atmospheric particulate matter, as a function of pulse-transmission time, is quantitatively related to (clear air and plume) optical parameters along the observed path according to the lidar (radar) equation

$$P = K E \beta R^{-2} T^2 \qquad (1)$$

where

P = received power from a scattering volume defined by the laser pulse

K = lidar calibration constant

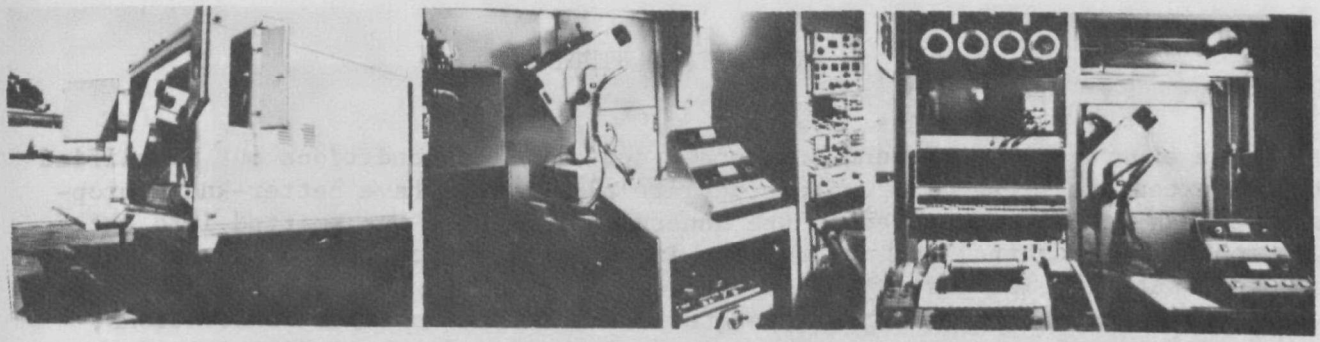
E = transmitted power

β = backscatter coefficient of the scattering volume

R = range from lidar of the scattering volume

T = transmission to the scattering volume

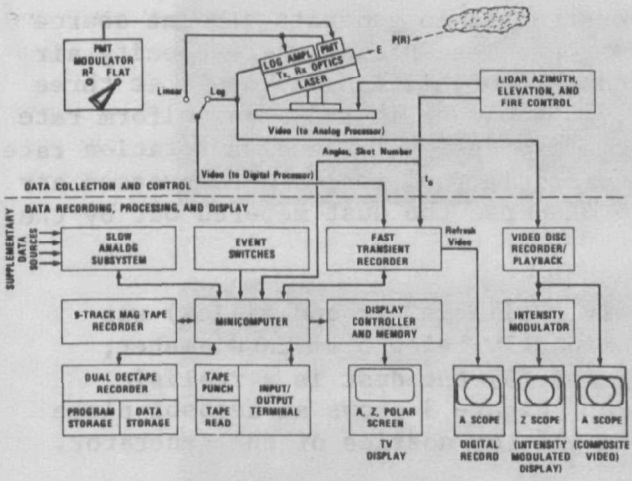
For relatively clear air, the attenuation term can normally be ignored ($T^2 = 1$) and P provides information on the distribution of particulate material in the atmosphere, as shown in Figure 2. However, the evaluation of physical-density terms (such as mass concentration) requires relating the optical parameter β to the density. Unfortunately, backscatter-to-concentration ratios are dependent on the distributions of particle size, composition, and shape. Before these factors, and the ways in which they can limit the accuracy of quantitative evaluations of plume density, can be considered, the lidar observations must be shown to respond in a predictable manner to increases of plume density for a given aerosol type. An experimental field program using presently available research equipment has been conducted for this purpose. The design of the field program is discussed below, followed by some of the preliminary results.



(a) MARK IX LIDAR VAN (b) ANALOG DATA AND FIRE CONTROL ELECTRONICS (c) DIGITAL DATA ELECTRONICS AND TV DISPLAY

EXTERIOR AND INTERIOR VIEWS OF THE LIDAR VAN

BLOCK DIAGRAM OF THE LIDAR SYSTEM



LIDAR SPECIFICATIONS

TRANSMITTER

6943Å Wavelength
0.5 mrad Beamwidth
1.0 J Pulse Energy
30 ns Pulse Length
60 ppm Maximum PRF

RECEIVER

6 inch Newtonian
1 to 5 mrad Field of View
5Å Predetection Filter
RCA 7265 PMT Detector
4-decade, 35-MHz Logarithmic Amplifier, Inverse-range-squared or step-function PMT modulation.

DATA SYSTEMS

Analog video disc recording (4.5 MHz) with A-scope and Z-scope real-time displays. Digital magnetic tape (data and programs) recording (25 MHz) with computer processing and real-time TV display (512 x 256 x 4 bit) of processed data.

MOUNT

Automatic azimuth and elevation fire and scan with 0.1° minimum resolution. Automatic reset. Mechanical safety stops.

FIGURE 1 SRI INTERNATIONAL'S MARK IX MOBILE LIDAR SYSTEM

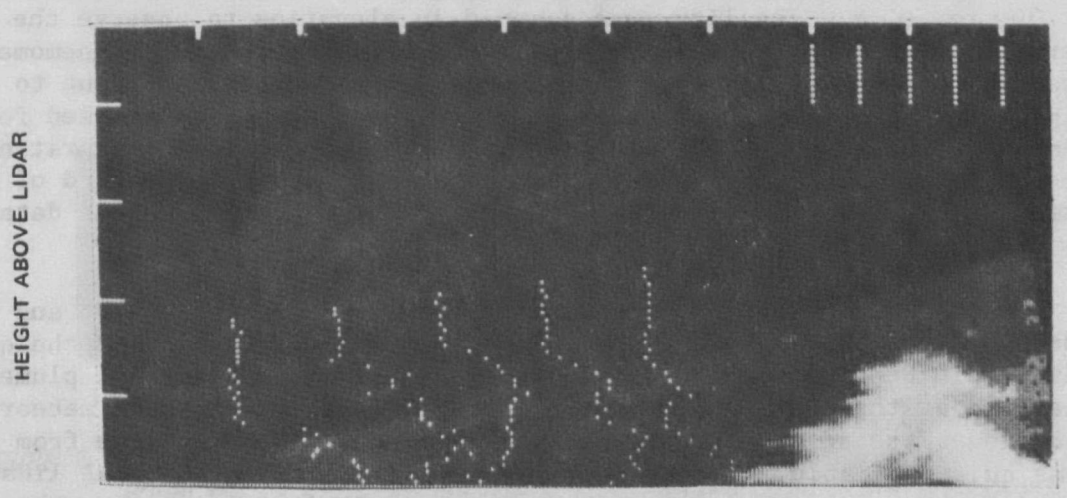


FIGURE 2 EXAMPLE OF COMPUTER-GENERATED PROFILES OF VERTICAL PLUME DENSITY Lidar is located at lower left corner. The height and distance scale is 75 m/div. Vertical concentrations of the plume (relative to clear air, with a scale of 10 dB/div) are plotted at the lower left and the horizontal position associated with each profile is plotted in the upper right.

III FIELD STUDY

The experiments were conducted under actual field conditions but simplified to the extent that the observed particulate plume would have better-known properties than could be expected in the general case. Dust was emitted into the atmosphere in a controlled manner. The intent was to show that observed backscatter spatially integrated across the aerosol plume downwind of the source is well correlated with the source strength. In addition to lidar observations, conventional wind measurements were made to determine downward dilution of the particulate plume; near-surface particulate sampling was conducted to investigate possible approaches for measuring actual fugitive emissions with unknown particulate properties.

An aerosol source was designed and constructed to generate a point source of dust by feeding a fine powder of known properties into a sonic-velocity air stream pointing downwind. The source is capable of emitting aerosols at three levels--1, 3, and 10 m above ground level. The powder is fed at a uniform rate by means of a grooved-disk feeder of proprietary design. The disk rotation rate can be continuously varied to control the particle feed rate. A compressed air stream of 40 ft^3/min at 52 psi was used to disperse the dust metered out by the feeder.

For the dust, Min-U-Sil (a commercially available* ground silica) was chosen because (1) the particles are predominantly below 5 μm in diameter; (2) the material is available in quantity; and (3) the dust is a realisic example of material encountered in practice. Figure 3 shows an aerosol plume generated by dust emissions from the bottom and top nozzles of the generator.

The Mark IX lidar van was positioned 300 to 500 m from the source so that the laser beam would intersect the plume perpendicularly at a downwind distance of about 300 to 500 m. The lidar was scanned in elevation to observe the cross-plume aerosol distribution, similar to that shown in Figure 2. An anemometer was installed on the Mark IX lidar van and its signal output was input to the lidar digital system so that both wind speed and direction were sampled for each lidar observation (firing). In addition, the output of an integrating nephelometer was similarly sampled. These data and the digital record of the backscatter signature were stored on magnetic tape for use during the data analysis program.

An initial data collection program was conducted during May 1978 and a second data collection period using slightly improved experimental techniques was conducted in September 1978. For each experimental run, aerosol plumes were generated at three different particulate feed rates. For some meteorological conditions, the cross-plume aerosol density at a given distance from the source was quite variable. Therefore, a series of about ten vertical lidar cross sections were collected for each particulate feed rate, so that the value of the time-integrated cross-plume backscatter could be determined. Preliminary results obtained from the field program are presented in the next section.

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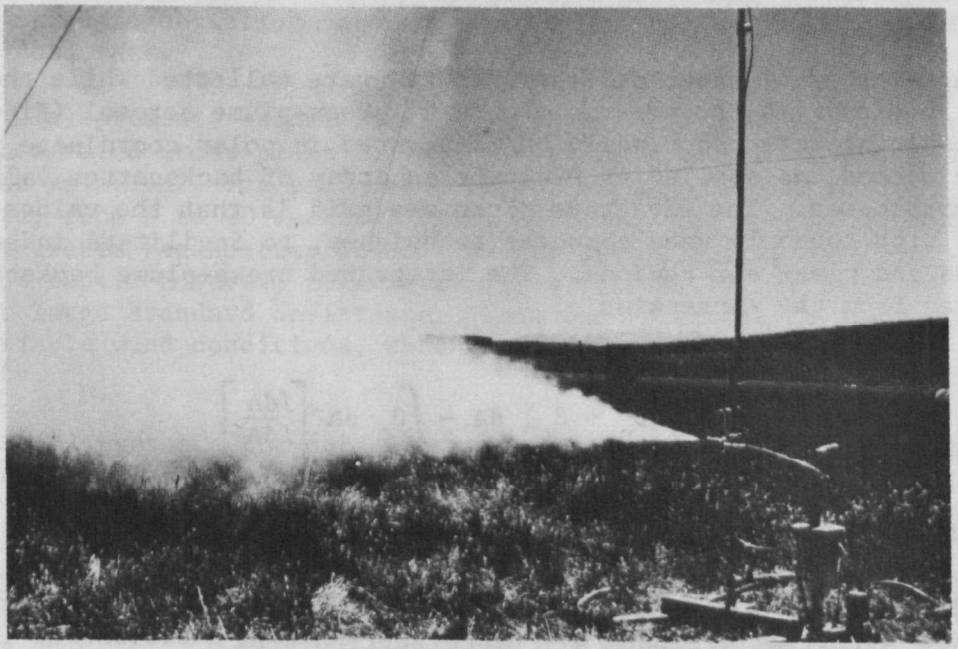
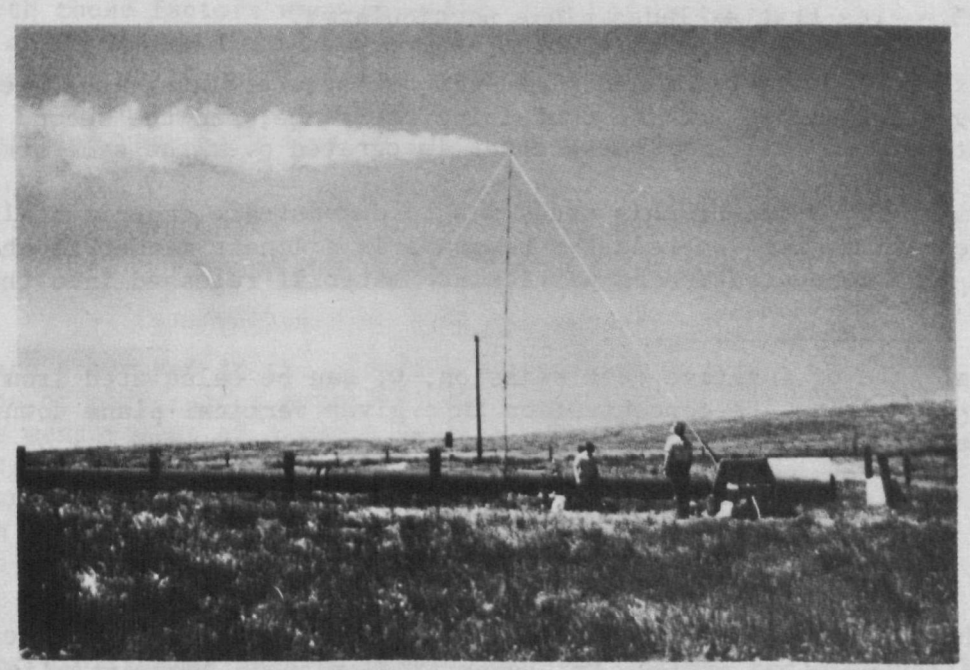
*Pennsylvania Glass Sand Corp., Pittsburgh, PA.



(a)



(b)

FIGURE 3 CONTROLLED DUST PLUMES GENERATED TO REPRESENT FUGITIVE EMISSION SOURCES

(a) Emission 1 m above surface

(b) Emission 10 m above surface

IV PRELIMINARY RESULTS

The range-dependent backscatter signatures were collected while the lidar was scanned in elevation, to observe the total cross-plume aerosol (Figure 2). While the resulting data can readily be integrated in polar coordinate form, an objective method was applied to evaluate an array of backscatter values in Cartesian coordinates. The advantage of an x-y grid is that the values can be displayed with commonly used computer techniques, to facilitate indentification of plume and clear air regions. The integrated cross-plume backscatter was determined from the expression

$$\int \beta_p \, dA = \int (\beta_p + \beta_c) \, dA - \int \beta_c \, dA' \left[\frac{\int dA}{\int dA'} \right] \tag{2}$$

where β_p = plume backscatter coefficient [see Equation (1)]

β_c = clear-air backscatter coefficient

A = area that includes all plume particulates

A' = area that excludes plume particulates.

The first term represents plume and clear-air backscatter integrated over an area of the vertical cross section containing the plume, and the second term is an estimate of the clear-air backscatter integrated over the same area.

The primary objective of this study was to demonstrate experimentally that the integrated cross-plume backscatter responds in a linear manner to changes in the total mass concentration of particulate material released into the atmosphere.

The total rate of fugitive dust emission, w, can be calculated from the distribution of particulate concentration in a given vertical plane downwind of the source by

$$w = \int c \, u \sin\Theta \, dA \tag{3}$$

where c is the concentration in the area segment dA at which the wind velocity is u and is oriented at an angle Θ to the vertical plane. Assuming c is proportional to the lidar backscatter coefficient ($c = K\beta$), and assuming a constant wind speed and direction at all parts of the vertical plane observed by the lidar, we have

$$w = K \, u \sin\Theta \int \beta_p \, dA \tag{4}$$

For each observation (vertical cross section), the backscatter records were processed in terms of an array of backscatter values for identifying appropriate clear-air and plume regions, to evaluate Equation (2). The integrated

cross-plume backscatter values were computed and statistically analyzed for the average and standard deviation for each set of observations made at a fixed particulate feed rate.

Figure 4(a) presents experimental results for two observational periods, using the integrated cross-plume backscatter as an estimate of the rate of fugitive dust emission. For each case, three emission rates were used, and for each rate, from 8 to 20 vertical cross sections were made. The data show that the integrated backscatter generally increases linearly with the particulate emission rate. However, relatively large scatter of the data points occur--i.e., large standard deviations. This can be explained partly on the basis of variable wind conditions, which were ignored in this estimate of emission rate.

Figure 4(b) presents the data shown in Figure 4(a) after wind-speed (but not direction) corrections were applied; these were based on anemometer measurements made at the lidar site during the data-collection period. The wind corrections improve the linear relationship between the lidar observations and the particulate feed rates (especially for the second case). However, the standard deviations remain about the same. This variability is thought to result partly from small-scale, uncontrolled variations in the particle feed rates and turbulent conditions resulting in a nonuniform aerosol concentration downwind of the source. Both these factors were visually observed to cause backscatter variations, but an estimate of their relative importance could not be made. Nevertheless, these data illustrate that the lidar technique provides a downwind measurement that responds in a predictable (linear) manner to the total particulate emission rate as evaluated from feeder calibration data.

V ABSOLUTE PLUME DENSITY MEASUREMENTS

The data presented in the previous section of this paper indicate that for a given type of particulate material, the lidar can be calibrated to provide an absolute measurement of aerosol density of a downwind pollution plume. However, fugitive dust emissions from a multiplicity of source types may consist of a heterogeneous complex of particle size, shape, and composition distributions. The lidar wavelength(s) or observational techniques must be carefully selected to minimize errors of density measurement associated with uncertainties of particle characteristics.

Earlier experiments used a large-scale chamber that was especially designed for making remote lidar observations of generated aerosols of known particle size, shape, composition, and concentration (Lapple and Uthe, 1976). Observations of aerosols generated from fly-ash particulates of different size categories showed that the backscatter-to-mass concentration ratio is less dependent on particle size at a lidar wavelength of 1.06 μm than at 0.7 μm. Recent transmissometer experiments (Uthe, 1978) have shown that extinction measurements in a wavelength interval of 3 to 4 μm provide a good indicator of aerosol volume concentration regardless of particle size, shape, or composition (for the range of typical pollutants). While backscatter certainly is sensitive to changes in particle shape or composition, measurements in the 3- to 4-μm region may result in a higher correlation between backscatter and aerosol mass concentration when

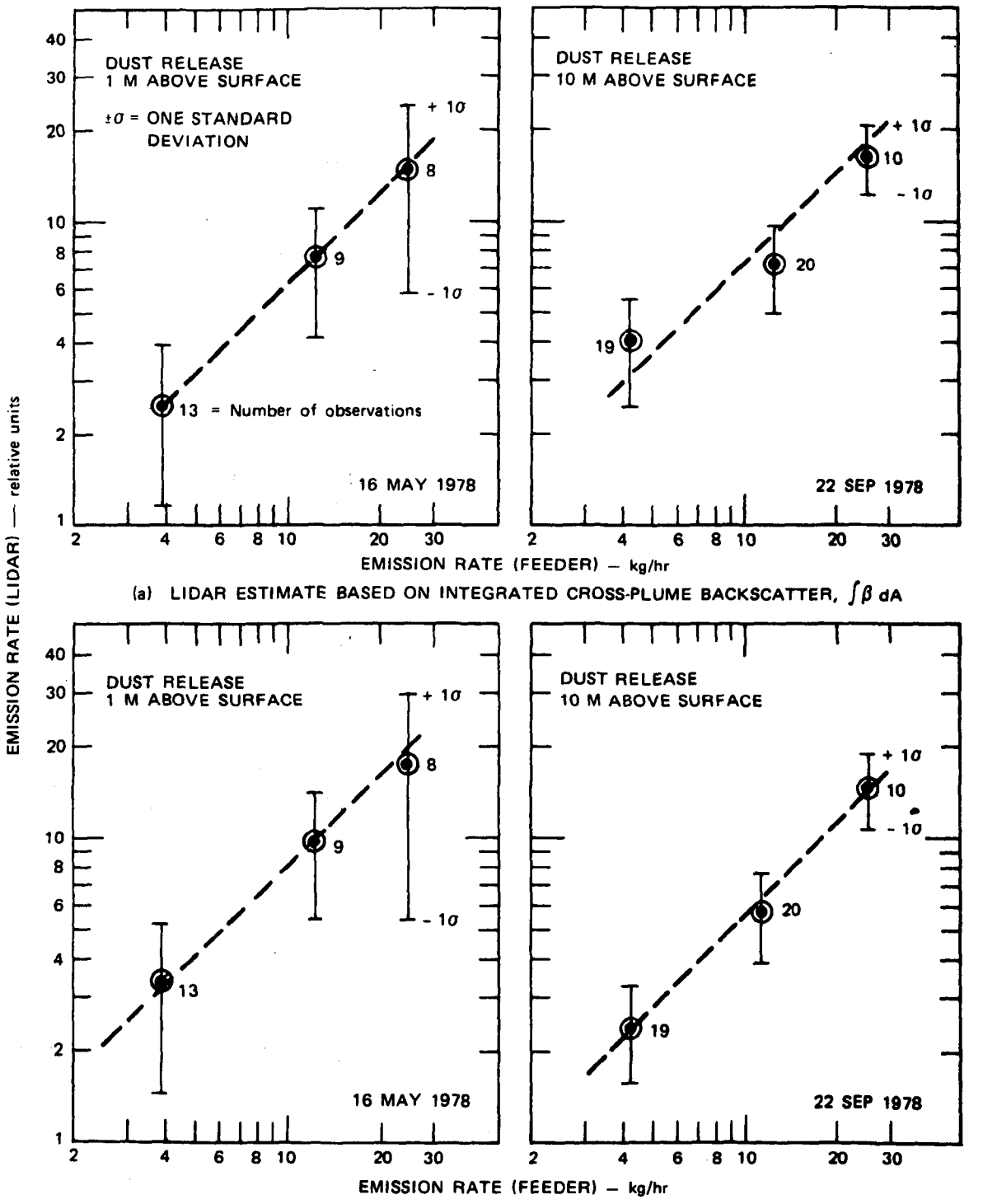


FIGURE 4 PARTICULATE EMISSION RATE EVALUATED FROM LIDAR PLOTTED AS A FUNCTION OF EMISSION RATE EVALUATED FROM PARTICLE FEEDER
Dashed line is best-fit linear relation.

the particle size is variable. Clearly, additional experiments are needed to define optimum wavelength regions and to establish uncertainties in plume density measurements because of unknown particle characteristics.

A downwind in-situ measurement of the backscatter-to-mass concentration ratio may provide the basis for converting backscatter observations to fugitive particulate concentrations. One possible approach is to sample the aerosol mass concentration at a location near the path of the laser beam. For example, a filter sample could be taken over an extended time period while repetitive lidar observations are made of nearly the same atmospheric volume. The time-integrated backscatter from this volume and the mass concentration measurement would provide an estimate of the backscatter-to-mass concentration ratio required to quantitatively evaluate the integrated cross-plume density from the lidar-observed, above-surface backscatter distributions. While this method may provide the necessary information to interpret backscatter data quantitatively in terms of aerosol density, an especially designed instrument for measuring the backscatter-to-mass concentration ratio is desired. Of course, if an observational technique can be developed to reduce to within acceptable standards the dependence of the density measurement on the uncertainties of particle characteristics, these supporting observations will not be necessary.

VI CONCLUSIONS AND RECOMMENDATIONS

Using an existing mobile lidar system, a field program has been conducted to demonstrate that for a single particulate type, the cross-plume integrated backscatter is a good indicator of source strength of particulate emissions. The positive results obtained from this first field program encourage the further development of this methodology for measuring fugitive dust concentrations.

We plan to test the lidar technique using sources of different particle size, shape, and composition distributions. For example, the next test may be of the emissions from a liquid-particulate generator capable of producing both black and white oil smokes. Other areas to be investigated include:

- Selection of optimum wavelength(s) regions to minimize mass measurement errors associated with uncertainties in particle size, shape, and composition distributions.
- Development of an eye-safe lidar technique.
- Evaluation of wind-measuring lidar techniques to evaluate actual flow of observed pollutants.
- Development of an instrument to measure backscatter-to-mass concentration in situ.
- Further experimental evaluations of proposed methodologies using sources of known particulate properties.

ACKNOWLEDGMENT

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DEVELOPMENT OF A FUGITIVE ASSESSMENT SAMPLING TRAIN FOR PARTICULATE AND ORGANIC EMISSIONS

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Abstract

The measurement of fugitive emissions from sources that preclude the capture of emissions before their diffusion into the ambient atmosphere poses some unique problems. Devices designed to obtain ambient samples for area studies generally do not provide samples of particulate matter large enough for meaningful quantification and qualification analyses in a reasonable sampling period, and are usually completely non-directional in their sampling.

This paper describes a program for the development of a prototype portable Fugitive Assessment Sampling Train (FAST) designed to obtain a 500 milligram particulate matter sample in an 8 hour sampling period downwind of most industrial sources. The development of the design criteria, establishment of operating parameters, selection and design of hardware components, fabrication and initial testing of the FAST are described in detail, and the program for the qualification testing of the completed unit is outlined.

A considerable portion of the air-polluting particulate matter and organic vapor emissions from industrial and energy-related processes is generated by sources that do not permit the capture of their emissions for measurement purposes before their diffusion into the ambient atmosphere. Obtaining samples of such fugitive emissions of sufficient size to perform statistically significant analyses of their concentration, particle size distribution, physical characteristics, chemical composition or biological activity presents a problem not readily solved using existing devices and traditional sampling techniques.

Standard high volume samplers, for example, can provide some information about the average particulate matter concentration at a sampling site over a long sampling period, but do not provide samples large enough for other than total mass determinations. Cascade impactors can provide particle size distribution information for a relatively small sample and cyclone separators can collect a fair-sized particulate matter sample in a few size ranges to provide essentially the same information. Grab sampling of gases or vapors for subsequent gas-chromatographic analysis can provide data on the chemical composition and approximate or relative concentration of these emissions, but is subject to the influence of interaction between emissions or aging of the samples. No single sampler exists that can collect a particulate matter sample large enough or a vapor sample stable enough to provide information in all areas.

This paper describes the progress made to date in the development of a Fugitive Assessment Sampling Train (FAST) designed to fill the requirements for a sampler capable of providing a large sample of particulate matter emissions from the atmosphere in a relatively short sampling period. A smaller organic vapor sample is obtained from the sampled stream.

The development effort was conducted by TRC-THE RESEARCH CORPORATION of New England under a contract (68-02-2133) with the Process Measurements Branch of the United States Environmental Protection Agency's Industrial Environmental Research Laboratory at Research Triangle Park, North Carolina. Discussions between TRC personnel and the EPA Project Officers, Dr. Robert M. Statnick and D. Bruce Harris, resulted in a target design specification for an ideal sampling train as the development starting point. This ideal sampler was described as being able to obtain, from the ambient air in the vicinity of an industrial fugitive emissions source, a 500 milligram sample of suspended particulate matter and a similar-sized sample of organic vapors in an eight-hour sampling period. The particulate matter sample would be separated into respirable (smaller than 3 micrometer) and non-respirable (larger than 3 micrometer) fractions. The sample sizes were selected to correspond to the then-considered minimum for complete analysis including bio-assay. The sampler was also to be self-contained and portable; it would require minimum power and, using commercially available components wherever possible, cost less than $10,000 to fabricate in the prototype version.

An extensive computerized literature search and review was conducted in the hope of obtaining sufficient information on ambient concentrations of industrial fugitive emissions as particulate matter and organic vapors to prepare a realistic system design specification for the FAST. While this search and review revealed almost no data on ambient concentrations, it did provide a wealth of information on emission rates from a wide variety of

industrial processes. A series of calculations based on the atmospheric diffusion equations in Turner's Workbook (1) for a range of atmospheric, topological and wind conditions was then performed to relate the published emission rates to ambient concentrations. These calculations indicated that an ambient concentration of 200 micrograms per cubic meter can be found within 100 to 500 meters of sources emitting between 0.6 and 23 kilograms per hour--a range covering about 90 percent of the industrial sources for which data is available.

This 200 microgram per cubic meter concentration was used to determine the sampling rate required to obtain a 500 milligram sample in an eight-hour period of 5.2 cubic meters per minute (184 CFM) as the initial system design parameter. A Roots lobe-type vacuum blower, capable of moving the required volume of air against a pressure drop of about 10 cm Hg, was selected as the particulate sampling prime mover. A system of drive belts and pulleys was utilized to operate the blower at the required 3800 RPM from a three horsepower drive motor. The drive system also provides enough flexibility to adjust the speed and the sampling rate up to about 20% if required.

To provide for the separation of the particulate matter sample into respirable and non-respirable fractions, an Air Correction Design 6UP Sanitary Cyclone Separator was selected. Its design capacity of 6.3 cubic meters per minute (222 CFM) provides a D_{50} at about 2 micrometers at a pressure drop of about 0.6 cm Hg. The cyclone was selected as preferable to filter- or impactor-type collectors since the sample is removed from the sampling stream and minimizes the degradation in sampling rate or effectiveness caused by the deposition of particulate matter on flow-through filters or impactor plates.

Consultations with Mr. Kenneth Cushing of the Southern Research Institute, under contract to the Process Measurements Branch in the area of particulate matter sampling, indicated that Reeves-Angel 934AH glass fiber filter material would be about 99.95% effective in collecting the fraction of the particulate matter sample down to about 0.3 micrometers passed through the cyclone. A circular format was selected for the filter material to provide the most even distribution of the sample on the filter surface and minimize the pressure drop buildup. A circular filter holder was designed to accommodate a 929 square centimeter (1 square foot) filter, limiting the pressure drop across the unloaded filter to 3.7 cm Hg. A louvered inlet section was also designed to reject particles larger than 100 micrometers to complete the particulate matter sampling section of the train.

To provide stable samples of airborne organic vapor emissions, it was decided to utilize an adsorbent resin in a removable canister that could be easily transported from the sampling site to a laboratory for extraction and analysis of the sample. Dr. Philip Levins of Arthur D. Little, Inc., under contract to the Process Measurements Branch in the organics sampling area, provided consultation to TRC on the resin. The best available resin, XAD-2 which is almost 100% effective in retaining organic vapors C_6 and higher, was determined to require a canister containing about 75 kilograms to provide a 500 milligram sample. This was prohibitive from the standpoints of size and cost, and the design criterion was revised to obtain the minimum sample required for a Level 1 assessment of 14 milligrams. This sample size requires only 2.1 kilograms of resin and a sampling rate of only 0.14 cubic meters per minute (5 CFM). A canister was designed and an oil-less Gast vacuum pump

selected to draw the sampling stream from the main stream after the particulate matter is removed.

The system design was reviewed and approved and procurement and fabrication efforts started. At this time, the EPA's Health Effects Research Laboratory suggested that an additional size fraction of the particulate matter sample be included to help in the assessment of the inhalable (less than 15 micrometer) portion of the emissions. It was decided to add a battery of six single stage Sierra Instrument impactors to the system to effect this additional fractionation between the inlet and the cyclone. These impactors were designed to provide a D_{95} for 15 micrometer particles at the system sampling rate with a pressure drop of only 0.05 cm Hg, and could therefore be added without affecting the system design.

The final system design is shown schematically in Figure 1. Design flow rates and pressure drops for each system element are shown enclosed in brackets. Samples retained by each element are shown in parentheses.

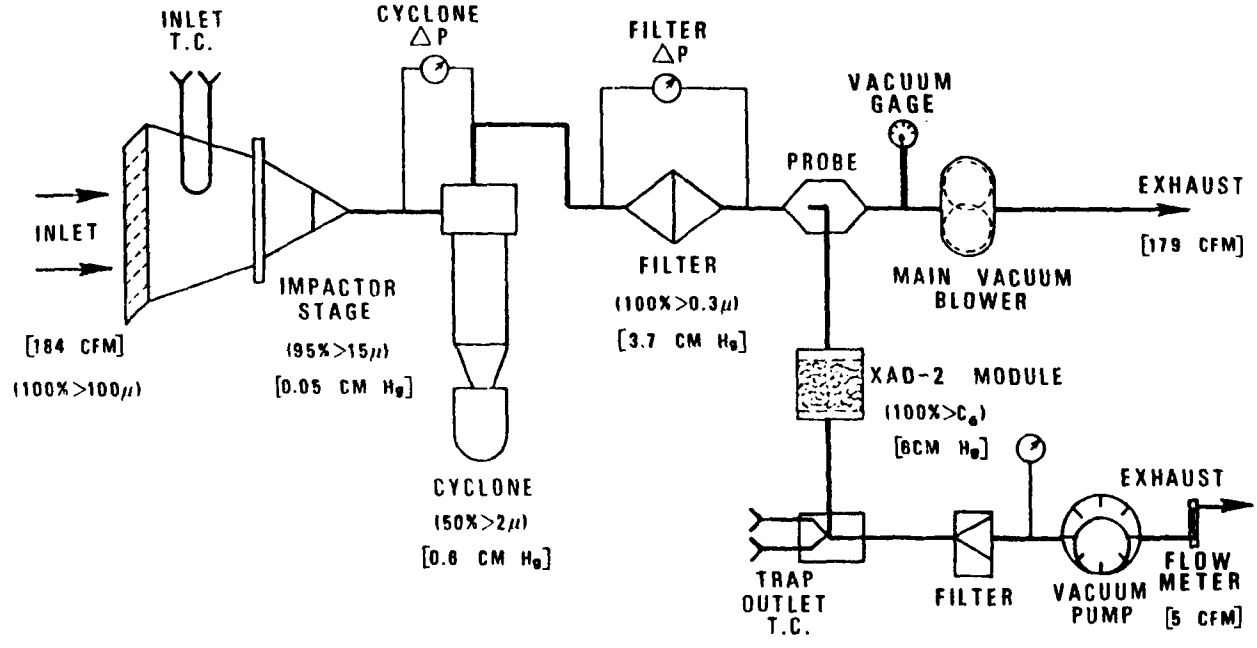

FIGURE 1 - FUGITIVE ASSESSMENT SAMPLING TRAIN
DESIGN OPERATING CONDITIONS

The procured and fabricated elements of the prototype system were then loosely packaged onto a space frame about 75 cm (2.5 feet) square by 183 cm (6 feet) high to allow easy access to the elements during development testing. The main sampling blower and the organic vapor sampling pump were separately mounted to improve the system's portability and permit the location of the

blower and pump exhausts away from the sampling inlet. The packages are shown in Figure 2 as they appeared for the initial operational tests.

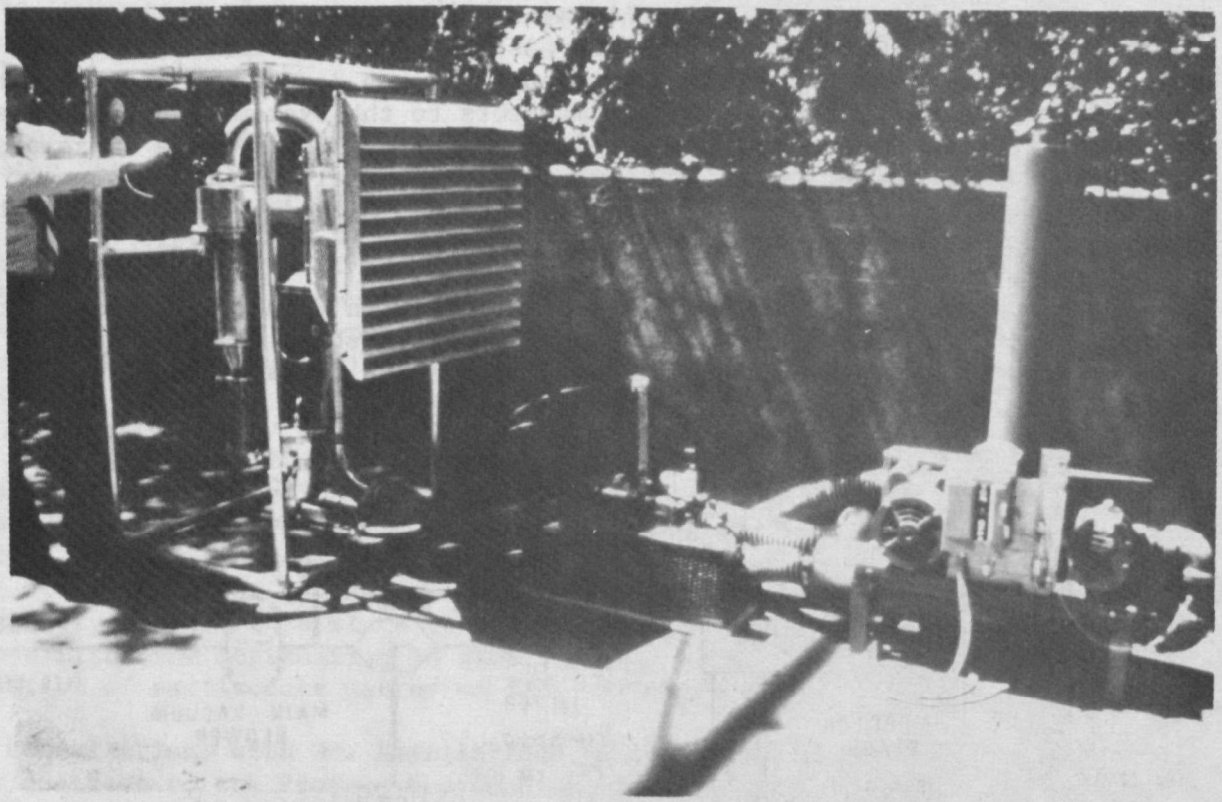

FIGURE 2

PROTOTYPE FAST SYSTEM INITIAL ASSEMBLY

After a successful operational test and a few modifications to the system were completed at TRC, the FAST was shipped to the Southern Research Institute's laboratory for calibration testing of the particulate sampling section. Tests were run by Southern using monodisperse ammonium fluorescein aerosols provided by their vibrating orifice aerosol generator at 3, 10 and 15 micrometers. The test results for the cyclone, shown in Figure 3 as points plotted on the manufacturer's design curve, are in very good agreement. The test results for the impactor, also shown in Figure 3, indicate good agreement with the design curve for smaller particles but are considerably lower than expected for the 15 micrometer particles of major concern.

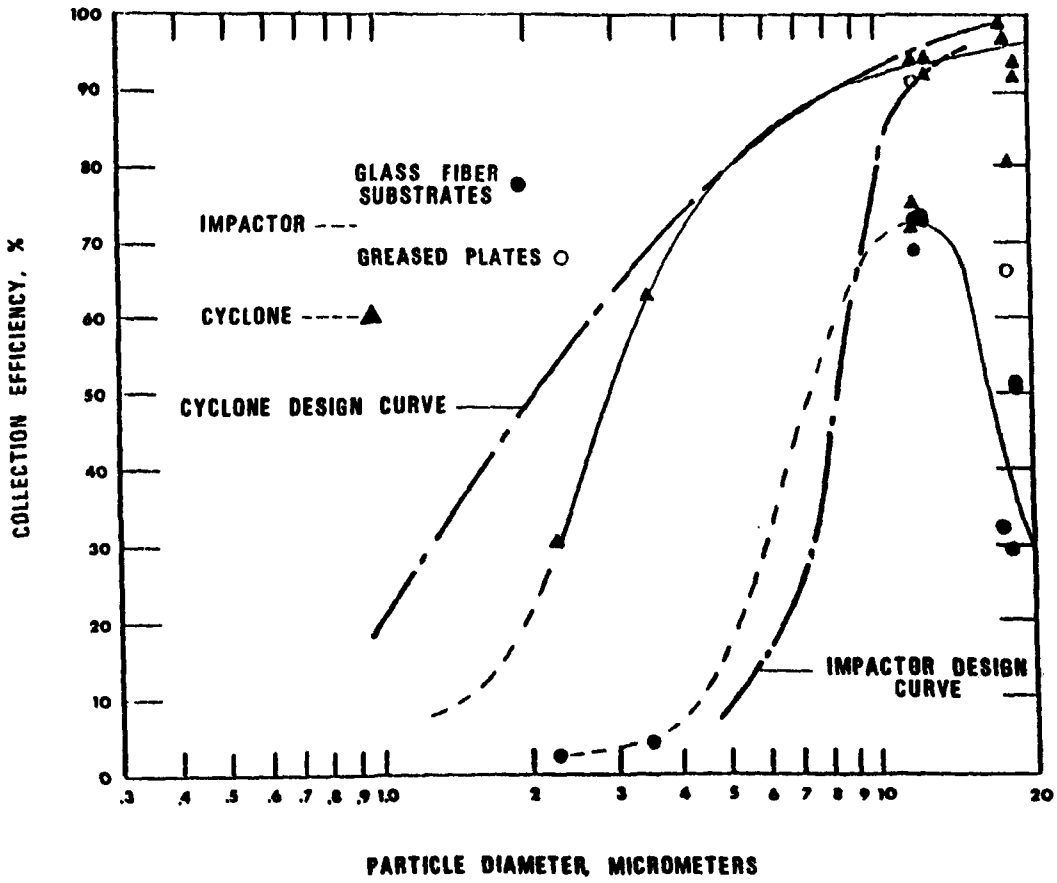

FIGURE 3: COLLECTION EFFICIENCY VERSUS PARTICLE DIAMETER FOR THE FUGITIVE ASSESSMENT SAMPLING TRAIN PRE-SEPERATOR IMPACTOR AND CYCLONE

Since it was felt that this discrepancy was caused by bouncing of the larger particles off the glass fiber substrate used in the impactors, a test was run using a grease substrate in an attempt to reduce the bounce. This resulted in a slight improvement in performance, but not to a level considered satisfactory for further development. A joint effort by TRC and Southern Research has been initiated to design and fabricate an elutriator to replace the impactor as the 15 micrometer fractionator. It is expected that the elutriator, envisioned as a battery of parallel settling chambers, will replace and perform the functions of both the inlet louvers and the impactors.

A field test of the FAST has been planned for the near future at a coke oven battery, where the system will be tested simultaneously with two Battelle mega-vol samplers and a standard hi-vol sampler in the measurement of emissions from coke pushing operations. This test, to be run prior to the fabrication and installation of the elutriator, should provide sufficient data for continued development of the cyclone and filter sections as well as an initial indication of the effectiveness of the adsorbent canister train design.

It is expected that the FAST will require some modifications and a repeat of the calibration and field testing cycle after the addition of the elutriator. Verification tests of the total system will be performed after the modifications and additions have been completed and an operating procedures manual will be prepared and published for the final version.

The efforts to date in the development of this Fugitive Assessment Sampling Train have been quite successful and encouraging. The completion of the planned effort will provide a useful tool for rapid, reasonable assessments of fugitive particulate matter and organic vapors from a wide variety of industrial sources.

Reference

(1) Turner, D. Bruce. Workbook of Atmospheric Dispersion Estimates. Public Health Service Publication No. 999-AP-26, U.S. Department of Health, Education and Welfare, Cincinnati, Ohio. Revised 1969. 84 pp.

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16. ABSTRACT

The proceedings are a compilation of technical papers prepared for presentation at the Third Symposium on Fugitive Emissions, October 23-25, 1978, at San Francisco, CA. The papers discuss the scope and impact of fugitive emissions (non-point sources) and present techniques which have been used to measure the emissions. Fugitive emission control technologies are also discussed.

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