

October 1974



**PREFERRED
STANDARDS PATH REPORT
FOR POLYCYCLIC
ORGANIC MATTER**

**U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Air Quality Planning and Standards
Strategies and Air Standards Division
Durham, North Carolina**

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STRATEGIES AND AIR STANDARDS DIVISION
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EXECUTIVE SUMMARY

Polycyclic Organic Matter (POM) is an aromatic hydrocarbon group that includes certain compounds which are proven carcinogens at elevated levels in laboratory animals. Some of these compounds have been linked with the occurrence of cancer in humans, but none have been directly related to cancer from exposures to ambient air. Nevertheless, it seemed prudent to consider POM a potential air pollution problem and to evaluate the need for additional formal regulatory action. The objective of this report is to assess the POM problem, determine the requirement for additional regulatory action under the Clean Air Act, and identify the most appropriate regulatory tool if such action is necessary.

POM includes thousands of compounds which vary widely in physical and chemical characteristics and in their capacity to act as carcinogens. Data on total POM are extremely limited. Most source emission estimates are for a single POM compound, benzo(a)pyrene (BaP), and not for total POM. BaP is a proven carcinogen commonly used as a surrogate for total POM. No analytical techniques exist for ambient air measurements of total POM. The majority of these measurements are for BaP only. Limited dose-response data that have been assembled also are for BaP and not total POM.

Sources of BaP normally are associated with the incomplete combustion of organic compounds, especially coal. The nationwide emissions of BaP are estimated at about 900 tons per year based on 1971-1973 data.

Major source categories and their estimated contribution to this total are listed below.

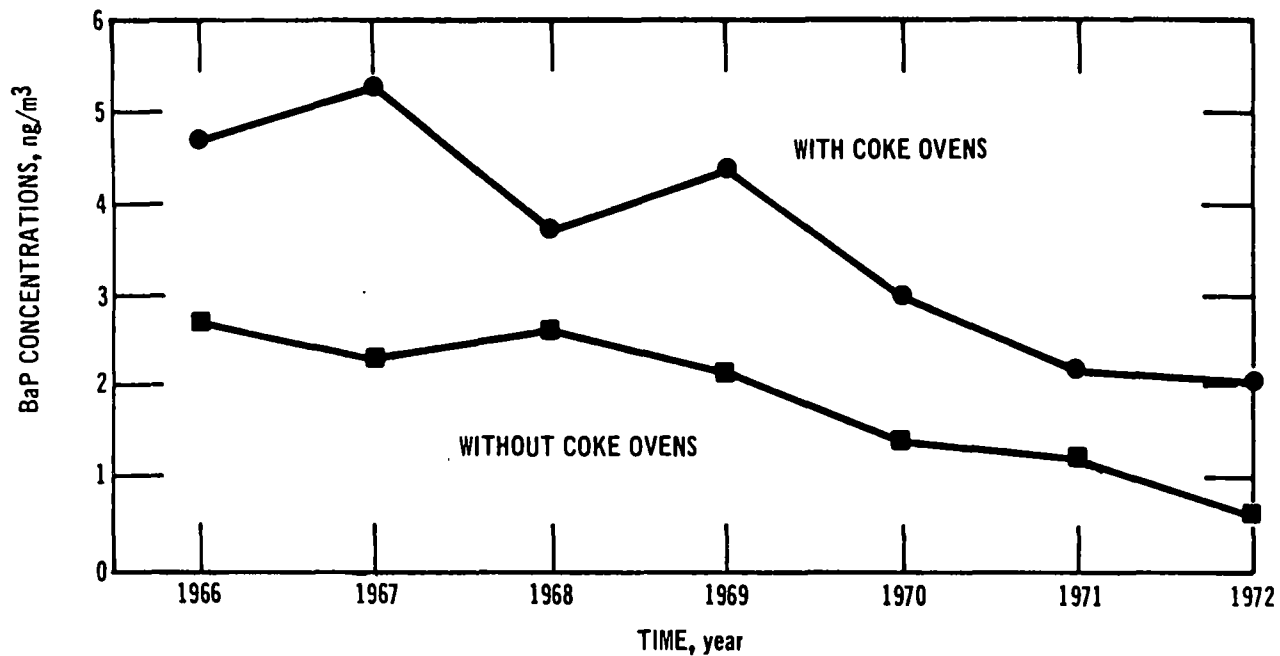
<u>Source Type</u>		<u>BaP Emissions</u> <u>(tons/year)</u>	<u>% of</u> <u>Total</u>
		(1971-1973)	
1.	Coal Refuse fires	310	34.7
2.	Residential furnaces, coal (hand-stoked)	300	33.6
3.	Coke production	170*	19.0
4.	Vehicle disposal (open burning)	25	2.8
5.	Wood burning (fireplaces, etc.)	25	2.8
6.	Mobile sources, gasoline	11	1.2
7.	Forest and Agricultural burning	11	1.2
8.	Tire degradation	11	1.2
9.	Open burning (domestic--municipal)	10	1.1
10.	Intermediate coal furnaces	7	0.8
11.	Petroleum refineries	7	0.8
12.	Enclosed incineration (apartment--municipal)	3	0.3
13.	Other	<u>4</u>	<u>0.5</u>
TOTALS		894	100.0

*A range of emissions has been given for this value. Appendix F contains the details.

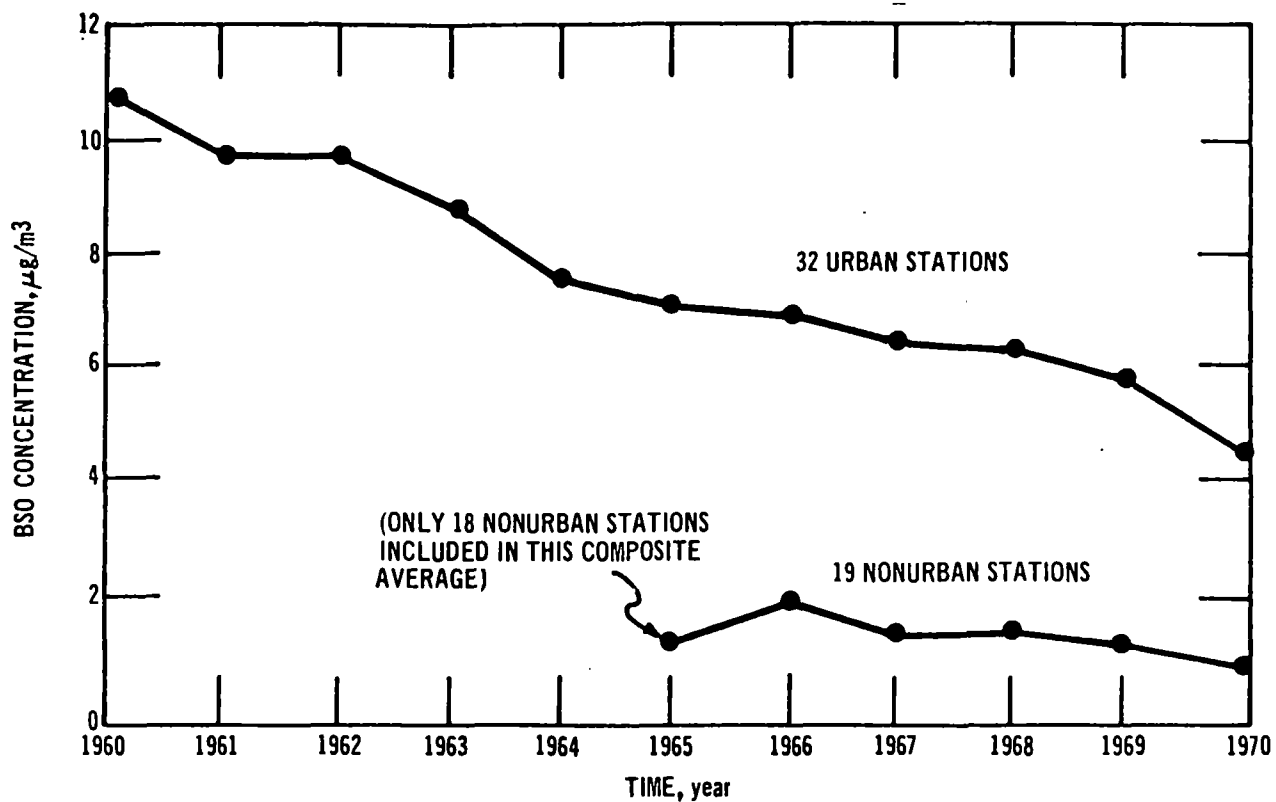
As illustrated by this list, stationary sources account for about 97% of the nationwide emissions of BaP. The inefficient combustion of coal is the largest single source of BaP. Thus the three largest emission sources are coal refuse fires, residential coal furnaces, and coke ovens. Together, they comprise more than 85% of national emissions of BaP.

Annual concentrations of BaP range from 0.07 to about 17 nanograms per cubic meter of air sampled (ng/m^3). Urban levels are higher than non-urban levels by as much as 10 times under some conditions. Annual ambient air concentrations of BaP have declined significantly over the past few years. In order to assess these declines, BaP trends data were compiled for selected cities from 1966 through 1972. Cities containing large coke oven facilities were singled out as a separate category since coke ovens are a major source of POM and were considered for special regulatory action. In addition, trends in the benzene soluble organic fraction (BSO) of total suspended particulates were analyzed for urban and non-urban sites since data for this fraction are more complete than data for BaP. Both BaP and POM are found in the benzene soluble portion of total particulates.

Trends data for BaP (shown in the following figure) indicate a significant decline in ambient concentrations. From 1966 to 1972, concentrations have decreased about 55% in coke oven cities and 77% in non-coke oven cities. For all cities sampled, the urban composite average BSO concentrations (shown in the second figure) have decreased



Trends in annual BaP concentrations in cities with and without coke ovens.



Trends in BSO and in BSO percentage of TSP at 32 urban and 19 nonurban stations.

from 10.6 ug/m³ in 1960 to 4.8 ug/m³ in 1970, or a 55% decrease.

These decreasing trends are significant and can be attributed to the success of existing control programs as well as the move away from the use of coal in small, inefficient furnaces.

Coke ovens are a special problem. Based on available data, they are suspected of being a major source of BaP and are difficult to control. Demonstration projects sponsored by the Environmental Protection Agency and industry groups indicate that particulate emissions from coke ovens can be controlled by a combination of process alteration and equipment design modifications. Preliminary results show that application of good particulate emissions control will also reduce BaP emissions by 85-90%. Federal emission standards for particulates being developed for new coke ovens, and enforcement actions underway against existing ovens, ensure adequate regulation of this remaining significant source of BaP.

Existing Regulatory Control Program

Coal refuse fires, residential coal-fired furnaces, and coke ovens (the three major source categories of POM emissions) are being controlled, or are scheduled for control, through the following actions:

1. Coal refuse fires are regulated in the four States which include the majority of coal refuse fires. The nine other States with coal refuse fires can control emissions from this source by open burning regulations or other regulations. In addition, the Bureau of Mines, Department of Interior, has proposed Federal regulatory action for this source. They presently require control of spontaneously started fires under existing Federal regulations.

2. Emissions from coal-fired residential sources decreased 28% between 1968 and 1972 and this trend is expected to continue for the following reasons:

- a) Socio-economic conditions have established a trend away from coal-burning in the home because of its dirtiness and inconvenience.
- b) All states except one are imposing particulate emission limits on coal-fired residential furnaces. In many cases, these limits, by requiring particulate control systems, discourage the continued use of coal for residential heating.
- c) Residential coal-fired furnaces are banned directly in some urban areas such as Chicago, St. Louis, and Milwaukee. Others have indirect bans through stringent sulfur regulations which preclude the use of most, if not all, sources of coal.

3. New coke ovens will be controlled by national particulate emission standards being developed for this source. Control equipment has already been installed on several existing coke ovens as a result of local regulations and Federal enforcement actions. EPA has fourteen enforcement actions against coke ovens for particulate controls. Preliminary data from EPA demonstration projects indicate that about 90% control can be achieved for particulate emissions during charging operations, which historically has been responsible for most emissions from this source. These same data show an equivalent degree of BaP emission control can be expected.

Most of the remaining sources for BaP are small, numerous and widely dispersed. Several of these sources are not possible to control effectively (e.g., home fireplaces, forest and agricultural burning). Others (petroleum refineries, vehicle disposal, and open burning) are being regulated through New Source Performance Standards and State Implementation Plans. Although some concern has been expressed regarding increased use of coal for electrical power generation under our current national energy policy, such increased use is not anticipated to cause increased emissions of POM because power plants utilize very efficient combustion systems and emit essentially no POM.

Conclusions and Recommendations

The health effects of ambient air concentrations of POM or BaP are not well documented and information is not available for selecting harmful air concentrations. Although the presence or magnitude of the threat from POM cannot be determined at this time, selected polynuclear organic materials have been shown to be carcinogenic in some situations. Therefore, it seems prudent to minimize emissions of POM and to use all practical means to reduce levels of POM in the air.

Ambient concentrations of BaP and benzene soluble organics have declined significantly over the period for which trends data are available. From 1966 to 1972, BaP concentrations have decreased 55% in coke oven cities and 77% in non-coke oven cities. This decline in ambient levels is attributed to current particulate control programs and the phase-out of coal-fired home furnaces. Although these actions have not been oriented specifically to reducing BaP emissions, they do have a direct effect on those sources responsible for most BaP emissions

and are an effective national control strategy. An analysis of additional regulatory steps does not suggest other effective measures.

Control regulations specifically for BaP or POM are not warranted nor practical at this time. Ambient monitoring for BaP should be undertaken in selected cities to ensure the continued adequacy of the control program. Also, special emphasis should be given to the prompt enforcement of existing regulations for incinerators, open burning, coal combustion, burning refuse piles, and coking operations.

PREFERRED STANDARDS PATH REPORT FOR POLYCYCLIC ORGANIC MATTER

1. INTRODUCTION

Polycyclic organic matter (POM) and benzo(a)pyrene (BaP) were two of several pollutant groups for which control strategies were to be considered as listed under National Objectives in the Administrator's "Guidelines for Development of Fiscal Years 1973-1978 Program Plans." Since BaP is a specific POM compound and is generally used as a surrogate for POM the two tasks were combined.

The procedure used in preparing this report and arriving at recommendations included:

- (1) studying control options available under the 1970 Clean Air Act, an internal EPA procedure known as a preferred standards path analysis.
- (2) reviewing and evaluating comprehensive studies available on POM, including:

- (a) "Particulate Polycyclic Organic matter"^{3*} by the National Academy of Sciences, 1972.
 - (b) "Draft NERC/RTP Position Paper on Particulate Polycyclic Organic Material (PPOM)"¹⁶ by PPOM Task Force Panel, U.S. EPA, NERC/RTP, 1972.
 - (c) "Preliminary Air Pollution Survey of Organic Carcinogens"¹¹ by Litton Systems, Inc., for HEW, 1969.
 - (d) "Sources of Polynuclear Hydrocarbons in the Atmosphere"⁸ by R.P. Hagebrauck, et al., AP-33, 1967.
- (3) evaluating available ambient air data.

After thorough evaluation of this information, the following abridgement is presented.

1.1 Background

Polycyclic organic matter (POM) is organic matter with a multiple ring (two or more) structure, as shown in Figure 1. A typical structure of five rings is shown in Figure 1 with the dashed circle representing the carbon-carbon (C-C) bond and the numbers around the outside representing points where, in this case, hydrogen atoms are attached. Other atoms (or groups of atoms) may be substituted for carbon at the numbered points. When such a substitution occurs, polycyclic azaheterocyclic compounds (aza-arenes) are produced. Other names which are given to POM include (1)

* Numbers shown as superscripts refer to references at the end of the Report.

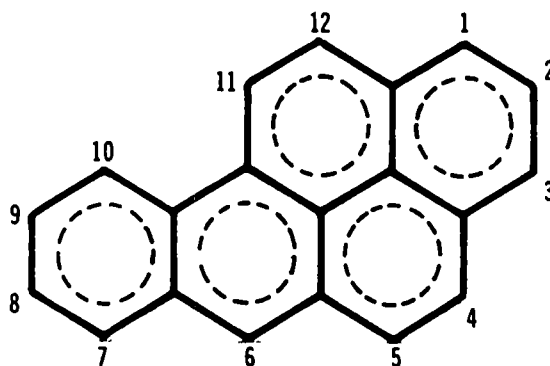


Figure 1. Typical structural formula of POM, benzo(a)pyrene .

polynuclear aromatic hydrocarbons (PNAH, PAH); (2) polynuclear aromatics (PNA); (3) polycyclic aromatic hydrocarbons (PAH); or simply, (4) aromatic ring compounds. Generally, when any of these names, or variations thereof, are used the broad class of POM is being referenced.

The primary reason for interest in POM is that some of these compounds have been proved to be carcinogens in experimental animals. They may also act as cocarcinogens, or be active in the presence of other cocarcinogens. (A carcinogen is a substance or agent which produces or incites cancer. Agents which induce altered physiological states that may increase the risk to carcinogenesis are cocarcinogens). Furthermore, epidemiological evidence connects lung cancer etiology with occupational exposure to certain particulate POM (PPOM) compounds in specific industries. A few POM compounds which have been identified as definite carcinogens are :

7,12-Dimethylbenz(a)anthracene
Dibenz(a,h)anthracene
3-Methylcholanthrene
Benzo(c)phenanthrene
Benzo(a)pyrene
Dibenzo(a,h)pyrene
Dibenzo(c,g)carbazole

The last compound in the list is an example of a carcinogenic aza-arene and is illustrated in Figure 2.

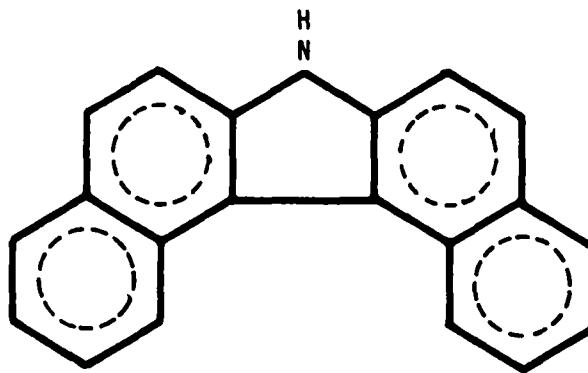


Figure 2. Typical structural formula of an aza-arene, dibenzo(c,g)carbazole.

1.2 Scope of Problem

Under the Clean Air Amendments of 1970 the EPA was provided with alternative approaches to control strategies for candidate pollutants. The process of looking at these alternatives is known as a preferred standards path analysis (PSP), which is explained in greater detail in Appendix A. The PSP analysis in this report uses BaP as an indicator for PPOM. Reasons for limiting this analysis to BaP as an index for PPOM are:

- a. Existing data support using BaP as a rough indicator for PPOM (Figure 3 and Section 5). If effective control is achieved for BaP other POM compounds should also be controlled to a large degree because their characteristics are similar to BaP.
- b. Lack of available data for POM:
 - 1. Emissions estimates are for BaP and not for POM; some scientists have limited data on total POM.
 - 2. Majority of ambient air measurements are for BaP only; selected research studies have produced limited ambient data for POM.
 - 3. Limited dose-response information does exist for BaP but none is available for total POM.
- c. BaP has received primary emphasis in measurement techniques development for specific POM compounds; none are available for total POM.

Thus, this report presents the preferred standards path for BaP as a surrogate preferred standards path for POM. The NAS report, the ORM/NERC/RTP position paper on PPOM, extensive personal contacts, and other literature have been used for support of the scenario contained in this report.

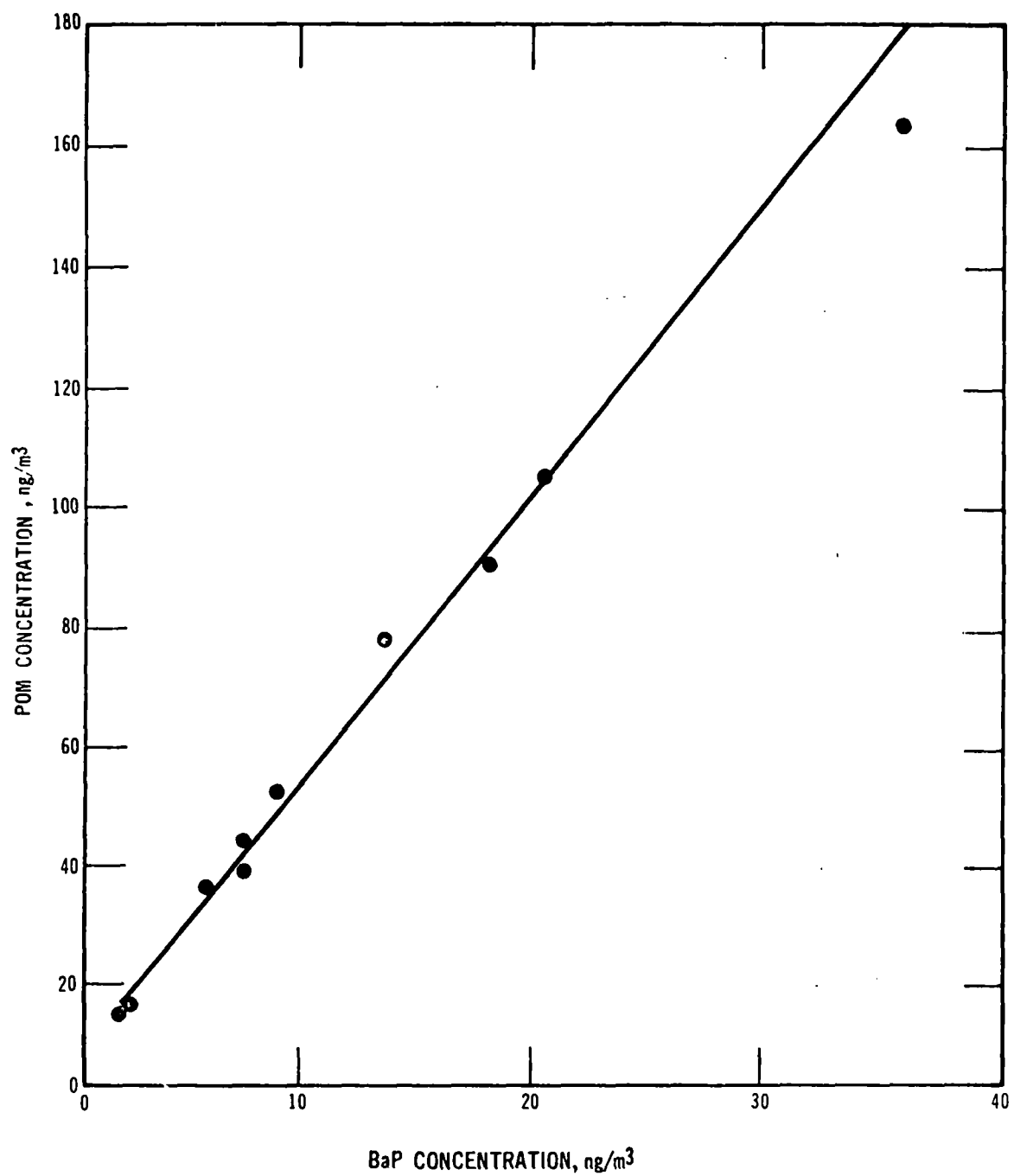
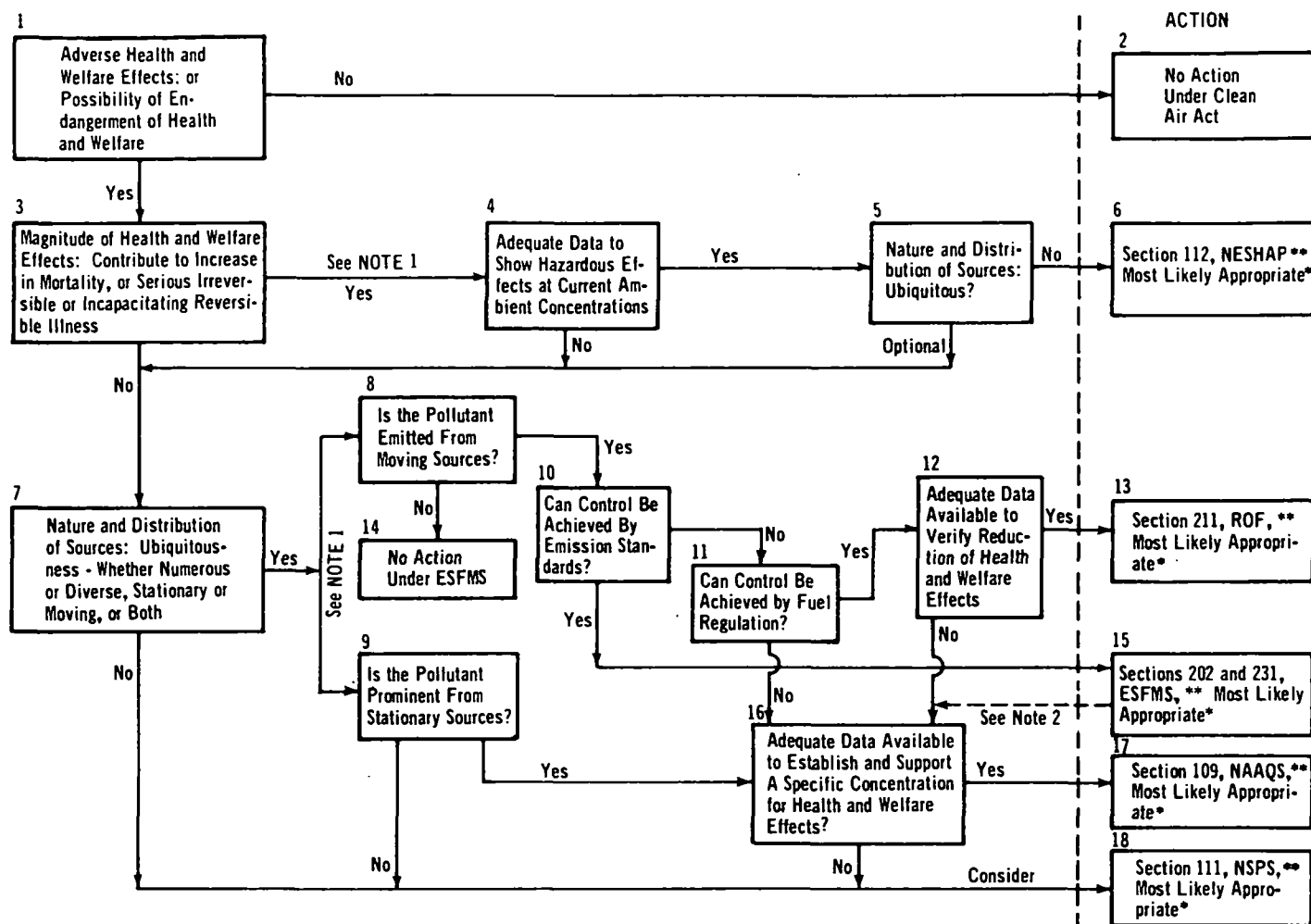


Figure 3. Annual POM concentrations (10 compounds) versus annual BaP concentrations in Birmingham, Ala., 1964 - 1965.

2. RATIONALE AND RECOMMENDATIONS FOR EPA STRATEGY FOR BaP

2.1 Criteria under the Clean Air Amendments of 1970

Criteria available to the Environmental Protection Agency (EPA) for recommending a preferred standards path for any pollutant from either a stationary or moving source, as described in the Clean Air Amendments of 1970 (Act), include the following factors: (1) Presence and magnitude of health and/or welfare effects of a pollutant; (2) Nature and distribution of pollutant sources; and (3) Supporting data (implied). Those sections of the Act in which these factors are included, along with the six basic standard setting options, are described and then discussed below. The characteristics of a candidate pollutant are compared with criteria for each option given (Figure 4). This chart should not be considered a strict decision-making tool but merely a guide that provides one of many inputs into the selection process. Analysis must begin with an estimation of health effects since this is the common thread among all six options. (See block 1 of Fig. 4.) Once the determination is made that a candidate pollutant may contribute to adverse health and/or welfare, one or more of the options for control should be chosen. The approach to a control strategy requires different procedures depending upon the chosen option.



NOTE 1: A yes answer here simply means the pollutant is a candidate for regulatory action; however, it does not denote mandatory action.

NOTE 2: The dashed line indicates that significant pollution from stationary sources may remain even after utilizing ESFMS. Land use and transportation controls may need to be implemented in addition to Section 202.

*MOST LIKELY APPROPRIATE simply means that the indicated option may be the most logical without considering external factors. However, other ramifications may preclude its use. Also, more than one option (e.g., Sections 109 and 202; Sections 109 and 111; etc.) may be used in combination for any given pollutant in order to achieve the full intent of the Act. (Also see NOTES.) Section 111(d) is applicable to non-criteria pollutants.

** Acronyms explained in text.

Figure 4. Preferred standards path: Guide for determination of regulatory action.

2.1.1 National Emission Standards for Hazardous Air Pollutants
(NESHAP - Section 112)

Under the Act the Administrator is allowed to use his judgment to determine whether a pollutant is hazardous, i.e. "may cause, or contribute to, an increase in mortality or an increase in serious irreversible, or incapacitating reversible, illness" [Section 112(a)(1)]. Within 180 days after publishing a list of suspected hazardous pollutants the Administrator must publish proposed regulations setting emission standards together with a notice of public hearings. "Not later than 180 days after such publication, the Administrator shall prescribe an emission standard for such pollutant, unless he finds, on the basis of information presented at such hearings, that such pollutant clearly is not a hazardous air pollutant, [then] the Administrator shall establish any such standard at the level which in his judgment provides an ample margin of safety to protect the public health from such hazardous air pollutant" [Section 112(b)(1)(B)].

A major consideration in making a preferred standards path choice is to assess the largest magnitude of adverse health effects, i.e., possible mortality or serious illness. This requires evaluating the candidate pollutant for the hazardous option (Section 112) initially. Here, the effect of atmospheric emissions on health must be analyzed in relation to (a) current and expected ambient levels (concentrations) and (b) what an ample margin of safety is. In practice, this relationship is affected by several factors, including terrain, number of sources, extent of buildup or persistence in the environment, etc. Setting a national emission standard despite these variables may logically require

a level of emissions to be either zero or based upon the worst significant situation observed or anticipated. If in applying these criteria, specific measured or expected ambient concentrations of the candidate pollutant are shown to be hazardous, the hazardous pollutant option may be the choice. If so, a national emission standard which provides an ample margin of safety must be proposed and promulgated. The standard will apply to stationary sources, new and existing. The Act does not specifically require that the Administrator have information on control technology to initiate action under this option. However, from a practical point of view, since the Administrator must oversee implementation and enforcement of emission standards knowledge of control technology is required.

2.1.2 National Ambient Air Quality Standards (NAAQS -
Sections 108-110)

The Act requires the promulgation of primary "ambient air quality standards the attainment and maintenance of which in the judgment of the administrator, . . . allowing an adequate margin of safety, are requisite to protect the public health" [Section 109(b)(1)]. Similarly, secondary NAAQS are required to "protect the public welfare from any known or anticipated adverse effects" [Section 109(b)(2)]. Ambient air quality standards are based upon criteria which delineate "all identifiable effects on public health or welfare" from a pollutant whose "presence . . . in the ambient air results from numerous or diverse mobile or stationary sources" [Section 108(a)]. The Act further requires each State to "adopt and submit to the Administrator . . . a plan which provides for implementation, maintenance, and enforcement of such . . .

standard in each air quality control region . . . within such State" [Section 110(a)(1)].

In controlling pollutants under the ambient option the effect of existing ambient concentrations on health and welfare must first be analyzed. Such data must be published in a criteria document simultaneously with a proposed national standard for a specific ambient concentration which can be supported. Then the States are left to establish the relationship between ambient concentrations and emission levels from sources. This relationship is affected by such factors as terrain, number of sources, and effect of buildup or persistence of the candidate pollutant in the environment. States are responsible for prescribing and enforcing emission standards, procedures for control of number or location of sources, etc. However, the Administrator must issue control techniques information simultaneously with criteria documents [Section 108(b)(1)]. Also from the practical point of view, development of implementation plans by States and the Administrator's review and approval of such plans require some knowledge of control technology.

2.1.3 Standards of Performance for New Stationary Sources (NSPS - Section 111)

The Act specifies that the Administrator include a category of sources on a proposed list for standards of performance "if he determines it may contribute significantly to air pollution which causes or contributes to the endangerment of public health and welfare" [Section 111(b)(1)(A)]. Further, within 120 days "the Administrator shall propose regulations, establishing Federal standards of performance for new sources within such category" and ". . . promulgate within 90 days" [Section 111(b)(1)(B)].

The Act further requires the Administrator to "prescribe regulations . . . under which each State shall submit . . . a plan which (A) establishes emission standards for any existing source for any air pollutant (i) for which air quality criteria have not been issued or which is not included on a list published under Section 108(a) or 112(b)(1)(A) but (ii) to which a standard of performance under subsection (b) would apply if such existing source were a new source, and (B) provides for the implementation and enforcement of such emission standards" [Section 111(d)(1)].

When a limited number of source categories or limited number of predominant categories exist, the section on standards of performance for new sources may be the most practical approach to controlling a candidate pollutant. If Section 111 is used, the effect of atmospheric emissions of the candidate pollutant on health and welfare must be analyzed first. However, the Act does not require calculating a relationship between ambient concentrations and emissions since the standard will reflect the best demonstrated system of emission reduction for the affected new source (taking cost into account). Thus, the level of control is not related directly to health and welfare effects of the candidate pollutant. Analysis for health and welfare effects do or could exist. For unmodified existing sources, States must enact emission standards using available practical approaches, subject to EPA review and approval [Section 111(d)].

2.1.4 Emission Standards for Moving Sources (ESFMS - Title II)

Title II of the Act, Emission Standards for Moving Sources (ESFMS), provides for control of air pollutants from moving sources. This title presents criteria for motor vehicles, fuel regulations, and aircraft.

a. Motor Vehicle Emission Standards (MVES - Section 202)

Under this section of the Act "the Administrator shall by regulation prescribe . . . standards applicable to the emission of any air pollutant from any class or classes of new motor vehicles or new motor vehicle engines, which in his judgment causes or contributes to, or is likely to cause or contribute to, air pollution which endangers the public health or welfare. Such standards shall be applicable to such vehicles and engines for their useful life" [Section 202(a)(1)]. The Act further requires the Administrator to allow time for "development and application of the requisite technology, giving appropriate consideration to the cost of compliance within such period" [Section 202(a)(2)]. Subsection (b) of Section 202 presents exceptions which apply to standards and time requirements for hydrocarbons (HC), carbon monoxide (CO), and nitrogen oxides (NO_x).

b. Regulation of Fuels (ROF - Section 211)

"The Administrator may, . . . by regulation, control or prohibit the manufacture, . . . or sale of any fuel or fuel additive for use in a motor vehicle or motor vehicle engine (A) if any emission products of such fuel or fuel additive will endanger the public health or welfare" [Section 211(c)(1)(A)]. The Act further provides that control under the subsection just quoted is prohibited "except after consideration of all relevant medical and scientific evidence available to him, including consideration of other technologically or

economically feasible means of achieving emission standards under section 202" [Section 211(c)(2)(A)]. Furthermore, in connection with registration of fuels and additives the Act provides means whereby "the Administrator may also require the manufacturer of any fuel or fuel additive - (A) to conduct tests to determine potential health effects of such fuel or additive (including, but not limited to, carcinogenic, ... effects)," [Section 211(b)(2)(A)].

c. Aircraft Emission Standards (AES - Section 231)

The Administrator shall issue, based on studies conducted by him, "proposed emission standards applicable to emissions of any air pollutant from any class or classes of aircraft or aircraft engines which in his judgment cause or contribute to or are likely to cause or contribute to air pollution which endangers public health or welfare" [Section 231(a)(2)]. Provision is made for final regulations to be issued within 90 days after the proposal following public hearings within the most affected air quality control regions (AQCR) [Section 231(a)(3)]. However, the regulation shall not take effect until necessary time has elapsed to develop and apply requisite technology and consultation with the Secretary of Transportation [Section 231(b) and (c)].

In considering the nature and distribution of sources of the candidate pollutant, one of the first questions is whether emissions are from moving sources. If so, then additional questions must be asked

(block 8 of Figure 4) because at least two options become available. However, regulation of fuels (Section 211) can be employed only after considering all possibilities of control by emission standards under Section 202, as well as relevant medical and scientific evidence. If emission standards are not, or can not be, effective without fuel regulation, then this option may be used also.

Emissions from aircraft must also be considered in connection with moving sources. If such emissions have a significant impact on health and welfare effects the Administrator, in consultation with the Secretary of Transportation, is authorized to set standards. Enforcement of these standards is the responsibility of the Secretary of Transportation, after consultation with the Administrator [Section 232(a)]. Standards can be set and enforced only after allowing what is, in the Administrator's [Section 232(a)]. Standards can be set and enforced only after allowing what is, in the Administrator's judgment, sufficient time for development and application of requisite technology. Cost of compliance must be considered also [Section 231(b)].

2.1.5 Combination of Options

Pollutants may be regulated under more than one section of the Act simultaneously (Figure 4). However, this approach cannot be used in at least two specific situations. These situations occur when pollutants are potential candidates for control under Section 112 (which cannot be applied to a pollutant for which an ambient air quality standard exists) or Section 111, subsection (d) (which cannot be applied to any pollutant for which ambient or hazardous standards exist). However, Section 111 may supplement Sections 109 and 112 to control the same pollutant if

specified criteria (as discussed in Subsections 2.1.1-2.1.4 above and Appendix A) are fulfilled. Once a pollutant is regulated under Section 111, Section 111(d) becomes effective for existing sources of the pollutant, unless ambient or hazardous pollutant standards apply.

Standards for moving sources, Sections 202, 211, and 231, are more likely to be used in conjunction with Section 109, NAAQS, than other possible combinations (Figure 4). For example, Section 202 of the Act specifies emission limitations for carbon monoxide, hydrocarbons, and nitrogen oxides from automobiles. Concurrently, ambient air quality standards have been set under authority of Section 109 for these same pollutants. However, standards for moving sources should not be expected to be used solely with Section 109. They could also be used with Sections 111 and 112 to control the same pollutant if specified criteria of the Act are satisfied.

2.1.6 Total Ban

If health data warrants, a total ban on emissions could be achieved directly under the hazardous standard option, indirectly under the ambient standard option through stringent levels, and directly under the new and/or moving source options if an adequately demonstrated system exists to achieve zero emissions (taking cost into account).

Using the preceding mechanisms, alternatives and criteria needed in selecting the preferred standards path (selecting a particular option) are illustrated in Figure 4. Decisions listed in Figure 4 are qualified as "most likely appropriate" because the Act is subject to interpretation (Appendix A). Since the Act allows flexibility, the choice of a particular option may create controversy because another alternative

could be just as valid. However, the first requirement for taking any action under the Act is the establishment of adverse health and/or welfare effects. Once this is confirmed, alternatives can be evaluated, using Figure 4 as a guide.

2.2 The Preferred Standards Path

Now, available information for BaP will be examined in relation to the first block in Figure 4, i.e., establishment of the possibility of adverse health or welfare effects. Specifically, the following reasons are sufficient justification to consider Federal control of BaP.

a) BaP is a proven carcinogen at elevated levels in animals under experimental conditions.

b) Epidemiological evidence connects lung cancer etiology with occupational exposure to BaP, and other coal tar volatiles, in specific industries. Data relating to each of the six options will now be presented in the order shown in Figure 4.

2.2.1 NESHAP

Although an apparent urban factor exists causing a 2.0 to 2.5 times increase in lung cancer incidence, no evidence exists to link this increase solely to BaP or other POM in the ambient air. Many areas with high lung cancer incidence (Appendix B) do not coincide with high ambient concentrations of BaP as measured by the NASN prior to 1970 (Appendix C). Unfortunately, many cities with high lung cancer incidence have not been sampled for BaP. Nevertheless, experimental animal studies and limited epidemiological studies do suggest that BaP may cause or contribute to lung cancer incidence. Naturally, lung cancer results in an increase in mortality and/or an increase in serious irreversible, or incapacitating reversible, illness.

However, data do not support that these hazardous effects (e.g. lung cancer incidence) are due to inhalation of (or other exposure to) existing or anticipated atmospheric concentrations of BaP. Neither do data define levels at which effects occur as no threshold of effects can be conjectured. In short, with current data and extrapolation techniques BaP has not been shown with certainty to be hazardous at ambient levels. Of course, the Act does not require that ambient levels be the deciding factor, but from a practical point of view, effects should be related to levels that people are likely to receive in routine activities. If these are occupational levels then EPA would not be the agency to establish standards. Also, the nature and distribution of sources (block 5) should have a bearing on the decision reached. If sources other than stationary contribute a significant amount of BaP to the total then Section 112 of the Act would not be adequate remedy. Thus, the conclusion is that the use of Section 112, NESHAP, for BaP control cannot be justified at this time.

2.2.2 NAAQS

For BaP the NAAQS option (block 7) must be examined because both source classifications, moving (block 8) and stationary (block 9), contribute to ambient concentrations. The moving source classification also satisfies the criteria of numerous or diverse. However, available data do not support a specific concentration for an ambient standard. Establishment of an ambient air quality standard requires the identification of a defensible air quality number (including an adequate margin of safety) where adverse health effects are observed. Arbitrarily establishing a number without sound medical support for it would be

inconsistent with the Clean Air Act. Thus, the conclusion is that Section 109, NAAQS, is not an appropriate regulatory tool for controlling BaP because evidence to support an ambient air quality standard for BaP is not available.

2.2.3 ESFMS

If the decision block (no. 8) for ESFMS in Figure 4 is chosen because moving sources are a significant source of the candidate pollutant, two options become available. The first option, emission standards, does not appear feasible because emissions from mobile sources are insignificant as shown in Table I. This estimate does not include emissions from aircraft or diesel-burning moving sources. No estimate has been attempted for aircraft but emissions are generally believed to be slight. Emissions from diesel engines have been estimated to be minimal.

If the emission standards option were not selected, it would cause a shift to the regulation of fuels option shown in the diagram (block 11). Based on negligible emissions of BaP from gasoline burning engines the ROF option can not be justified. Thus, based on available data, emission standards and fuel regulation do not appear feasible.

2.2.4 NSPS

While it is concluded that data do not support action by any of the previous options, the question remains whether Federal action should be considered under Section 111 of the Act. Data presented thus far verifies that Section 111 would be the only feasible regulatory option at this time if additional regulatory action is needed.

Section 111 of the Act authorizes the Administrator to establish standards of performance which reflect best systems of emission reduction for categories of new sources. The Act further requires States to establish emission standards for existing sources of the same categories for pollutants not listed under Sections 108 or 112. Section 111 can be used if the source category "may contribute significantly to air pollution which causes or contributes to the endangerment of the public health or welfare." To be defensible, performance standards under Section 111 must be applicable to significant categories of BaP. Sources in these categories are identified in Table I based on available emission estimates (Section 3.0).

Quantitatively, emissions of BaP to the ambient air are minimal (Table I). However, since BaP is a proven carcinogen (and/or cocar-

Table I. MAJOR SOURCES OF ESTIMATED BaP EMISSIONS IN THE UNITED STATES BY DESCENDING QUANTITY OF CONTRIBUTION (1971-73)

Source type	BaP emissions (tons/year)	% of total
1. Coal refuse fires	310	34.7
2. Residential furnaces, coal (hand-stoked)	300	33.6
3. Coke production	170*	19.0
4. Vehicle disposal (open burning)	25	2.8
5. Wood burning (fireplaces, etc.)	25	2.8
6. Mobile sources, gasoline	11	1.2
7. Forest and agricultural burning	11	1.2
8. Tire degradation	11	1.2
9. Open burning (domestic - municipal)	10	1.1
10. Intermediate coal furnaces	7	0.8
11. Petroleum refineries	7	0.8
12. Enclosed incineration (apartment - municipal)	3	0.3
13. Other	4	0.5
TOTALS	894	100.0

*A range of emissions has been given for this value. Appendix F contains the details.

cinogen) under given conditions, it may be considered to contribute significantly to air pollution. This contribution of BaP is from about three major categories of sources that are amenable to control, using either Section 111 or other means (e.g. local ordinance). Emissions of BaP should be reduced because it is a carcinogen.

2.2.4.1 Alternatives to Control Under Section 111

Two major alternatives under this option are:

- a) Control all major sources of BaP by §111, assuming adequately demonstrated control techniques, or
- b) control only selected sources of BaP.

Either of these alternatives could be used but would result in a completely new national strategy for BaP only, thus invoking §111(d) since BaP is not regulated directly under §§109 or 112. Strong evidence indicates that such a specific strategy is not warranted due to other control actions in progress, as discussed in the next section.

2.2.5 Alternative Control Strategy

In addition to options already discussed and dismissed, another alternative exists: no direct Federal action, based either on the lack of demonstrated control technology or that existing Federal, State, or local controls are adequate and effective. Control technology does exist for, and has been applied to, many of the sources listed in Table I; for others it is non-existent (e.g., forestry burning) or very difficult to apply (e.g., burning coal refuse piles).¹⁷ Federal, State, and local agencies are also making progress in controlling BaP from all sources.

Analysis of ambient BaP concentrations shows a definite declining trend in all cities (except one) for which data are available. Ambient

concentrations of BaP analyzed in twenty-three cities with coke ovens show a decrease of 55% over a six-year period (1966-1972). Furthermore, trends analysis for the benzene soluble organics (BSO) fraction of particulate samples (that portion which includes several POM compounds) shows a 55% decrease for a ten-year period (1960-1970). (See Section 5.0.) Such definite declines as these indicate that the following positive actions by control agencies are having concurrent effects on ambient concentrations as measured at receptor sites.

1. Source No. 1, coal refuse fires, is regulated in four States which include the majority of coal refuse fires. Other States can control emissions from this source by open burning, or other, regulations. In addition, the Bureau of Mines, Department of Interior, has proposed Federal regulatory action for these sources and are requiring spontaneously started fires to be controlled under existing Federal regulations.
2. Emissions from coal-fired residential sources decreased 28% between 1968 and 1972^{3,30} and this trend is expected to continue for the following reasons:
 - a) Socio-economic conditions have established a trend away from coal-burning in the home. Urban renewal is underway in practically every U.S. city. All of these projects avoid the use of coal for heating because it is dirty, relatively inconvenient to use, and in many places cannot meet air quality emission standards. The manufacturer of units which burn

solid fuel for home heating has declined to the point that an industry directory for 1973 did not list coal-fired units as being manufactured.

- b) All States except Arkansas have promulgated general particulate emission regulations for small fuel burning sources. These regulations, in effect, require particulate collectors which are not cost-effective for residential sources. Five States limit maximum emission from these sources to 0.8 lbs of particulates per million BTU; this rate is slightly less than estimated emission rates for coal-fired home furnaces. Of the remaining forty-four States, twenty-two limit emissions to 0.6 lbs per million BTU, and the rest require less than $0.6 \text{ lb}/10^6 \text{ BTU}$. For example, Oregon limits particulate emissions to $0.33 \text{ lb}/10^6 \text{ BTU}$ and Connecticut has a limit of $0.2 \text{ lb}/10^6 \text{ BTU}$ while Alaska limits emissions to $0.1 \text{ lb}/10^6 \text{ BTU}$. These regulations have been a contributing factor in the phase-out of coal-fired residential furnaces.
- c) Residential coal-fired furnaces are banned directly in some urban areas, such as Chicago, St. Louis and Milwaukee. Others have such stringent sulfur content regulations that coal is banned indirectly. For example, Detroit, Philadelphia, and New York, allow a maximum of 0.3% sulfur content.

3. Emissions from all other sources listed in Table I are in the process of being reduced, or are scheduled to be in the near future. The remainder of the sources contribute less than one-third of the total quantity of BaP nationwide and are dispersed fairly widely across the U.S. Moreover, the majority of these remaining sources are amenable to control and will likely be controlled by various means in meeting primary ambient air quality standards for particulates or other pollutants. For example, by-product coke ovens (Appendix D) are being controlled through Federal enforcement actions as well as State and local actions. At least fourteen enforcement actions are in progress and more are expected. In addition, EPA is gathering data through its demonstration projects in preparation for proposing performance standards for new sources (Section 111). These NSPS's for particulate and hydrocarbon controls are expected within the next twelve months.

These actions that are in progress and downward trends of ambient concentrations indicate that major sources of BaP are being controlled.

2.2.6 Advisability of Federal Control for BaP

The advisability of Federal control should be based on an evaluation of the magnitude of effects of ambient BaP as well as effectiveness of existing and planned actions. Available data have

already been presented which identify those factors of importance in deciding whether Federal action should be specific for BaP. These data are restated here for emphasis. First, evidence to show that ambient POM is a health problem does not exist. Secondly, a significant negative trend in ambient BaP concentrations indicates that such a relationship to adverse health effects will be even more difficult to establish. Controls by State/local agencies and those resulting from Federal enforcement actions should assist in continuing this downward trend. Finally, NSPS for particulates and hydrocarbons expected by late 1975 should also result in further control of new sources.

2.3 Recommendations

As a result of appraising available options and supporting data and considering the advisability of Federal action, the following recommendations are made.

1. In order to minimize POM emissions EPA should continue to support those national and local actions that are responsible for the downward trends in BaP levels. However, a Federal regulatory program designed specifically for control of BaP is not warranted at this time.
2. Monitoring of ambient concentrations of BaP in selected cities should be continued on a routine basis. Specifically, quarterly composited NASN hi-vol samples for BaP concentrations at the same 30-40 sites studied for this report (Appendix C, Table C-2) should be analyzed routinely. Section 7.0 gives additional information on current research under way by NERC/RTP.

3. Special emphasis should be given to prompt enforcement of existing regulations for coal combustion, incinerators, open burning (including coal refuse piles), and coking operations. The same emphasis should be placed on new regulations as they are promulgated for these and other sources of POM.

3. SOURCES OF BaP

The literature indicates that BaP usually is associated with particulate matter.³ Formation of BaP generally accompanies any incomplete combustion of fossil fuels or organic compounds, especially coal. The amount of BaP produced and emitted is postulated to be directly proportional to the efficiency of combustion and control techniques applied. Of course, controlled combustion is not the only source of BaP; some occurs from natural causes, e.g. forest fires. Also, natural synthesis of BaP has been postulated by several scientists.¹¹ The more common sources of BaP are listed in Table I and will be mentioned in more detail below. Values given in this section are estimates based on the best available information. Methods used to arrive at these estimates may be obtained from the references.

3.1 Stationary

The category of stationary sources of BaP encompasses a wide variety of processes that contribute to local BaP concentrations and accounts for 98% of the nationwide estimate. This category is subdivided into heat and power generation, refuse burning (which includes forest and agricultural burning), and industrial activities. The most comprehensive examination of stationary sources of BaP available is an EPA publication (AP-33) by Hangebrauck⁸ et al. Other POM compounds were measured also, but BaP received primary emphasis; this, results of emissions are reported as units of BaP.

3.1.1 Heat and Power Generation

The majority of heat and power produced and used in the United States comes from fossil fuels (coal, oil, gas, or wood) and accounts for about 37% of nationwide BaP emissions. Of these the primary source of BaP appears to result from coal combustion in hand-fired residential furnaces. Data from all four fuels, in terms of quantities of BaP produced, may be seen in Table II. Emissions are dependent on efficiency of combustion rather than the type of fuel used. This is shown by the low emission factor for intermediate size coal units and coal-fired steam power plants, in relation to the gross heat input. Oil and gas have relatively low emission factors, while the emission factor for wood is low when compared with hand-stoked residential furnaces. These data are consistent with knowledge of POM formation processes, i.e., reducing conditions caused by insufficient oxygen. As may be expected, local variations in quantities of emissions will exist in given areas of the United States depending on the type fuel, number of industries, and other local conditions.

Total BaP emissions given in Table II must be regarded as subject to change because of several developments. The two main developments relate to two residential heating practices, woodburning fireplaces and residential heating with coal. These type units have the highest emission rates and are affected by trends of an affluent society. Wood-burning fireplaces are on the increase in popularity in new urban areas; local air pollution control agencies are banning, or attempting to ban, residential coal burning. Based on available data the total BaP emissions from heat and power generation in the United States are estimated at approximately 340 tons annually.³⁰

Table II. ESTIMATED BENZO(a)PYRENE EMISSIONS
FROM HEAT AND POWER GENERATION SOURCES IN THE U.S., 1972*

Type of Unit	Gross Heat (BTU/hr)	Benzo(a)pyrene Emission Factor (ug/10 ⁶ BTU)	Benzo(a) pyrene Emissions (tons/year)
Coal			
Hand-stoked residential furnaces	0.1 x 10 ⁶	1.7-3.3 x 10 ⁶	300
Intermediate units (chain- grate and spreader stokers)	60-250 x 10 ⁶	15-40	7
Coal-fired steam power plants	1,000-2,000 x 10 ⁶	20-400	<1
Oil			
Low-pressure air-atomized	0.7 x 10 ⁶	900	2
Other	0.02-21 x 10 ⁶	100	
Gas			
Premix burners	0.01-9 x 10 ⁶	20-200	2
Wood	-	5 x 10 ⁴	25

*Adapted from references 3 and 30.

3.1.2 Refuse Burning

In refuse burning, as in coal burning, efficiency of combustion governs BaP emissions. Inefficient combustion occurs in all open burning and also in most small incinerators; thus, formation of BaP results. On the other hand, enclosed incinerators designed and operated for specific tasks should result in efficient combustion and less BaP emissions. Presently, this category contributes about 40% to the national total.

Hangebrauck et al⁸ give BaP emission factors for: (1) municipal and commercial incinerators for certain wastes; (2) municipal and agricultural open burning; and (3) junked vehicle disposal. As ex-

Table III. ESTIMATED BENZO(a)PYRENE EMISSIONS
FROM REFUSE BURNING IN THE UNITED STATES
(1968)*

Source of Benzo(a)pyrene	Benzo(a)pyrene Emissions (tons/year)
Enclosed incineration	
Municipal	<1
Commercial and industrial	23
Institutional	2
Apartment	8
Open burning	
Municipal	4
Commercial and industrial	10
Domestic	10
Forest and agricultural	11
Vehicle disposal	50
Coal refuse fires	340

*Adapted from reference 3: an estimate based on 1972 data indicates a total of approximately 340 tons being emitted, ^{29.30} accounted for as follows:

- a) coal refuse fires - 310 tons
- b) forest and agricultural - 11 tons
- c) vehicle disposal - 5 ²³ tons
- d) other open burning - 10 tons
- e) enclosed incineration - 3 tons

The method of estimating emissions may have varied for the two years.

pected, these emission factors vary widely and again show the importance of efficient combustion. Large municipal incinerators (50-250 tons of refuse/day) have a BaP emission rate of 0.1 - 6 mg/lb of charged refuse. Commercial incinerator (3-5 tons/day) emission factors ranged from 50-260 mg/lb. Emission factors for municipal open burning are given an average value of 150 mg/lb while vehicle disposal results in about 13000 mg/lb of refuse. Higher emissions of BaP from refuse burning than originally estimated (20 tons/year) by Hangebrauck et al have been

projected recently. These later emissions, shown in Table III, are based on higher estimates of nationwide refuse burning, and the inclusion of coal refuse fires, rather than significantly different emission factors. The single largest contributor identified is the coal refuse pile under "Open Burning." Such waste piles are common in coal mining areas and are frequently referred to as culm (or gob) piles (or banks).

The given estimates should be regarded as an order-of-magnitude approximation because of the uncertainty of factors used to estimate emissions. Therefore, approximately 340 tons/year of BaP from refuse burning is the best available value.

3.1.3 Industrial Activity

Some industrial processes are conducive to direct source measurement while others must use indirect means, often imprecise, for estimating emissions. The former category includes catalytic cracking of petroleum and air-blowing of asphalt, probably the most obvious sources of BaP in the petroleum refining industry. The latter category of emissions estimating, indirect sampling, may be illustrated by coke production in the steel industry; carbon black, coal-tar pitch, and asphalt hot-road-mix processes; and general chemical processes. The procedure generally used consists of sampling the ambient air as near the expected source, or complex of sources, as possible and back-calculating emissions. This technique necessarily yields much less accurate results because of background ambient concentrations and other factors.

Industrial emissions of BaP are summarized in Table IV and account for about 20% of the national total. Experience has indicated that those industrial processes that must be sampled indirectly contribute insignificant quantities of BaP to the environment, with the exception of coke production. Estimates of BaP emissions from coke ovens range from about 0.06 tons per year to approximately 170 tons/year (Appendix F). The higher value is used here because it is based on data from the U.S. The high BaP discharge from coke production appears to be associated with the effluent from the charging and coking processes. A crude emission factor of 2.5 g of BaP/ton of coke produced was applied to estimate BaP emissions from coke production. A more accurate assessment of coke oven emissions is being developed.

Table IV. SUMMARY OF ESTIMATED INDUSTRIAL
BENZO(a)PYRENE EMISSIONS IN THE UNITED STATES
(1972)*

Source of Benzo(a)pyrene	Benzo(a)pyrene Emissions (tons/year)
Petroleum	7
Asphalt air-blowing	<1
Coke production	(0.06-) 170

*References 31; 8; and Appendix F.

Obviously, other industrial processes are sources of BaP (and POM) but are more localized and less significant as a nationwide problem. Thus, the current best estimate for BaP emissions from industrial sources is about 175 tons/year in the United States.

3.2 Mobile

Generally, when one thinks of mobile sources, thoughts of automobiles immediately enter one's mind. Of course, sources other than the automobile contribute to this group; for this reason, many people prefer to call this category "transportation" sources rather than "mobile." Because of difficulties associated with the type testing required, literature^{2,4,7,32} on vehicular sources of BaP emissions is sparse.

3.2.1 Gasoline-powered Vehicles

Of the various transportation sources contributing to emissions of BaP, the automobile has been studied the most. Contributions of gasoline-powered vehicles can be separated into vehicular characteristics and fuel composition effects. The first category includes the effects of the air:fuel ratio; emission control devices; driver operating modes; engine deterioration; and combustion chamber deposits. The second category is composed of fuel aromaticity, BaP levels, additives, and lubricants.

As with other sources of BaP, efficient combustion apparently results in less BaP emissions. Air:fuel ratios greater than stoichiometric, i.e. use of excess air, promote efficient combustion in the cylinders of internal combustion engines. Recent data indicate that BaP production decreases about 30-fold with an increase of the air:fuel ratio from 10:1 to 14:1.²

Various researchers^{4,7,32} have shown that other factors mentioned above also increase emission quantities of BaP. Engine age causes deterioration of operating parts, thus permitting more oil consumption and concomitant BaP emissions. Also, excessive wear creates more space for deposits to accumulate in the combustion area. These deposits, through a complex process, cause increased BaP emissions.

Since BaP is composed of aromatic compounds, it stands to reason that the more basic aromatics in the fuel the easier for BaP to be formed and emitted. However, based on recent research, higher fuel aromaticity may be offset by using unleaded gasoline which changes the character of combustion chamber deposits.³² Other research in progress tends to dispute such a conclusion. Nevertheless, one thing appears to be certain, with the use of unleaded gasoline the distribution of BaP emissions from mobile sources will be different. Development of emission control devices introduces further complicating variables in the overall effect but studies thus far indicate these devices are beneficial.³²

3.2.2 Diesel-fuel-powered Vehicles

Studies on diesel-fuel-powered vehicles have been limited mainly to trucks and buses under laboratory conditions. Actual on-the-road operation of diesel-powered vehicles can result in high BaP emissions due to engine overloading, poor maintenance, and other factors. If diesel engines are not overloaded, laboratory tests have indicated that higher BaP emissions occur at idle than at any load condition. Furthermore, these same tests indicated that half-load produced highest BaP emissions during operation. Emissions dropped sharply at full load, presumably because of increased combustion chamber temperatures resulting in more efficient burning.³

Other mobile sources likely contribute to BaP emissions but they are more localized and quantitative data are lacking, especially for aircraft and turbine engines. Aircraft and turbine engines apparently have never been surveyed directly for BaP emissions. Some soil sampling investigations around airports undertaken by different researchers¹⁴ suggest aircraft may be a significant emitter of BaP. Degradation of automobile tires is likely the major non-combustion, mobile source of BaP. Carbon blacks, used in tire manufacturing, contain BaP and other high molecular weight organic compounds. A summary of estimated quantities of BaP from mobile sources is given in Table V.

Table V. ESTIMATED VEHICULAR BENZO(a)PYRENE EMISSIONS
IN THE UNITED STATES (1970)*

Vehicle source	Fuel consumed (gal/year)	Benzo(a)pyrene emission factor ($\mu\text{g/gal}$)	Benzo(a)pyrene emissions (tons/year)
Gasoline-powered			
Automobiles	56.4×10^9	170	10
Trucks	24.2×10^9	500	12
Diesel-fuel-powered			
Trucks and buses	5.8×10^9	62	0.4
Tire degradation	-	-	10
Total			32

*Adapted from reference 3; an updated estimate of BaP emissions from these sources for 1972 and 1973 is 22 tons annually,^{30,32} computed as follows:

- a) for gasoline-powered sources, a ratio was established between 1970 and 1973 gasoline consumption and related emissions with the assumption that 85 percent of the vehicles are controlled by 70 percent for BaP emissions in 1973.

$$\frac{80.6 \times 10^9 \text{ gals}}{103 \times 10^9 \text{ gals}} = \frac{22 \text{ tons}}{x}$$

$$\therefore x = 28 \text{ tons} \times 0.40 \text{ (noncontrolled)} = 11 \text{ tons BaP}$$

- b) for tire degradation the emission factor has been estimated at 0.3 lb BaP/day/ 10^6 population. 1972 population is 208×10^6 .

$$x = 0.3 (208) \left(\frac{\text{ton}}{2000 \text{ lb}} \right) \left(\frac{365 \text{ day}}{\text{yr}} \right) = 11.4 \text{ tons BaP}$$

- c) Diesel-powered sources were estimated to be about the same.

3.3 Natural

Data presented thus far provide good substantiation that BaP is primarily a product of incomplete combustion. However, a few sources of BaP may be defined as naturally occurring. The first of these has already been alluded to--it is bituminous coal. In addition to being analyzed for BaP content, coal has also yielded benzo(a)anthracene and other unspecified POM. Two of three types of asbestos used industrially contain appreciable quantities of natural oils. These oils have been found to contain BaP.¹¹

Two additional sources may be more difficult to verify but no question of their natural occurrence should arise. One researcher¹¹ has identified various molds in the environment as a source and BaP. Several scientists have indicated that BaP is synthesized naturally in the environment.

4. CONTROL TECHNOLOGY

The nature of emissions referenced above provides a broad hint to control procedures. Since inefficient combustion of fossil fuels is a primary source of BaP, by far the most important control technique is to burn fuels efficiently. Fuel or process substitution may be directly applicable to many processes of BaP emissions. For example, gas or oil are inherently more efficient than coal; incineration is much more desirable than open burning. These two methods have been adopted widely recently.

A desirable end result may be possible only by adding control equipment in line with the regular process. In particular, reduction of BaP from a source may be achieved by treating the effluent gas stream. Bag filters, scrubbers, or electrostatic precipitators are standard devices for removing particulates upon which BaP is likely absorbed. Afterburners, catalytic mufflers, or condensation may be applicable techniques to control vapor phase POM.

4.1 Stationary

Since the most important contributors of BaP emissions in heat and power generation are hand-fired coal furnaces and wood burning, the preferred choice of control would be by alternative fuel sources. One might argue that substitution is the only means of

control for these two sources as neither is amenable to better engineering controls.

In regard to refuse burning, efficient incinerators are being installed for municipal, commercial, industrial, and apartment building sources. Relative contributions from these sources are diminishing in comparison to coal refuse banks. Existing burning refuse banks are required to be extinguished if burning began spontaneously. Federal regulations have been proposed for all burning coal refuse piles which will eliminate most emissions as these regulations are implemented. Future culm piles are being eliminated by more specific attention to refuse accumulation practices in mining areas.¹⁰

Benzo(a)pyrene emissions from catalytic cracking in the petroleum industry apparently are receiving necessary action through utilizing carbon monoxide (CO) waste boilers for effective control (see Table VI). From Table VI it is clear that CO waste-heat boilers have a significant beneficial effect on BaP emissions, and thus on other POM emissions.

Much of the BaP produced at by-product coke ovens (Appendix D) quite likely could be removed in the by-product stream, except for that escaping from a leaky system. However, BaP emissions from most coke processing still appears to be quite high, whether it originates from charging, leaks, or directly from the gas stream. Some possible approaches to controlling BaP from coke ovens are under investigation by EPA and industry groups; two demonstration projects for particulate

Table VI. ESTIMATED BENZO(a)PYRENE EMISSIONS
FROM CATALYTIC CRACKING SOURCES IN THE UNITED STATES
(1968)*

Type of Cracking Unit	Petroleum Consumption (million barrels/year)	Benzo(a)pyrene Emission (tons/year)
FCC		
no boiler	424	0.08
CO boiler	1,230	0.02
Subtotal	1,654	0.10
HCC		
no boiler	14	3.4
CO boiler	55	0.0
Subtotal	69	3.4
TCC (air-lift)		
no boiler	27	2.4
CO boiler	118	0.0
Subtotal	145	2.4
TCC (bucket-lift)		
no boiler	17	0.0
CO boiler	75	0.0
Subtotal	92	0.0
Total	1,960	5.9

*Adapted from reference 3; reference 31 gives 1973 petroleum consumption as 2,372 million barrels/year which yields about 7 tons of BaP annually.

control from coke ovens are in progress and BaP sampling is being done simultaneously.

The Control Systems Laboratory, NERC/RTP, began coke oven particulate control demonstration projects in 1972. These projects were designed to demonstrate the feasibility of controlling particulate emissions from coke ovens, on both the charging and pushing sides of the oven. Particulate control demonstration projects are, for the most part, being accomplished on existing coke ovens. The purpose of demonstrating the availability of control technology was to gather background information for standards of performance for new coke ovens.²² However, since the demonstration projects are on

existing sources, preliminary results indicate that particulate controls are technically feasible for existing as well as new sources. Since coke oven controls are complex and many questions must be answered, the various projects underway by EPA are divided into several phases. Only initial phases of both charging and pushing control demonstration have been completed. However, from observations during these phases, particulate control appears to be feasible.

In the charging-phase demonstration project, source sampling for BaP was performed in conjunction with particulate sampling. Preliminary analysis of sampling results indicate that about 90 percent control of particulates is achieved.²³ With this level of particulate control, a similar degree of reduction in BaP emissions was expected. Preliminary evaluation of the BaP fraction of the particulate from the controlled versus the non-controlled larry car (vehicle that drops coal into the oven) indicates that approximately 85-90 percent control is achieved.²³ If these results are obtained for other types of particulate control (e.g., pipeline charging*), control processes should also reduce BaP emissions. Such results confirm previous expectations of control engineers that particulate control will significantly reduce BaP emissions.

Presently, no data are available on the amount of BaP control achievable on the pushing side of the coke oven. The second phase of a demonstration project is underway to obtain control estimates on the pushing side. Based on observations of installed systems, engineers

* Pipeline charging is a method for pre-heating coal and charging it into coke ovens via pipelines in a totally enclosed system; licensed by Coaltek Associates.²⁴

estimate that control on the pushing side should work as well as controls on the charging side of the oven.²¹

Thus, we see from Tables I-IV that BaP emissions originate primarily from stationary sources, accounting for about 97 percent of total nationwide BaP emissions. However, due to inadequate sampling techniques these emissions should be considered estimates of relative ranking. Furthermore, since stationary sources account for the majority of BaP, these emissions are highly localized. Table VII depicts some degree of localization for the three major sources. Such localization indicates those areas where control efforts should be emphasized.

4.2 Mobile

Before the current concern for reducing vehicle emissions was instigated by the Clean Air Act, most vehicles operated with fuel-rich carburetion to promote smooth performance and quick power response. Preliminary modifications prior to the 1970 Clean Air Amendments resulted in leaner fuel-air mixtures and lower BaP emissions. This trend is indicated in Table VIII by the emission factors estimated in 1972 for the periods indicated. "Advanced systems" in Table VIII refer to prototype emission control devices spurred into development by the 1970 Act. Generally, advanced systems utilize catalytic converters and thermal reactors.

Emission control devices will effect control of BaP concomitantly. Data to support this view are based on current exhaust sampling techniques.³² If such a high degree of control is achieved in actual practice, mobile source pollution caused by BaP will assume a much

Table VII. CONTRIBUTIONS TO NATIONAL TOTALS OF
BENZO(a)PYRENE BY SOURCE AND STATE
(1972)*

Source of benzo(a)pyrene emission	State	Percent of U.S. total from given source contributed by given state
Coal refuse burning	Pennsylvania	46
	West Virginia	37
	Ohio	16
	Kentucky	
	Virginia	
	Alabama	
Total		99
Residential coal-fired furnaces	Illinois	23
	Pennsylvania	16
	Kentucky	7
	Ohio	7
	Tennessee	7
Total		60
Coke production	Pennsylvania	27
	Indiana	15
	Ohio	15
	Alabama	21
	Michigan	
	W. Virginia	
Total		78

*References 28; 33; and 34.

less significant role. At any rate, one may logically expect a reduction of BaP emissions from light-duty vehicles as a result of the EPA motor vehicle control activity. A higher degree of uncertainty exists for other type transportation sources, e.g., buses, heavy diesel trucks, aircraft, etc.

Table VIII
AUTOMOTIVE BENZO(a)PYRENE EMISSION FACTORS
(1972)*

Source	Benzo(a)pyrene emission factors (μ g/gal of fuel consumed)
Uncontrolled car (1956-1964)	170
1966 Uncontrolled car	20-80
1968 Emission-controlled vehicle	6-24
Advanced systems**	< 1-2

*Adapted from reference 3 & 32

**Prototype emission control systems

4.3 Natural

Control of BaP from natural sources is even more limited than other types control because natural emissions are less well-defined. For bituminous coal, product substitution would be the logical approach to control. Of course, suitable substitutes for specific use and application must be found. Asbestos control under NESHAPs should preclude BaP from the environment also.

As for control of BaP from natural synthesis and molds, no suggestion has been forthcoming. Their contribution to ambient concentrations is so negligible that it is likely not worth time and money required for exploration.

5. AMBIENT CONCENTRATIONS

Nationwide ambient concentrations for most pollutants of interest, including BSO and BaP, are available through the National Air Surveillance Network (NASN). Concentrations available through this network provide only relative nationwide comparisons because of limited sampling sites in a given area. An additional problem in using NASN data for ambient BSO and BaP concentrations is that none of these sites were selected specifically to sample for organic pollutants. However, representative data are available from some suspended particulate sites in the NASN. In addition, summary reports on POM reference various special studies that have been undertaken since the late 1950's.

If a detailed breakdown of NASN data by quarter were examined, a seasonal trend for BSO and BaP would be obvious with few exceptions. Higher concentrations would be noticeable for the fall and winter quarters at most sites while the summer generally has the lowest concentrations. Studies conducted in the late 1950's indicated this same pattern.^{12,13}

5.1 Measurement Technique

To better understand the measurement of ambient BaP data, a brief description of the process should be helpful. Particulate samples are collected upon tared fiber glass (flash-fired) filters by

drawing air through the filters at a relatively high volume of air flow (approximately 50-60 cfm) during a 24-hour day. These filters are sent to a laboratory for preliminary weight analysis, after which organics are extracted from a portion of the filter by the Soxhlet method for about six hours using a suitable solvent [ordinarily benzene--thus the term benzene soluble organics (BSO)]. Subsequently, BaP is separated from total BSO by thin layer chromatography (TLC) followed by ultraviolet (UV) spectral analysis.

A minimum of 50 mg of BSO material is required for a BaP analysis using TLC and UV. In order to obtain a sufficient quantity of sample for this analysis several organic fractions from one station must be pooled. Usually each urban 24-hour hi-vol particulate sample will contain about 25 mg of benzene extract. Generally, samples for a three month period, consisting of 5-7 samples, are composited to obtain one data point. Thus, an annual station mean would consist of averaging four quarters of composite samples for one station (see Appendix C). Table IX is an annual summary of BSO, BaP, and TSP for the last three years of analysis from 121 selected NASN sites where BaP was analyzed on a routine basis. Table C-2 of Appendix C lists those sites which were analyzed after a special request for 1971 and 1972 BaP data. TSP is an abbreviation for total suspended particulates, which is obtained from a weight analysis of hi-vol filters and relating this to average air flow for the 24-hour sample period. Note from Table IX that BaP trends follow the average BSO content and not TSP sampled.

Table IX. THREE YEAR SUMMARY OF TSP^a, BSO,
AND BaP* FOR 121 NASN SITES IN THE
UNITED STATES - 1968, 1969, AND 1970

Year	Pollutant		
	TSP ^a ($\mu\text{g}/\text{m}^3$) ^b	BSO ^a ($\mu\text{g}/\text{m}^3$)	BaP ^a (ng/m^3) ^c
1968	89 (88) ^d	5.7 (5.5)	2.4 (1.6)
1969	84 (81)	5.0 (5.0)	2.3 (1.6)
1970	96 (88)	4.3 (4.3)	2.0 (1.2)

*See Appendix C for the annual average for each selected site for each year.

^aTSP = total suspended particulate; BSO = benzene soluble organics; and BaP = benzo(a)-pyrene.

^b $\mu\text{g}/\text{m}^3$ = micrograms of sample/cubic meter of air.

^c ng/m^3 = nanograms of sample/cubic meter of air.

^dNumbers in parens are median station values for the 121 sites.

No known routine studies by State or local agencies are being conducted on ambient levels of BSO and BaP. Several States have inquired about analytical procedures but have given no indication of follow-through. At any rate, no BaP data from State or local programs are recorded in OAQPS's national data bank.

5.2 Ambient BaP Concentrations

Figure 5 is a graphical representation of annual average BaP concentrations for coke-oven and noncoke-oven cities for 1966 to 1972. This figure is presented to show the general trend in the two type cities. The overall trend is decreasing in each case. With the exception of the dip in 1968 for the coke-oven cities, and 1967 for

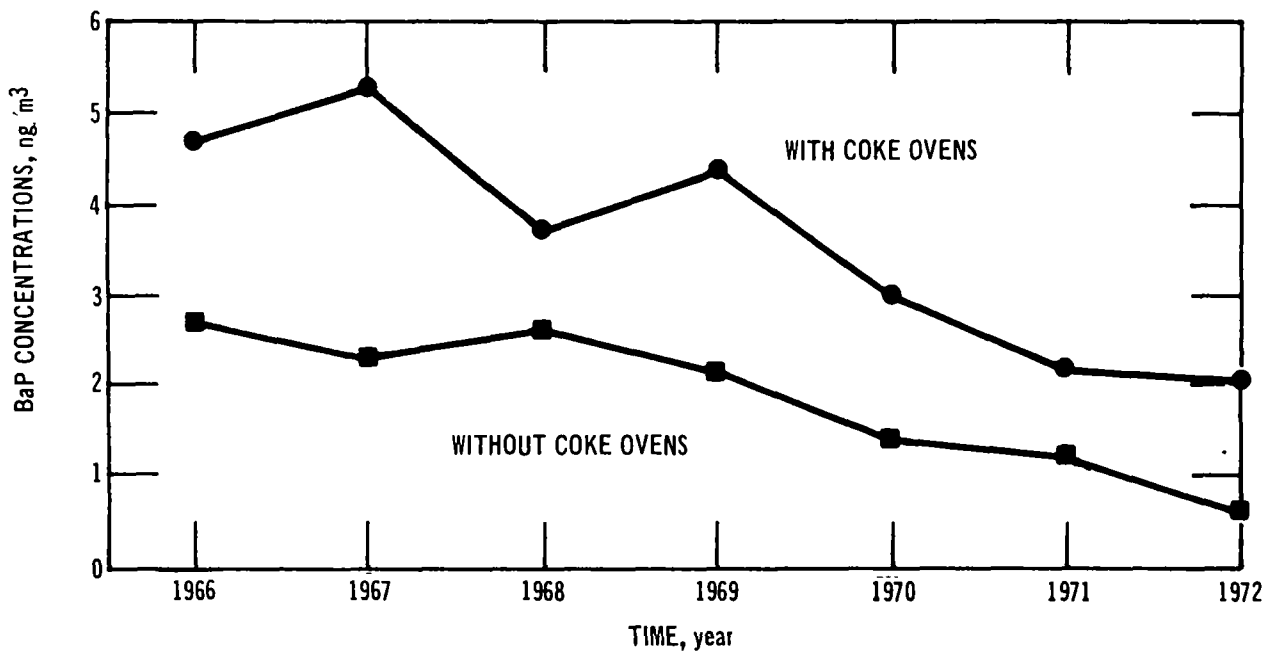


Figure 5. Trends in annual BaP concentrations in cities with and without coke ovens.

noncoke-oven cities, the trend is quite similar for both type cities. In each case, the number of cities included in the annual average varies from year to year because only those cities with a full year's data (4 valid quarters) were included (Appendix C, Table C-3).

Average BaP concentrations for coke-oven cities have decreased from 4.74 nanograms per cubic meter (ng/m^3) in 1966 to 2.14 ng/m^3 in 1972 (Figure 5 and Table C-3). This is a 55% decrease in a six-year period. The decrease in noncoke-oven cities is even more conspicuous: a 77% decline occurred over the same six-year period, with concentrations dropping from 2.76 ng/m^3 to 0.64 ng/m^3 .

Individual trends were decreasing for most of the 34 cities used in computing BaP averages. However, some cities showed an upward variation in some years. One city, Chattanooga, Tenn., showed no decline at all over the six-year period based on a rank correlation coefficient of zero. Two other cities, Pittsburgh, Pa. and Ashland,

Ky., showed a weak decreasing trend pattern with the rank correlation of BaP versus time of -0.14 and -0.40, respectively, because of positive fluctuations in the trend line.

Realizing that whatever is happening in the noncoke-oven cities is also happening in the coke-oven cities for the overall trend, one can still surmise certain occurrences from information available. Even though limited data do indicate that coke ovens are a major contributor to localized BaP concentrations, one must remain aware that coke ovens are not the sole source of BaP emissions. For example, the stockpiling of steel in 1967 in anticipation of a steel strike in 1968 likely contributed to the noticeable decrease of BaP concentrations in 1968 for coke-oven cities (Figure 5). On the other hand, an abrupt decline in BaP concentrations occurred in noncoke-oven cities in 1967, indicating the possibility that less coal was available for nonsteel industry use, especially small consumer use.

Again, one must remember that no readily evident method exists for identifying the specific contribution of coke ovens because the amount of coal delivered to individual cities is not available. However, the circumstantial evidence presented is quite strong. The trends analysis presented below establishes that a significant difference exists in BaP concentrations for cities with and without coke ovens.

5.3 Trends Analysis for BaP

Two statistical techniques were used to establish whether the apparently decreasing trend (Figure 5) in BaP concentrations was significant. These techniques were a linear regression and Spearman

rank correlation analyses. The Spearman rank correlation procedure, a nonparametric approach correlating the rank of BaP concentrations and time, was used to avoid the assumption of a certain distributional form for the data, and other requirements inherent in parametric correlation and regression analysis. This procedure was used on the data for each of the cities studied while the regression technique was applied to individual averages separately for the coke-oven and noncoke-oven cities. An analysis of variance (ANOVA) was used to establish whether a significant difference in BaP concentrations existed between coke-oven and noncoke-oven cities.

The general trend for BaP is decreasing. Regression analysis performed on the cities with and without coke ovens for BaP concentrations versus time verify that the trends shown in Figure 5 are significant, i.e., the decreasing pattern in BaP concentrations over time are unlikely to have occurred by chance alone under the hypothesis that a trend is not present in the data. Therefore, this suggests that the data support an alternative hypothesis of a trend component being present in the data. The linear regression for both the coke and noncoke cities could be described as parallel lines, i.e., lines with the same negative slope but being displaced by a significant difference in concentration between the two sets of lines representing the different cities (see Figure 5).

The Spearman rank correlation analysis was applied to the individual cities by looking at relative rankings of annual average BaP concentrations versus time (years). Of the 34 individual sites analyzed, all have negative correlation coefficients, except

Chattanooga which was zero. Of these, 12 sites were statistically significant. A statistically significant result ($\alpha = 0.05$) implies that an observed correlation coefficient was not likely to be derived from a population of coefficients whose true correlation is zero. Statistically, we can infer that a significant negative association exists between BaP and time for about 35% (12 of 34) of the sites. These results support the conclusion that a decreasing trend does exist for BaP concentrations over time.

In establishing a significant difference between cities with and without coke ovens an analysis of variance procedure was used to evaluate BaP concentration variations. With no way for considering the many additional uncertainties (other than time) which influence BaP concentrations in cities with coke ovens, the ANOVA shows that a statistically significant difference does exist between the two type cities. In fact, only a very small probability (0.0001) exists that the difference in the observed BaP concentrations between the two sets of cities could have occurred by chance alone, under the hypothesis that no difference exists in BaP concentrations between the two sets of cities. In other words, based on the seven year's available data, BaP concentrations are higher in cities having a coke oven, although it is impossible to evaluate the contribution of other sources of BaP to explain this difference.

5.4 National Urban and Nonurban Trends for BSO

Composite annual averages of BSO concentrations and BSO expressed as a percent of TSP are shown in Figure 6 for 32 urban and 19 nonurban NASN stations. The urban composite average concentrations

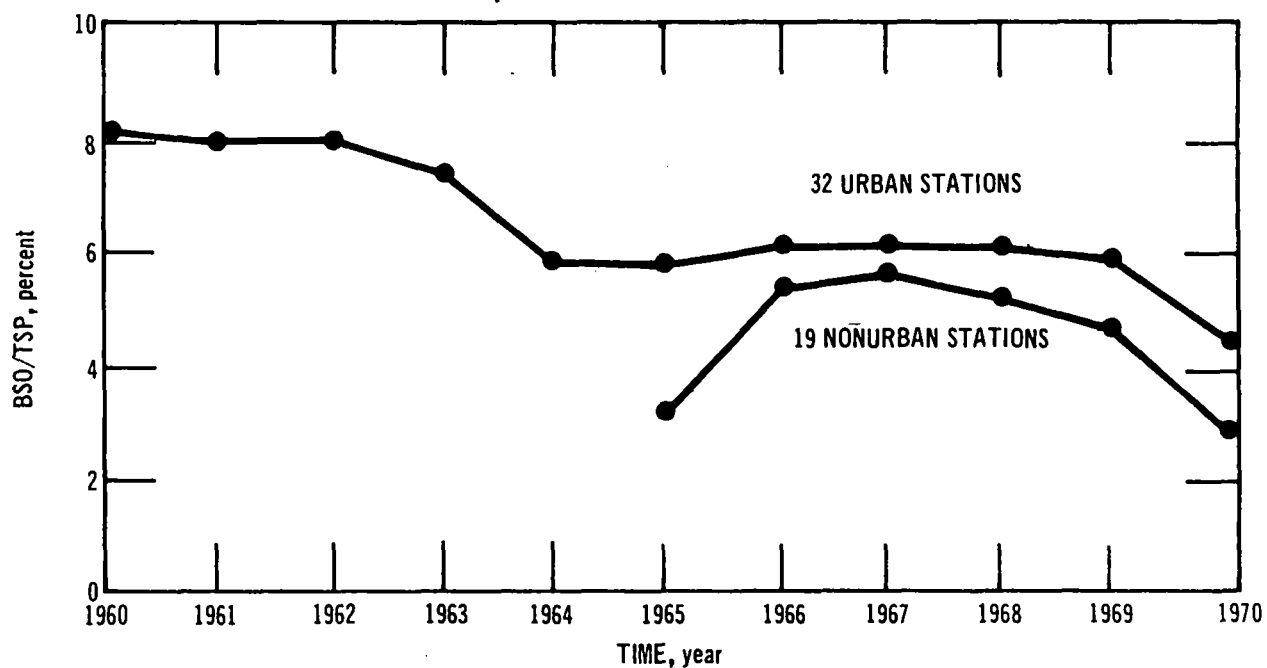
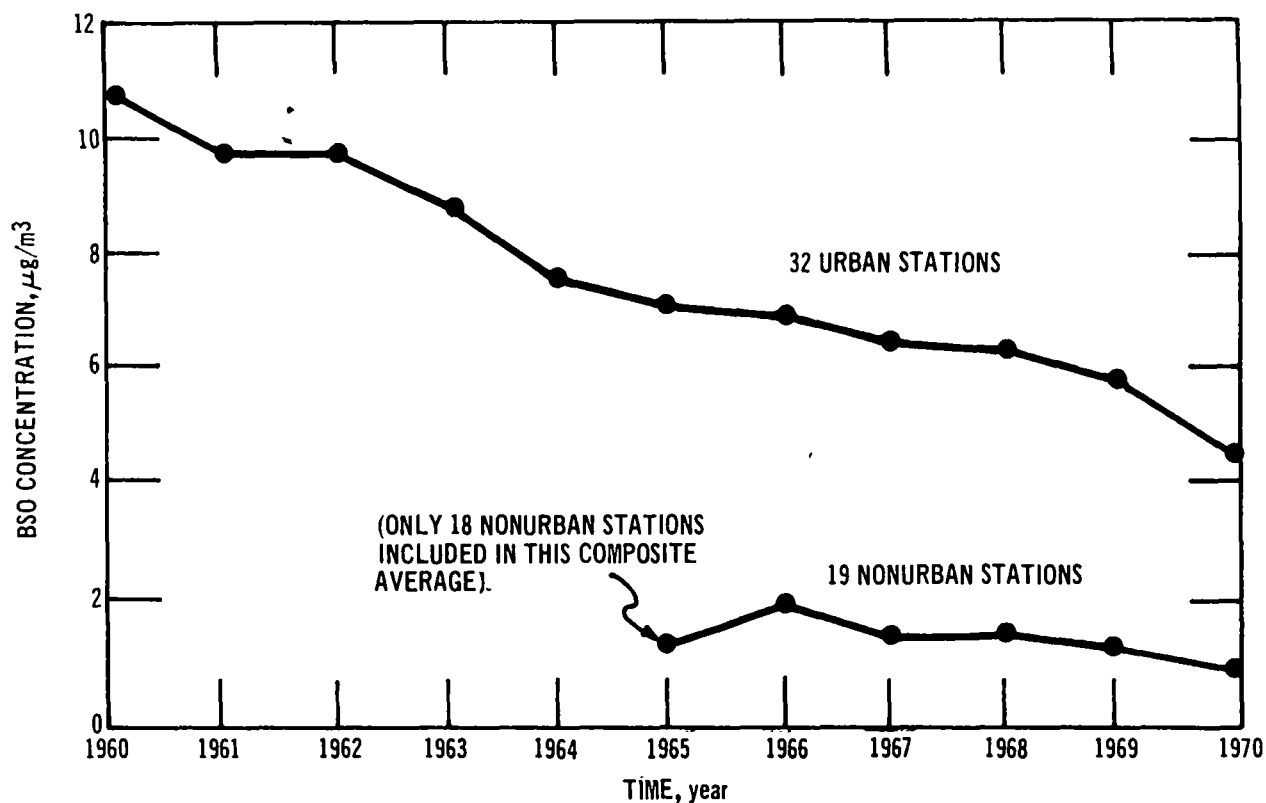


Figure 6. Trends in BSO and in BSO percentage of TSP at 32 urban and 19 nonurban stations. of BSO have decreased from $10.6 \mu\text{g}/\text{m}^3$ in 1960 to $4.8 \mu\text{g}/\text{m}^3$ in 1970. This represents a 55 percent decrease. The national trend in BSO concentrations shown closely parallels the trends observed at the vast

majority of the stations included in the composite sample.

Statistically downward trends were found for 27 out of 32 stations represented by this grouping. (The same statistical techniques described under BaP trends were used for BSO.) Only in Helena, Montana, was there some indication found of an increasing trend pattern. Portland, Oregon, did not exhibit a clear-cut trend pattern and was described as having "no trend," whereas Providence, Rhode Island, Seattle, Washington, and Milwaukee, Wisconsin, exhibited weaker decreasing trend patterns than was the general case. The overwhelming consistency in trend patterns points to some general phenomenon exclusive of geographical location within the country, city size, or industrial sources present in a particular city.

The decrease in the composite average concentrations of BSO in 1963 -- approximately $1 \mu\text{g}/\text{m}^3$ -- together with similar decreases in 1964 and 1970, represent the years with the largest concentration changes. The nonurban average BSO concentration, which is a factor of approximately 4 or 5 less than the urban averages, exhibits a more uncertain pattern. However, since 1966, nonurban BSO averages have decreased from $2.2 \mu\text{g}/\text{m}^3$ to $1.2 \mu\text{g}/\text{m}^3$ in 1970.

The BSO percent of TSP has also decreased for the urban stations over the period considered. The pattern is characterized by rather sudden decreases from 1963 to 1964 (8 to 6 percent) and from 1969 to 1970 (6 to 4.5 percent). In other years, the BSO percent of TSP remained relatively constant. It appears that significant decreases in national urban levels of both BSO and BSO percent of TSP have occurred, notwithstanding sizable, short-term, year-to-year fluctuations.

Nonurban trends in the BSO concentration and the BSO percent of TSP have also been downward during the 4-year period, 1966-1970. It is interesting to note that the averages of the BSO concentration and the BSO percent of TSP for the nonurban stations (with the exception of Curry County, Oregon) were lower in 1965 and 1966. However, insufficient pre-1965 data exist to judge whether trends in nonurban BSO concentrations have paralleled the concentration decreases at the urban sites.

6. EFFECTS OF BaP

Description of lung diseases related to dust or airborne particles was advanced first in the sixteenth century. Two other researchers demonstrated more than three hundred years later that lung cancer was a prominent pulmonary disease among miners.³ Now in the mind of the general public, lung cancer is associated with cigarette smoking because of superabundance of publicity, as well as "the warning on the label." The relation between cigarette smoking and lung cancer has stimulated interest in the role of air pollution in cancer. This interest has arisen because some pollutants found in urban areas, including BaP, are similar to those found in cigarette smoke. The necessity of defining all significant etiologic factors in lung cancer has become evident because of its rising incidence and poor prognosis. At this stage, primary prevention is the most effective means of lung cancer control.

6.1 Epidemiological Studies

Some types of comparison strongly support the hypothesis that increased lung cancer mortality rates are related to urban pollution. The epidemiologic method of studying air pollution effects on lung cancer incidence entails comparing lung cancer death rates in communities that have markedly different pollution levels, but are similar in other potentially causal factors. Comparisons between urban and rural areas within the same country are attractive in that, generally, fewer variables are involved and pollution level differences are maximized. The disadvantage

is that very little data on ambient concentrations of pollution are available for rural areas. Thus, comparisons between urban areas offer an advantage over comparisons between urban and rural areas but have other disadvantages in that different variables may affect the study. Each comparison requires a suitable adjustment for any known extraneous variables which may affect it.

6.1.1 Classification of Studies

Four groups of epidemiologic studies are used to arrive at evidence of environmentally related lung cancer incidence in humans.³ They are studies to:

1. Compare urban metropolitan populations with rural populations in relation to lung cancer mortality rates, usually without examining etiologic factors.
2. Compare lung cancer death rates in migrants to those in country of origin and those in country to which they migrate; and examine change in rate in relation to change in the environment.
3. Compare demographic units and study the relation between lung cancer death rates and various indices of pollution. Multiple regression techniques are generally used to separate environmental effects from other factors.
4. Sample characteristics of lung cancer decedents through family interviews and compare these with corresponding characteristics of the remaining population.

Through such studies the prospect is offered of identifying a relatively sharp distinction between strongly related factors and those incidentally associated with lung cancer.

Urban-rural differences undoubtedly offer clues to problems of pulmonary carcinogenesis in man. The general association between urbanization and increased lung cancer is not questioned. However, characteristics of the urban environment which are primarily responsible for lung cancer are a subject of controversy. The problem of identifying causal factors is amplified by difficulties of obtaining either accurate or extensive measures of exposure for most factors. Further intensification of the problem occurs from the close association of urban pollution factors with one another. For example, an area with high BaP levels will generally tend to have high concentrations of other hydrocarbons and sulfur dioxide.

6.1.2 Consideration of Other Variables

Special attention should be paid to imperfections or contradictions in the correlation of air pollution measurements and lung cancer incidence. General agreement apparently exists on the increment in the incidence of pulmonary cancer caused by smoking; interpretations differ, however. This is of immense interest, because urban air and cigarette smoke have carcinogenic substances, e.g. BaP, and some gases in common.¹³ Such similarity is likely the reason frequent reference is made to studies of cigarette smoking and pulmonary cancer. However, complexities of the problem of pulmonary carcinogenesis are compounded, rather than simplified, by that fact.

Considering the numerous variables that are known and anticipating that some are likely completely unknown, one should focus attention on the target tissue and consider the air as the primary point of contact with carcinogenic agents. Two specific variables which must

be considered in epidemiologic studies are irritant or toxic gases in the atmosphere and diseased tissues at the target site. These variables are difficult to quantify or to determine their specific relation to carcinogenicity. The common link between these two variables is their potentiating action on carcinogenic properties of BaP. Tissues altered by disease may incur a precancerous state which appears to contribute to carcinoma susceptibility.¹⁶

No attention has been given to effects on domestic animals because no information appears to be available. Of course, except for epidemiological studies, most data on the carcinogenicity of BaP have been obtained by using experimental animals. Various techniques have been used to verify carcinogenicity in these animals. A comprehensive review of these studies is presented in the NAS report³ and the NERC/RTP position paper on PPOH.¹⁶

6.1.3 Conclusions from Epidemiological Studies

Studies of lung cancer thus far have indicated the following conclusions, generally resulting from the four broad study classifications listed above.³

1. Lung cancer has emerged as the single greatest cause of cancer death in males and is a significant cause in females in the U. S. Its incidence has increased in the last 30 years.
2. A major etiologic factor appears to be cigarette smoking; however, smoking does not account completely for this increased incidence.

3. Urban dwellers have approximately twice as high an incidence of lung cancer as those living in rural areas after correcting for smoking. Within urban areas, incidence has been greater in areas of industrial pollution.
4. Benzo(a)pyrene, found in cigarette smoke in high concentrations, causes cancer of the lung and other organs in experimental animals. Also, it is present in the air at relatively high levels in industries whose workers have been found to have high mortality rates due to lung diseases, including cancer. Finally, it prevails at different levels in all urban communities investigated.
5. Generally, immigrants have an incidence of lung cancer between that noted for their countries of origin and that of the countries to which they migrate. The higher their ages at migration the nearer their incidence rates to their cohorts who do not migrate. In studies where the home country had a greater cancer incidence rate, rates of migrants decreased significantly, even if their cigarette smoking increased.

6.2 Other Effects

Other health effects of POM are related mainly to skin disorders from occupational exposure of man to high levels. Studies of these effects do not attempt to differentiate exposures by air from exposures by other means, e.g., contact. This aspect should be considered carefully in attempting to extrapolate the information derived for occupational problems to the possible hazards of ambient air pollution. No

documentation exists that particulate materials containing POM in ambient air have resulted in adverse skin effects.

Welfare effects of BaP are even more inconclusive because few specific studies have been conducted in this area. However, the probability of altering soil microbial populations appears to exist and such action may upset the ecosystem.¹⁶ The nature of BaP indicates that it may be produced in many plants or plant products; however, these data are speculative.

7. PROGRAMS TO STUDY BaP (AND OTHER POM)

Polycyclic organic matter has been studied for two hundred years among occupational workers. Descriptions of lung disease related to exposure to dust or airborne pollutants were said to exist as early as the 16th century. The first recorded description relating the burning of fossil fuel to occupational disease was written in 1775 by Percivall Pott. This was related to constant exposure to soot among chimney sweeps who had developed cancer of the scrotum. Then in 1879 two German scientists demonstrated that lung cancer was a prominent pulmonary disease among miners.³ Thus, occupational exposures provided the first evidence of cause-effect relations from airborne pollutants that now have been associated with BaP.

Since those early years many studies have been undertaken to define cause-effect relationships between airborne contaminants and lung disease and to identify the causal agent in airborne pollutants. In more recent years, beginning in the mid- to late 1950's, specific programs have been initiated to establish such a relationship with the frequency and complexity of studies increasing each year. Such has been possible because more and more people and institutions, or sponsors, have become interested in obtaining as much information as possible concerning BaP and other POM compounds. However, in all of this interest no forceful, common objective has been evident. As a result, much effort has been duplicated and much money utilized unnecessarily.

The purpose of this section is to give an analysis of those programs concentrating on BaP and other POM that are underway by NERC/RTP.

7.1 Current Research Projects

Table X is a synoptic listing of those programs which have a direct relation to emissions of BaP and/or other POM compounds. These projects were extracted from the NERC/RTP draft Position Paper on PPOM.¹⁶ Items of interest pertinent to the listed subjects will be given below.

7.1.1 Analysis Methodology

Several approaches to the improvement of analytical techniques are under investigation. The primary objective of most of these projects is to develop more rapid, and hopefully less expensive, analytical methodologies for various POM. Analytical techniques that can be easily applied are needed so that any agency can routinely monitor and measure BaP. Some analytical methods which are under investigation include projects 1, 2, 4, and 5 of Table X.

7.1.2 Effects Investigation

Project No. 11 has been delayed for about 1 1/2 years because exposure-room furnishings have been impeded by equipment delays. In the meantime, characterization of aerosols, design of containment systems, sampling techniques, etc. are continuing so that the inhalation study can be initiated immediately after the room is operable. The feasibility study listed as project 10 may be one of the more critical in relating BaP (and other selected POM) exposure to adverse human health effects. This project was begun in FY 1974.

Table X. APPLICABLE CURRENT POM RESEARCH PROJECTS¹

Project	Date begun (FY)	Scheduled Completion	Estimate of cost ² (\$1000)	Time required (Yrs.)
1. Development of fluorescent detector for analysis of carcinogenic HC	1972	1974	47	2
2. High pressure (up to 2000 psi) liquid chromatographic determination of total PAH	1972	1974	3	2
3. PAH profile in urban air (ratios of various compounds)	1972	1974	31	1.5
4. Rapid, direct gas chromatographic determination of BaP	1972	1974	54	1.5
5. Rapid GC determination of atmospheric PAH	1972	1973	55	1
6. Synthesis & purification of carcinogenic air standards	1972	1975	28	3
7. Characterization & control of air pollutant emissions from fuel combustion	1972	1977	200	5
8. Enclosed coke pushing & quenching demonstration	1972	1973	800	1
9. Smokeless coke & charging demonstration	1970	1973	950	3
10. Feasibility study: Determine which tissues best reflect PPOM exposures. Develop methodology for tissue analysis.	-	1975	200	1
11. Physiologic & pathologic responses of animals to inhalation of carcinogenic hydrocarbons	1973	1978	500	5
12. Development of stationary source sampling procedures for PNA	1973	1974	34.4	1

NOTES:

¹Taken from NERC/RTP Research Objective Accomplishment Plans (ROAP) and NERC/RTP Position Paper on PPOM (Draft), Dec. 1972.

²Estimates of costs for multi-year projects are for informational purposes only; they are not to be construed as accurate since an order of magnitude could separate estimates & actual costs. Single year projects scheduled to end in 1973 should be nearly accurate, discounting extension of completion time.

7.1.3 Stationary Sources

Four projects (#7, 8, 9, & 12) involving BaP (and other POM) from stationary sources were in progress during FY 1973. Three of them were initiated earlier by the Control Systems Laboratory. The fourth (# 12), which began the latter part of FY 1973, is a project to develop an improved source sampling procedure for BaP (and other POM). The initial phase will be for about one year with options to explore any problems in more detail if funds are made available. Project 8 was delayed approximately six months because of a job accident. Fuels referred to in project 7 are such things as methanol, low BTU gas, etc.

7.1.4 Ambient Analysis

Data available under this grouping should be of primary interest because ambient concentrations of POM compounds are ill-defined. Project 3 is of major importance in achieving a better understanding of ambient levels of BaP and other compounds. The objective of this study is to define, as accurately as possible in selected urban areas, specific ratios of BaP to other POM compounds and vice versa. Project 5 could easily fall in this category also, because standard compounds must be available for reliable analytical results. A limited number (about 6) of POM compounds were scheduled to be purified under the present contract.

7.2 Recommended Research Projects

A study which should receive early emphasis is the development of methodology to simplify and expedite ambient measurements. Analytical technique appears to be the primary area of need in expediting ambient measurements. The need for simplified analytical methodology was forcefully emphasized during preparation of this report when additional

ambient BaP concentrations for 1971 and 1972 were required for trends analysis in selected cities. A significant time delay was entailed because of time required to analyze for BaP. Simplified analytical methodology will expedite availability of ambient data for tracking trends of BaP (and other POM) concentrations. When new methodology is developed, it should be more easily applicable by State and local agencies also.

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APPENDICES

APPENDIX A - OVERVIEW OF A PREFERRED STANDARDS PATH

Generally, six major options for control of air pollutants are available to the Environmental Protection Agency (EPA) under the Clean Air Amendments of 1970 (Act): (1) National ambient air quality standards (NAAQS) - Sections 108, 109 and 110; (2) standards of performance for new sources (NSPS)- Section 111; (3) national emission standards for hazardous air pollutants (NESHAP)- Section 112; and (4) Title II, Emission Standards for Moving Sources (ESFMS) - Sections 202 (automobiles); 211 (regulation of fuels); and 231 (aircraft emissions). A seventh option for consideration, if data evaluation indicates it, is that of no Federal action. Other options may be identified by subdividing or combining the above seven options. The process of selecting the most appropriate option is called a preferred standards path (PSP) analysis. However, a PSP decision may not be possible if adequate data are not available for determining the preferred option. Thus, an eighth major option becomes available: postponement of a decision until more data become available. Of course, this latter option should carry with it recommendations of what data are needed and a suggested priority for obtaining such data.

A. Purpose of a Preferred Standards Path

The purpose of a PSP is to recommend a regulatory course of action to develop applicable standards resulting in control of

a given pollutant. Recommendations must be based on a thorough state-of-the-art assessment of emissions and health effects for the pollutant as related to objectives of the Act. Basic objectives of the Clean Air Act are described below.

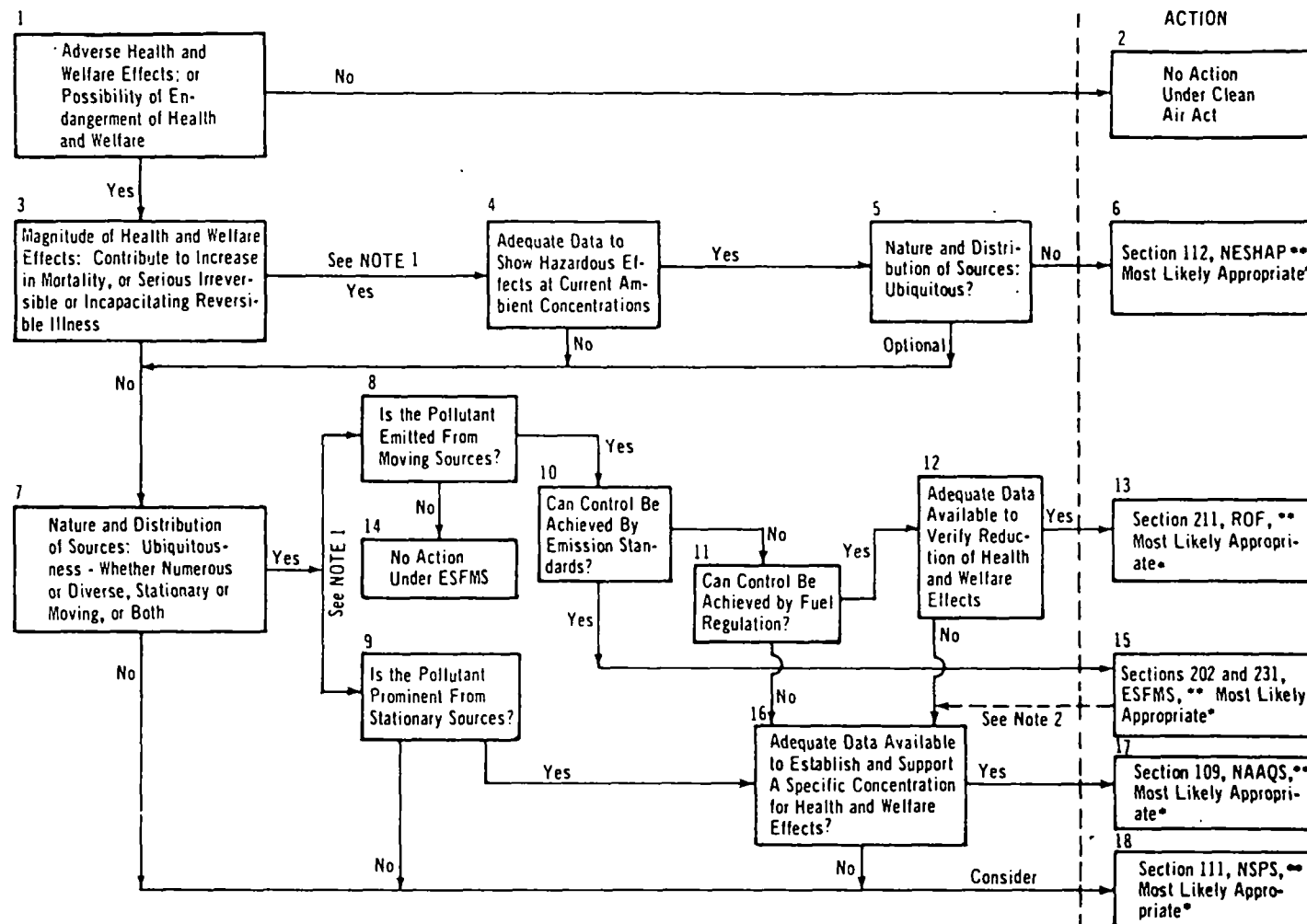
1. Protection of health and welfare by attainment and maintenance of a designated numerical ambient standard:
This objective is applicable to those pollutants for which adverse effects levels (health or welfare) may be determined and that exist in excessive concentrations which are relatively easily measureable in the ambient air. Usually, this goal is achievable by setting NAAQS following procedures set forth in Sections 108-110 of the Act.
2. Enhancement of air quality: This goal is for those pollutants which may be tolerated in exiguous quantities but their increase is limited in order to prevent the creation of new problems. As related to stationary sources, this goal may be achieved best by applying Section 111 of the Act. Title II--Sections 202, 211, and 231--will be used for moving sources.
3. Protection of life: This objective is the ultimate purpose of the Act. It applies to those pollutants that are present in the ambient air and must be reduced to safe levels. Generally, these pollutants cannot be readily measured and concentrations must be reduced to a minimum (or possibly to zero if the air pollutant is

hazardous as defined in the Act) to achieve the goal of protecting life. Thus, Section 112 of the Act would be applicable to such pollutants.

B. Necessary Analysis for a Preferred Standards Path

Information needed to develop a standard will depend to a large degree on the recommended control option, i.e., which section(s) of the Act is (are) to be used in attaining the prescribed objective. A thorough evaluation which leads to such a recommendation is the heart of a PSP analysis. Therefore, most items which should be considered in this type analysis are included in the following subsections.

1. Summary of the existing problem - Without an exhaustive knowledge of the existing situation no effective analysis can be undertaken. Specific factors to be examined in summarizing current conditions are described briefly herein. Figure A-1 is provided as a guide in the logic of analyzing a candidate pollutant for determining what control strategy may be applicable.
 - a. Information on the pollutant's effects: A quantitative criteria document is not required to determine a pollutant's effects on human health and welfare. However, health effects must be quantified for a candidate pollutant prior to implementing Sections 109 and 112 of the Act. Both these sections of the Act provide general guidance to the



NOTE 1: A yes answer here simply means the pollutant is a candidate for regulatory action; however, it does not denote mandatory action.

NOTE 2: The dashed line indicates that significant pollution from stationary sources may remain even after utilizing ESFMS. Land use and transportation controls may need to be implemented in addition to Section 202.

*MOST LIKELY APPROPRIATE simply means that the indicated option may be the most logical without considering external factors. However, other ramifications may preclude its use. Also, more than one option (e.g., Sections 109 and 202; Sections 109 and 111; etc.) may be used in combination for any given pollutant in order to achieve the full intent of the Act. (Also see NOTES.) Section 111(d) is applicable to non-criteria pollutants.

** Acronyms explained in text.

Figure A-1. Preferred standards path: Guide for determination of regulatory action.

nature and severity of effects needed before action can be taken under either section. Some differentiation between chronic and acute effects is needed. The objective is to estimate the likelihood of occurrence and severity of adverse effects.

- b. Ambient levels: Without a good estimate of ambient concentrations of the candidate pollutant little appreciation for its health and/or welfare effects is attained. An evaluation of ambient levels should determine whether the pollutant is ubiquitous or is related only to specific point sources. If sources are numerous or diverse and cause adverse health effects at ambient levels then Section 109 may be applicable. If effects are caused by limited point sources Section 111 may be recommended. Of course, Title II must be used for moving sources.
- c. Sources of the pollutant: A precise emission inventory is not required; however, the following information is needed: (1) an estimate of emissions from various sources, along with the probability of resulting ambient levels; (2) the expected growth and/or decline of the number and type of source categories that emit the pollutant; and (3) the number of individual sources within a given category. This information determines whether observed effects are caused by localized or numerous and diverse

sources. Contributions from mobile sources must be included also.

- d. Technology assessment: An evaluation of the state-of-the-art in monitoring and control technology must be available. Monitoring technology includes ambient and source sampling instrumentation. Control technology includes the feasibility of applying existing methods to various process configurations within an industry. Furthermore, this section should evaluate the effectiveness of reducing emissions beyond those levels achievable with current control methods.
- e. Environmental impact: A PSP should consider the impact of the suggested option upon the environment. The completeness of this consideration may be dictated by the option under consideration. For example, if a NESHAP option is being studied, cost is precluded as an item for consideration; health effects are of primary importance. Generally, this section should contain such things as the accumulation of the pollutant in the environment, its effect on the fate of other pollutants, and other impacts.

- 2. Summary of existing regulatory efforts - In this subsection, the status of existing regulatory efforts should be examined in relation to how the selected PSP would impact these regulations. State, local, and Federal control programs should be considered in order that

pertinent actions for a new pollutant would not be contradictory, or cause some additional obstacle in enforcing existing regulations. These possibilities may be considered in the following two ways.

- a. Existing State, local, or Federal regulations may achieve partial control for a candidate pollutant. If so, the degree of control may be determined by summarizing the number and type regulations and by determining their degree of enforcement.
- b. Some control of the candidate pollutant may result from emission reductions of other pollutants, voluntary controls by the emitter, or Federal policy. Here, EPA must decide whether this method yields the desired control, or if the procedure results in the best control.

3. Summary of options available - The three options available for controlling air pollution from stationary sources are described in greater detail in this subsection. As previously stated, "no control" or "postponement of a decision to control" are also valid options that may result from a PSP analysis. One of the latter options may be chosen if based on existing evidence, an adverse effect cannot be shown to result from a candidate pollutant. The Administrator of EPA has such broad, discretionary powers to reach a decision under the Act that more attention is given below to the options.

- a. National Ambient Air Quality Standards: This approach may be used if the candidate pollutant has "an adverse effect on public health and welfare" and ambient levels result ". . . from numerous or diverse mobile or stationary sources" [Section 108(a)(1)].

A NAAQS includes a primary and secondary standard. Primary standards must be at levels which, "based on [air quality] criteria and allowing an adequate margin of safety, are requisite to protect the public health" [Section 109(b)(2)].

Since ambient levels are specified, NAAQSs are not directly applicable to sources. However, the attainment of an ambient standard may require source control. States decide how to control source emissions in order to comply with the ambient standards. Each State prepares an implementation plan (SIP), subject to approval by the Administrator [Section 110(a)(2)]. SIP regulations may utilize emission standards, land use policies, transportation control measures, or other approaches. The State has wide leeway for controlling new and existing sources except for moving sources, which are specifically exempted [Section 116]. If a State submits an unacceptable plan, EPA is required to promulgate regulations that will achieve the standard(s) [Section 110(c)]. Likewise, if a State does not enforce its regulations this responsibility reverts to the Federal government [Section 113(a)].

The milestones prescribed by the Act for NAAQS are more complex than for the other options and thus can result in longer delays before control is achieved. Significant milestones are listed below.¹

<u>MILESTONE</u>	<u>ACTION</u>
1. Start	Inclusion of candidate pollutant on Section 108 list.
2. Within 12 months	Issue air quality criteria and proposed standards (primary and secondary).
3. Within 3 more months	Promulgate standards.
4. Within 9 more months ⁽²⁾	Submit SIP's
5. Within 4 more months	Approval/disapproval of SIP. If not submitted or disapproved, prompt proposal of plan by EPA. ⁽³⁾
6. Within 2 more months	Promulgate EPA plan if SIP remains inadequate.
7. Within 3 more years ⁽⁴⁾	Attainment of primary ⁽⁵⁾ standard.

¹The Act delineated deadlines for initiating the regulatory process in each option (including the other two described below) for certain pollutants and sources (e.g., the six pollutants with existing air quality criteria documents, Group I NSPS, etc.). It also provided for subsequent inclusion of pollutants on relevant lists "from time to time." Once begun, however, the statutory timetable binds EPA to the described milestones.

²The Administrator may extend the deadline for submitting SIP's for secondary standards an additional 18 months.

³If a State fails to revise an existing SIP within 60 days, or a longer prescribed period, upon notification by the Administrator, EPA revisions are to be proposed promptly.

⁴If adequate technology is unavailable, an extension of up to two years may be granted by the Administrator for the attainment of a primary standard.

⁵Secondary standards must be attained within a reasonable time as specified within the SIP.

b. New Source Performance Standards: This option may be used if a category of sources "may contribute significantly to air pollution which causes or contributes to the endangerment of public health or welfare" [Section 111(b)(A)]. National standards established under Section 111 are applicable to (1) new stationary sources for specific categories and (2) modifications resulting in new emissions of air pollution. The Administrator can distinguish among classes, types, and sizes within categories. Section 111(d)(1) requires States to control existing sources covered by NSPS applicable to the specified category unless the pollutant is covered under NAAQS or NESHAP. If the pollutant is covered by Section 108(a), NAAQS, the States can prevent construction or modification of stationary sources which may interfere with attainment or maintenance of a NAAQS [Section 110(a)(4)]. With proper enforcement, SIP's control existing sources as necessary to attain and maintain the NAAQS. Moreover, if a pollutant has been designated as hazardous the standard will apply to all designated sources--existing, modified, and new.

The level of the standard "reflects the degree of emission limitation achievable through the application of the best system of emission reduction which (taking into

account the cost of achieving such reduction) the Administrator determines has been adequately demonstrated" [Section 111(a)(1)]. Thus, the degree of control (which likely will not be more stringent than the standard) is not related directly to health and/or welfare. States also may use best demonstrated control methods in setting standards for existing sources.

Enforcement of NSPS is a Federal responsibility initially [Section 113(a)(3)] but may be delegated to States if acceptable enforcement procedures are adopted [Section 111(c)(1)]. In addition, each state must adopt plans to regulate unmodified existing sources of the same category, provided the pollutant is not a criteria or hazardous pollutant. These plans are similar to procedures required to achieve NAAQS [Section 111(d)].

Milestones for NSPS do not allow for time extension as under NAAQS. Milestones as given in the Act follow.

<u>MILESTONE</u>	<u>ACTION</u>
1. Start	Include category of sources on Section 111 list.
2. Within 120 days	Issue proposed standards. ⁽⁶⁾
3. Within 90 more days	Promulgate standards. Effective upon promulgation.

⁶Presently no guidance is available to states in meeting specific deadlines for controlling emissions from unmodified existing sources for a given category. However, such guidelines have been proposed.

c. National Emission Standards for Hazardous Air Pollutants:

This approach may be used if the pollutant "may cause, or contribute to, an increase in mortality or an increase in serious irreversible, or incapacitating reversible, illness" [Section 112(a)(1)]. Standards under Section 112 are applicable to new and existing stationary sources, with the provision of a 90 day delay for existing sources [Section 112(c)(1)]. Authority to make distinctions or classifications, as in NSPS, is subject to question. These emission standards are established at a level which "provides an ample margin of safety to protect the public health from such hazardous air pollutant" [Section 112(b)(1)(B)]. Technology or cost of control can not be considered, except perhaps in deciding what margin of safety is ample. In order to assure an ample margin of safety the worst case expected in practice should be considered. This will involve reviewing the total number of sources, ambient concentrations, persistence of the pollutant, build-up in body tissue, etc. If an ample margin of safety, based on review of the above factors, required zero emissions a total ban could be accomplished under Section 112. (A similar result may be possible under Sections 108-110 if the ambient standards were

set low enough. Zero emissions can be achieved under Section 111 if technology to achieve 100 per cent control is adequately demonstrated.) Enforcement of NESHAP is a Federal responsibility [Section 113(a)(3)], but the Administrator may delegate authority to States under certain conditions [Section 112(d)]. However, this does not preclude the Federal enforcement prerogative. Again, provisions for time extensions are granted in the Act [Section 112(c)(1)(B); (c)(2)]. Milestones outlined for NESHAP result in the second quickest means for attainment of standard. However, with provision for extension and exemption for two-year periods, no definite time for attainment of standards can be estimated. Milestones designated in the Act are as follows:

MILESTONE	ACTION
1. Start	Include candidate pollutant on Section 112 list.
2. Within 180 days	Issue proposed standards.
3. Within 180 more days	Promulgate standards. Effective for new source upon promulgation, and 90 days later for existing sources.

C. Discussion of Various Options

Under this section the ramifications and reasoning for choosing a specific option should be given. Support for the recommended control option as decided by the PSP analysis must be substantiated by items discussed in this section. In other

words, a detailed discussion of factors outlined in "B" above must support the chosen control option. Insight into legal or other challenges should be given in detail. Except for those cases where prohibited by statute (e.g., Section 112) the economic impact for each option should be included.

D. Recommendations

The objective of this Section is to succinctly state the recommended control option and develop an accomplishment plan to implement the recommended approach.

APPENDIX B - LUNG CANCER MORTALITY IN SELECTED SMSAs

Table B-1 is a ranking of Standard Metropolitan Statistical Areas (SMSAs) by descending order of deaths from lung diseases. In comparing the principal cities of the SMSA (column 2) in this table with sampling sites of Table C-1 in Appendix C, one may see that the majority of those cities with relatively high lung cancer deaths have not been sampled for BaP in the NASN.

A definitive program to sample and analyze the ambient air for BaP in selected cities where high lung cancer incidence is established needs to be undertaken. This should be considered a first step in defining the relationship between the urban factor and carcinogens, specifically BaP or other POM.

Table B-1. WHITE MALE DEATHS AND DEATH RATES PER 100,000 POPULATION
PER YEAR BY SMSA* FOR MALIGNANT NEOPLASM OF TRACHEA, BRONCHUS, AND LUNG
1959-1961**

Rank	SMSA	Death rate	Total deaths	Rank	SMSA	Death rate	Total Deaths
1	Charleston, S. C.	65	75	27	Albany-Schen., N.Y.	48	492
2	Albany, Ga.	64	23	28	Beaumont, Texas	48	134
3	Galveston, Texas	63	79	29	Memphis, Tenn.	48	214
4	Lake Charles, La.	63	65	30	Omaha, Neb.-Ia.	48	280
5	New Orleans, La.	61	468	31	Richmond, Va.	48	184
6	Newport News, Va.	59	85	32	Buffalo, N.Y.	47	831
7	Montgomery, Ala.	58	61	33	Decatur, Ill.	47	75
8	Jersey City, N.J.	56	556	34	Savannah, Ga.	47	61
9	Shreveport, La.	56	121	35	Dallas, Texas	46	495
10	Baton Rouge, La.	55	74	36	Des Moines, Iowa	46	162
11	Norfolk, Va.	55	214	37	Philadelphia, Pa.-N.J.	46	2482
12	Jacksonville, Fla.	54	206	38	St. Louis, Mo.-Ill.	46	1178
13	Birmingham, Ala.	53	278	39	Atlanta, Ga.	45	377
14	Dubuque, Iowa	53	55	40	Cincinnati, Ohio	45	610
15	Baltimore, Md.	52	945	41	Erie, Pa.	45	155
16	Jackson, Mich.	52	92	42	Flint, Mich.	45	185
17	Tyler, Texas	52	49	43	New York, N.Y.	45	7025
18	Houston, Texas	51	536	44	Paterson, N.J.	45	752
19	Monroe, La.	51	44	45	Syracuse, N.Y.	45	354
20	Mobile, Ala.	50	111	46	Tampa, Fla.	45	682
21	Pensacola, Fla.	50	80	47	Topeka, Kansas	45	79
22	Charleston, W. Va.	49	141	48	Washington, D.C.	45	723
23	Honolulu, Hawaii	49	55	49	W. Palm Beach, Fla.	45	160
24	Miami, Fla.	49	683	50	Wheeling, W.Va.-Ohio	45	141
25	Portland, Me.	49	134	51	Wilmington, Del.	45	186
26	Toledo, Ohio	49	313				

*Standard Metropolitan Statistical Area.

**Reference 20.

APPENDIX C - NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR MEASUREMENTS FOR BENZO(a)PYRENE

Data given in this appendix are arithmetic averages of the four quarterly composites of the listed urban sampling site. The annual average is computed only if 3 or more quarterly composite values exceed the minimum detectable concentration of 0.2 nanograms/cubic meter (ng/m³).

Table C-1 contains annual averages for 121 selected NASN urban sampling sites. These sites were selected from the total NASN system based on valid data for each site for all three years. Each site had to meet minimum requirements of the NASN for each quarter in order to have a valid annual average for the given site. In order to be included in the summary, each sampling station must have a minimum of five scheduled samples collected and analyzed for any given quarter.

Table C-2 is a listing of 40 sites selected to update BaP concentrations in cities with and without coke ovens. Three sites were selected in National Parks so nonurban background readings would be available. A limited number (40) was chosen because of time and resource restrictions since routine analysis of NASN BaP was discontinued in 1970.

Table C-3 gives valid annual averages of BaP for the sites listed in Table C-2 for all years of record (1966-1972). Numbers in parenthesis are the number of valid sites included for the year.

Table C-1. NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR MEASUREMENTS
FOR BENZO(a)PYRENE^a, 121 SELECTED SITES, 1968-1970

Location, state and city	Benzo(a)pyrene [ng/m ³ (25°C)]		
	1968	1969	1970
Alabama			
Gadsden	2.37	1.76	2.50
Huntsville	2.74	1.82	1.56
Montgomery	2.93	2.04	1.31
Alaska			
Anchorage	1.68	1.28	0.77
Arizona			
Grand Canyon National Park	0.19	0.18	0.10
Maricopa County	0.47	0.27	0.28
Tucson	0.67	0.53	0.41
Arkansas			
Little Rock	0.89	1.05	0.65
Montgomery County	0.23	0.18	0.13
West Memphis	2.24	2.41	0.59
California			
Glendale	1.55	1.63	0.97
Humboldt County	0.31	0.49	0.11
Long Beach	2.09	2.27	1.00
Los Angeles	1.83	1.86	1.23
Oakland	1.62	1.61	0.95
Riverside	1.29	0.82	0.67
Sacramento	1.43	1.79	0.72
San Bernardino	1.00	0.90	0.83
San Diego	1.22	1.38	0.65
San Francisco	1.83	1.15	0.63
Colorado			
Denver	2.25	2.51	2.20
Connecticut			
Hartford	1.42	1.95	1.37
New Haven	1.38	2.10	1.21
Florida			
Jacksonville	2.94	2.30	1.35
Tampa	1.46	1.00	0.46
Georgia			
Atlanta	1.83	1.86	0.92
Hawaii			
Honolulu	0.55	0.57	0.19
Idaho			
Boise City	2.01	5.96	1.14
Butte County	0.17	0.09	0.07

Table C-1 (continued). NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR
MEASUREMENTS FOR BENZO(a)PYRENE^a, 121 SELECTED SITES, 1968-1970

Location, state and city	Benzo(a)pyrene [ng/m ³ (25°C)]		
	1968	1969	1970
Illinois			
Chicago	3.10	3.88	2.00
Springfield	1.06	1.27	0.85
Indiana			
East Chicago	4.90	6.75	5.26
Hammond	2.08	3.29	1.67
Indianapolis	4.07	5.18	2.32
Monroe County	0.54	0.25	0.16
Parke County	0.43	0.26	0.39
Iowa			
Des Moines	1.12	0.92	0.69
Kansas			
Topeka	0.69	0.44	0.31
Wichita	1.04	0.67	0.50
Kentucky			
Ashland	9.30	10.89	6.67
Covington	3.56	4.11	4.38
Louisiana			
New Orleans	1.55	1.52	1.14
Maine			
Acadia National Park	0.33	1.12	0.20
Maryland			
Baltimore	2.31	2.76	2.06
Massachusetts			
Worcester	1.65	1.48	1.63
Michigan			
Detroit	5.13	3.91	2.56
Flint	0.78	1.69	1.49
Grand Rapids	3.44	1.70	0.87
Trenton	1.44	1.58	0.84
Minnesota			
Duluth	2.73	2.08	1.09
Minneapolis	1.14	1.43	0.62
Moorhead	0.85	1.04	1.59
St. Paul	1.82	1.75	1.01
Nebraska			
Omaha	1.91	1.55	1.01
Thomas County	0.20	0.13	0.12

Table C-1 (continued). NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR
MEASUREMENTS FOR BENZO(a)PYRENE^a, 121 SELECTED SITES, 1968-1970

Location, state and city	Benzo(a)pyrene [ng/m ³ (25°C)]		
	1968	1969	1970
Nevada			
White Pine County	0.14	0.07	0.12
New Hampshire			
Concord	0.98	0.65	0.61
Coos County	0.20	0.11	0.14
New Jersey			
Camden	1.57	2.41	1.92
Glassboro	1.20	1.09	1.18
Jersey City	2.35	2.68	4.65
Newark	2.14	1.82	1.53
Paterson	1.95	1.24	1.20
Perth Amboy	1.17	1.20	1.01
Trenton	1.04	1.46	1.10
New Mexico			
Albuquerque	1.77	1.12	1.06
New York			
Jefferson County	0.19	0.25	0.24
North Carolina			
Cape Hatteras National Park	.22	.11	.21
Charlotte	5.56	4.85	1.85
Durham	7.96	3.38	3.38
North Dakota			
Bismarck	.86	.96	.44
Ohio			
Cincinnati	1.77	2.90	2.58
Cleveland	3.00	3.75	2.78
Columbus	2.21	2.73	1.57
Dayton	2.36	1.88	1.48
Toledo	1.80	1.49	1.38
Youngstown	5.64	9.86	7.12
Oklahoma			
Cherokee County	.21	.18	.22
Oklahoma City	.73	.71	.88
Tulsa	.77	.47	.79
Oregon			
Curry County	.14	.08	.09
Portland	4.13	2.60	2.31

Table C-1 (continued). NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR
MEASUREMENTS FOR BENZO(a)PYRENE^a, 121 SELECTED SITES, 1968-1970

Location, state and city	Benzo(a)pyrene[ng/m ³ (25°C)]		
	1968	1969	1970
Pennsylvania			
Allentown	1.17	1.92	2.40
Altoona	17.96	22.28	19.25
Bethlehem	2.05	2.00	2.71
Clarion County	.97	1.23	1.23
Harrisburg	1.32	1.45	1.53
Philadelphia	2.87	4.03	2.44
Pittsburgh	6.31	13.75	5.86
Reading	2.36	1.75	1.56
Scranton	6.08	7.65	2.88
Wilkes-Barre	1.58	1.54	1.30
York	1.91	2.00	1.21
Rhode Island			
East Providence	1.17	1.21	1.22
Providence	1.95	2.15	2.11
South Carolina			
Greenville	18.55	7.00	3.39
Tennessee			
Chattanooga	7.39	4.17	5.54
Memphis	1.34	.74	1.36
Nashville	5.96	2.80	3.61
Texas			
Matagorda County	.16	.12	.27
San Antonio	.85	.63	.95
Utah			
Ogden	.82	.67	2.49
Salt Lake City	.97	.65	1.44
Vermont			
Burlington	.71	.48	.74
Orange County	.31	.28	.16
Virginia			
Danville	2.47	1.79	2.69
Hampton	1.53	.88	1.08
Lynchburg	8.72	6.28	4.49
Norfolk	4.90	3.91	1.67
Portsmouth	10.17	3.39	4.94
Roanoke	7.65	5.30	6.20
Shenandoah National Park	.30	.31	.21

Table C-1 (continued). NATIONAL AIR SURVEILLANCE NETWORK AMBIENT AIR MEASUREMENTS FOR BENZO(a)PYRENE^a, 121 SELECTED SITES, 1968-1970

Location, state and city	Benzo(a)pyrene[ng/m ³ (25°C)]		
	1968	1969	1970
Washington Seattle	1.97	1.57	1.51
West Virginia Charleston	4.57	2.63	2.11
Wisconsin Kenosha	1.41	1.74	1.31
Madison	1.33	9.62	1.07
Milwaukee	4.65	4.03	2.46
Superior	3.30	1.58	1.50
Wyoming Casper	.90	.60	.44
Cheyenne	.60	.46	.43

^aAnnual average.

Table C-2. LISTING OF 40 NASN SITES SELECTED FOR 1971-72 BaP ANALYSIS

Coke oven cities		Noncoke oven cities
*1. Birmingham	16. Cleveland	1. Montgomery
2. Gadsden	17. Toledo	2. Jacksonville
3. Chicago	18. Youngstown	3. Honolulu
4. Gary	19. Bethlehem	4. Hammon
5. Indianapolis	*20. Erie	5. Baton Rouge
6. Terre Haute	21. Philadelphia	6. New Orleans
7. Ashland	22. Pittsburgh	*7. Acadia National Park
8. Baltimore	23. Chattanooga	8. New York
9. Dearborn	24. Houston	9. Newport News
10. Detroit	25. Spokane	10. Norfolk
11. Trenton	26. Milwaukee	*11. Shenandoah Nat'l. Park
*12. Duluth		12. Seattle
13. St. Paul		13. Charleston, W. Va.
14. St. Louis		*14. Grand Canyon
15. Buffalo		

*Not included in Figure 5. Birmingham and Erie had insufficient data; Duluth had known extraneous influences in 1971 and 1972.

Table C-3. ANNUAL BaP AVERAGES FOR SELECTED CITIES
1966-1972

Year	Coke	Non-coke
1966	4.74 (15)*	2.76 (7)
1967	5.34 (15)	2.29 (8)
1968	3.75 (18)	2.64 (8)
1969	4.41 (23)	2.14 (11)
1970	3.02 (21)	1.41 (11)
1971	2.18 (11)	1.22 (8)
1972	2.14 (19)	0.64 (11)

*Number of cities included in average (no. with full year's valid data).

APPENDIX D - DESCRIPTION OF BY-PRODUCT COKE PRODUCTION

Coke is the carbon residue of certain grades of bituminous coal after destructive distillation. It is used as a fuel, primarily in making pig iron which is an essential ingredient in steel. Destructive distillation (coking) is accomplished principally in by-product ovens and consists of driving certain volatile matter off coal, leaving a residue with a high percentage of carbon and relatively small amounts of impurities.

A brief description of the by-product coking process is presented here as extracted from reference 26 of the text. By-product ovens are usually constructed in groups called batteries. They consist of a block of many long, narrow firebrick ovens with heating chambers made of similar brick located between the ovens, so that a battery of ovens is a huge block of coking cells separated from an intricate system of combustion chambers (Figure D-1). The fuel used to heat coke ovens may be blast furnace gas, coke oven gas, or natural gas, each fuel requiring a different set of burner adjustments. None of the coal being coked is burned to provide heat for the coking operation.

The carbonization of the coal begins soon after crushed and sized coal is loaded into red-hot ovens. The gaseous products and condensates are conveyed continuously via the large collecting mains from the coke oven battery to an adjacent by-product plant. Usually each oven has a steam jet aspirator which aids in conveying the gaseous carbonization products into the collecting main.

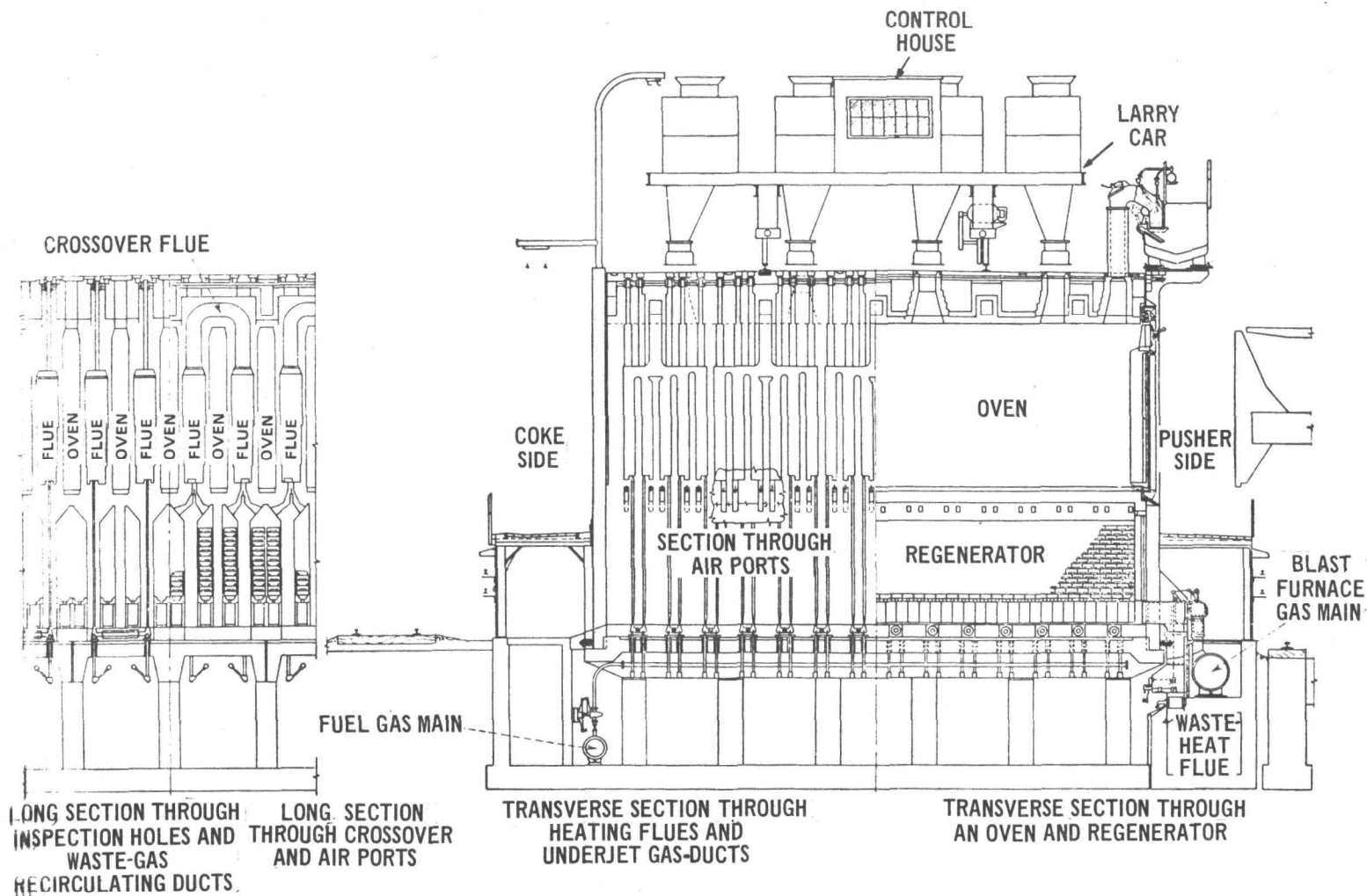


Figure D-1. Koppers-Becker underjet low-differential combination coke oven with waste-gas recirculation (Courtesy of Koppers Company, Inc.)²⁷

The charging holes on the top of the battery are closed almost immediately after charging to minimize escape of gas or dust into the atmosphere. Spacing of the charging holes, and sleeves on the charging car hoppers, have been used to minimize such emissions. Smoothness of operation and the physical characteristics of the coal being used aid in the reduction of these emissions. Most operators attempt, through door maintenance programs and operating procedures, to further reduce the escape of gas through the charging holes. Various methods of controlling particulate and gaseous emissions during charging have been investigated. More widespread application of these methods has occurred in Europe and Japan than in the U. S. However, recent attempts at application have been undertaken by several U. S. companies, with varying degrees of success.

In the by-product plant, tar, benzene, naphtha, and other commercial products are segregated and separately removed from the coke oven gas. The by-product recovery plant has processes similar to those in petroleum refineries and chemical plants. The cleaned gas, having a medium heating value, is customarily used as fuel in boilers and other furnaces, including those of the coke oven. Since sulfur has an undesirable influence in steelmaking, the steel industry has always endeavored to use materials with low sulfur content. Therefore, low sulfur coal is used for coke making. Coke oven gas usually contains some sulfur-bearing compounds derived from the sulfur in the coal.

After the coking period, the incandescent coke is pushed from the oven into a quenching car where burning in air takes place. This car is a large special-type railroad car with perforated sides, which is used

to carry the flaming coke to an area where a huge quantity of water is sprayed onto the coke to quench it, i.e., extinguish its incandescence. During this operation, which stops the burning of the coke, large quantities of steam are generated. Hot coke extinguishing is usually performed in a quenching tower. The quiescent coke is then conveyed to a cooling wharf where the coke is spread out so that the excess water may drain away. After cooling, the coke is sized in a plant similar to that for screening rock and ores. As with the charging phase of coke production, various attempts at controlling emissions from the pushing of finished coke have been attempted with various degrees of success.

APPENDIX E - COSTS OF CONTROL AND GROWTH PATTERNS FOR COKE OVENS

Most polycyclic organic matter (POM) is emitted when coal is charged to coke ovens, during the coking process, or when the finished coke is pushed out of the oven.

Charging Controls

The charging process is not generally controlled at present. Two possibilities for control are described. One is to equip the larry car (vehicle that dumps coal into the ovens) to prevent escape of fumes. This process is now in the demonstration stage but estimates indicate that the capital cost for retrofit of an existing 70 oven battery would approximate \$1,500,000. Several things could increase this cost: (1) the necessity to add additional vents to each oven; (2) adding holes in the roof for vents; and (3) strengthening the battery structure if necessary to bear the weight of the new larry car, which weighs 100 tons compared to 50 tons for a non-controlled car. Operating costs have not been compiled as yet. This type of information is expected to be available by the end of CY 1974.

Another control method already in use is pipeline charging. This method eliminates the larry car and the dumping cycle. The entire charging operation is confined within a pipe. For a new 70 oven battery costing about \$24,000,000, the control equipment can be added for \$8,000,000. However, since the coke is preheated in the process, claims are made that the production capacity of the battery is increased by 50%. To obtain the same capacity without pipeline charging would cost

\$36,000,000. Therefore, the control system results in a 10% capital saving at the greater production figure.

Retrofit costs will run \$12 to \$14 million for a 70 oven battery. However, if a market is assumed for the additional coke produced, installation of control equipment would cost about the same as adding ovens. Claims are also made that operating savings from the use of a higher percentage of cheaper, high volatile coal; the increase in yield of blast furnace coke; and the shorter cycles would more than compensate for the operating costs involved.

Coking Controls

Gases generated during the coking process are vented to a by-products plant and thus are not released to the air. However, considerable leakage often occurs around door seals. At present the only remedy is careful operation and adequate maintenance, including cleaning the seals at each cycle.

Oven Discharge Controls

Although most of the POM evolves during the coking cycle, it is possible that some may evolve during the discharge cycle. One company is testing an indexing hood connected to a scrubber to control particulates. The company is continuing its tests of this control method for charging and discharging. At this stage of testing, insufficient data are available to judge the adequacy of performance or to accurately estimate costs. Company representatives estimated total costs would approximate \$10,000,000 to retrofit their batteries that contain a total of about 220 slots.

Cost Summary

Subject to reservations stated above, the tables below detail retrofit control costs²⁵ for a 70 oven battery. These costs are estimates only.

Table E-1. CAPITAL COSTS
(in \$1,000)

Pipeline charging	
Material and field costs	\$11,858
Modification to ovens and installation of charging main	546
Coordination and start-up	388
Subtotal	\$12,792
Pushing	
Material and field costs	1,619
Overhead and profit	26
Coordination and start-up	35
Subtotal	\$ 1,680
Total	\$14,472

Table E-2. ANNUAL COSTS
(in \$1,000)

Pipeline charging	
Maintenance at 6% Inv.	\$ 768
Utilities	318
Labor	229
Taxes, Ins. at 2% Inv.	256
Interest (8%)	1,023
Depreciation (15 yrs)	853
Subtotal	\$3,447
Pushing	
Maintenance at 6% Inv.	101
Utilities	92
Labor	0
Taxes and Ins. at 2% Inv.	34
Interest (8%)	134
Depreciation (15 yrs)	112
Subtotal	\$ 473
Total	\$3,920

Assuming that a 70 oven coke battery produces 630,000 tons of coke annually, the average cost per year to achieve about 85-90% BaP control is approximately \$6.25 per ton of coke. This compares with the cost of coke at \$40.70 per ton in 1972 (latest published data),³³ or about 15% of the market value. This annualized cost of retrofit is significantly greater than that for a new coke battery installing pipeline charging, where the estimated cost is about \$0.10 per ton of coke.

Capital costs for retrofitting a battery with a modified larry car have been estimated at 1/5 to 1/10 those for pipeline charging.²¹ However, operating costs have been projected to be much greater, even to the extent

of accounting for the difference in capital costs. These operating costs are only estimates and are not based on operational data because the first phase of the demonstration project has just been completed. Additional time will be required before operating costs can be obtained. Therefore, no definitive conclusion can be stated for costs at this time. Nevertheless, if the above cost estimates are reasonable one can immediately see the large differences between retrofitting coke ovens and installing controls on a new battery.

Growth Patterns

Despite the historic growth of the steel industry, the consumption of coke per ton of steel poured continues to drop. In 1960, 1542 pounds of coke were used per ton of steel; by 1970 the amount of coke dropped to 1248 pounds; and by 1980 a further drop to 1100 pounds is projected. Thus, even though hot metal production is expected to increase at a normal rate, coke production will not change appreciably.

However, because of air pollution regulations and obsolescence many existing batteries will have to be replaced. The table below was taken from a by-product coke oven survey of Koppers Company.³⁰

Table E-3. SUMMARY OF PROJECTED NEW OVEN CONSTRUCTION
UNITED STATES

Estimated date for operation	Number of batteries	Number of ovens	Annual capacity total coke (1000 tons)	New coke capacity (1000 tons)
1974	2	139	1,881	457
1975	5	386	4,440	1,445
1976	7	455	5,260	810
1977	5	326	3,470	700
1978	3	196	2,370	270
1979	2	109	1,465	---
1980	5	465	6,230	---

APPENDIX F - EMISSION ESTIMATES FOR COKE OVENS

Emission data for BaP from coke ovens are limited. Estimates of emissions which have been made from available data vary extensively, e.g., from 0.06 to 166 tons per year. Presently, crude sample data from uncontrolled plants in the United States are available.¹ Some data have also been reported from the U.S.S.R and Czechoslovakia.⁴ Demonstration projects are in progress at several plants within the U.S. Data on BaP emissions from uncontrolled and controlled sources are being gathered by contractors; these data should be available soon. Until these data are available, the following calculations, based on crude data, are presented to demonstrate the method of arriving at the estimate contained in the report. Total tons of by-product coke produced in the U.S. are used in the calculations because total nationwide emissions are of interest. However, beehive oven production is excluded because in 1972 it accounted for approximately only 1% of the U.S. total. All units are in U.S. net tons.

A. Estimates Based on U.S. Data:

1. Given:

a. Ratio of BaP to Total Particulates

$$\approx 1.734 \times 10^{-3}$$

(Reference 1, Smith)

b. Through-put loss of particulates in the coking

$$\text{operation} \approx 1.1 \times 10^{-3} \text{ tons of particulate}$$

per ton of coal charged (minus unloading and
quenching)

(Reference 2, AP-42)

c. Quantity of coal consumed per ton of coke

produced ≈ 1.45

(Reference 3, Bureau of Mines)

d. Tons of coke produced in 1972 $\approx 59.9 \times 10^6$

Tons produced at furnace plants $\approx 54.2 \times 10^6$

(Reference 3, Bureau of Mines)

2. Find:

Estimated BaP emissions from Coke ovens

$$\left(1.734 \times 10^{-3} \frac{\text{ton BaP}}{\text{ton particulate}}\right) \left(1.1 \times 10^{-3} \frac{\text{ton particulate}}{\text{ton of coal}}\right) \\ \left(1.45 \frac{\text{tons of coal}}{\text{tons of coke}}\right) \left(59.9 \times 10^6 \frac{6 \text{ tons coke}}{\text{year}}\right)$$

$\approx 166 \text{ tons of BaP/yr.}$

B. Estimate Based on Soviet Data:

1. Given:

Ratio of BaP/Total Particulates $\approx 1.05 \times 10^{-3}$

(This value is the average of the maximum ratios
of two Soviet coal-coke plants.)

(Reference 4, Mašek, Table 2)

2. Find:

Estimated BaP emissions for U.S. coke ovens
assuming B.1 is the accurate ratio.

$$\left(\frac{1.05 \times 10^{-3}}{1.734 \times 10^{-3}} \right) \left(166 \frac{\text{tons of BaP}}{\text{year}} \right) = 101 \text{ tons of BaP/yr.}$$

C. Estimate Based on Czechoslovak Data:

1. Given:

Ratio of BaP/Total Particulates $\approx 6 \times 10^{-7}$

(This estimate is the average value of BaP from
dust on the upper floor of the battery.)

(Reference 4, Table 5)

2. Find:

Estimated BaP emissions from U.S. coke
plants assuming C.1 is the accurate ratio.

$$\left(\frac{6 \times 10^{-7}}{1.734 \times 10^{-3}} \right) \left(166 \frac{\text{tons of BaP}}{\text{year}} \right) = 0.06 \text{ ton of BaP/yr.}$$

REFERENCES FOR APPENDIX F

1. Smith, William M., "Evaluation of Coke Oven Emissions," Presented to the 78th General Meeting, AISI, New York, N.Y., May 28, 1970. (Also presented at the 63rd Annual Meeting of APCA, St. Louis, Mo., June 14-18, 1970.)
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4. Mašek, Václav, "The Composition of Dusts from Work Sites of Coke Ovens," Staub - Reinhaltung Luft, Vol. 30, No. 5, [English edition] pp. 34-37, May 1970.