

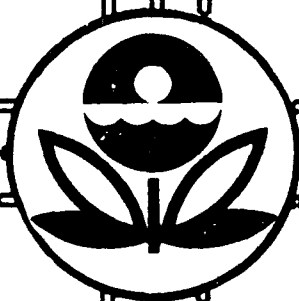
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REGULATORY AND TECHNICAL CONTROL  
STRATEGIES FOR FINE PARTICLES



U.S. ENVIRONMENTAL PROTECTION AGENCY

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**Regulatory and Technical Control Strategies for Fine Particles**

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REGULATORY AND TECHNICAL CONTROL STRATEGIES FOR FINE PARTICLES, J. Padgett and J.D. Bachmann, U.S. Environmental Protection Agency, Research Triangle Park, N.C.

This paper discusses the implications of available information for the development of technical and regulatory control strategies for fine particulate matter. Although it has been widely assumed to be a desirable goal, results of research activities in recent years have led to questions regarding the effectiveness and desirability of an undifferentiated fine particulate standard. Atmospheric particulate mass appears to be distributed into two fractions, which have distinct origins and properties. The fine particle mode is dominated by the products of gas to particle conversions, mostly sulfates, nitrates, and organics. Control of these substances means additional SO<sub>2</sub>, NO, and HC control. Some directly emitted particulates in this fraction have low concentrations, but high toxicity. The composition of fine particles varies markedly in different regions of the country. A general fine particulate standard, unless set at an unrealistically low level, would not ensure against effects from specific chemical compounds and would not obviate the need for separate regulatory programs for the more toxic particles. Control of specific categories of fine particles, with continued reliance on the current TSP, may be a more effective regulatory approach. EPA's current research programs are weighted towards specific pollutant categories, but regulations are several years away. In the meantime, existing regulatory programs can limit increases in emissions of fine particles and their precursors.

## REGULATORY AND TECHNICAL CONTROL STRATEGIES FOR FINE PARTICLES

### Introduction

The need for control of fine particulate matter (roughly defined as less than  $2\text{ }\mu\text{m}$ ) has been a topic of continued concern to EPA. The likelihood that these small particles are responsible for most of the adverse effects associated with total suspended particulate matter (TSP) was recognized at the time current particulate standards were established.<sup>(1)</sup> However, insufficient information existed to relate adverse effects to specific fine particulate levels. In 1972 the Administrator of EPA identified the establishment of National Ambient Air Quality Standards for fine particulate matter as a national priority objective in his guideline policy statement for the development of 1973-1978 program plans. The EPA Office of Research and Development (ORD) committed a substantial portion of its resources to studying fine particles and their control. An air quality standard for fine particles still cannot be set with the limited information available at this time. Furthermore, results of EPA and other research activities in recent years have led to questions regarding the effectiveness and desirability of an undifferentiated fine particulate standard.

This paper discusses the implications of available information for fine particulate control strategies. Based on current information, it appears that even with a fine particulate standard it would still be necessary to regulate certain classes of toxic fine particulate compounds such as lead, sulfates, and nitrates. Quite possibly the current TSP standard augmented by regulatory programs for specific toxic particulates will be adequate and preclude the need for a new general fine particulate standard with attendant regulatory disruptions.

### Discussion

#### Current Information

Although available information is still insufficient to initiate a regulatory program, the results of fine particulate research in recent years has led to important advances in our understanding of the origin and impacts of particles in the atmosphere. Figure 1 summarizes many of the important characteristics of atmospheric particulates typically observed in a variety of recent studies which determined particle size distributions in a number of areas.<sup>(2,3)</sup> The plot is constructed so that the area under any section of a curve is proportional to the concentration in that size range. It is clear that very fine particles make up most of the total suspended particulate in terms of numbers of particles and surface area. The mass (inferred from volume) shows a bimodal distribution.

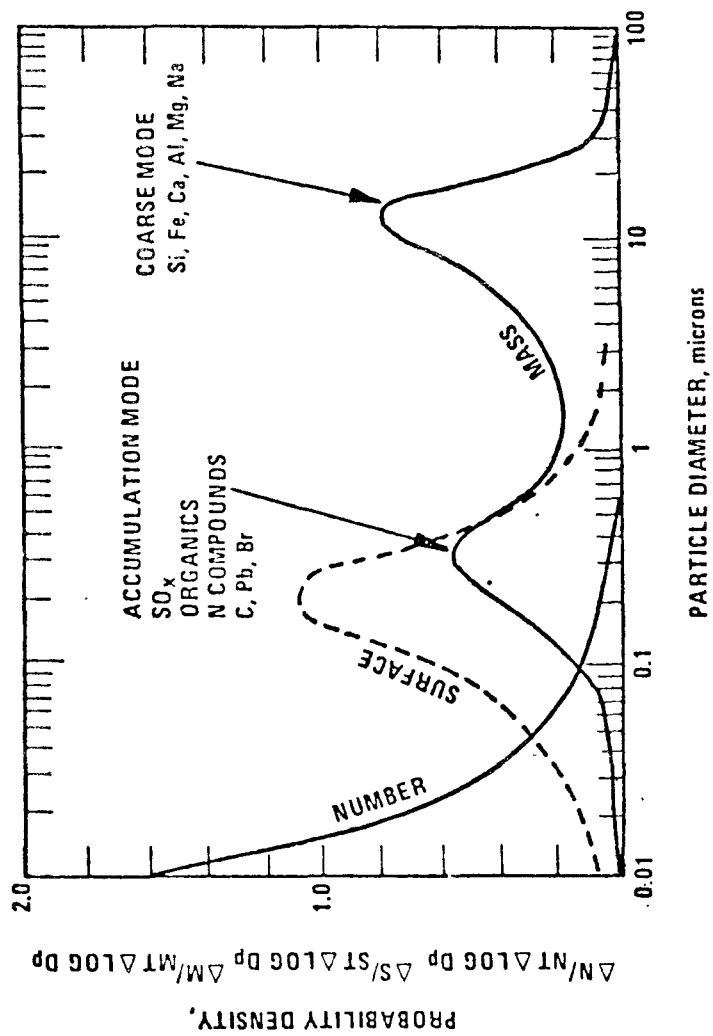


Figure 1. Normalized particle size distributions showing typical grouping of major chemical components.

The distinct minimum at about  $2\text{ }\mu\text{m}$  between the two modes provides a convenient size classification for atmospheric particles. The size range between  $0.1$  and  $1.0\text{ }\mu\text{m}$  is called the "accumulation" mode and typically makes up about one third of the total suspended particulate mass. Fine particulate mass "accumulates" in the  $0.1$  to  $1\text{ }\mu\text{m}$  range by coagulation of smaller particles or by condensation of gases on existing particles. It is thought that most of the mass in the accumulation mode is formed by physical and chemical processes which convert gases into particles, including trace element fumes from high temperature sources and the transformation products of sulfur oxides, nitrogen oxides, and organic compounds.<sup>(3)</sup> The large particle fraction of the bimodal distribution is termed the "coarse" mode. Coarse mode particles are generally formed by mechanical processes such as grinding or rubbing, for example industrial processes, soil, street dust, and rubber tire wear.<sup>(3)</sup> Chemical composition in this range is dominated by compounds of soil and mineral derived elements such as silicon, iron, and aluminum.<sup>(4,5)</sup> There appears to be very little exchange of mass between the fine and coarse particle ranges in the atmosphere because the far greater number and surface area associated with the accumulation mode dominate the condensation and coagulation growth processes.<sup>(3)</sup> Thus, the fine and coarse particle modes generally have distinctly different origins and chemical compositions.

Fine particles of varying chemical species share a number of important properties as a result of their physical size.<sup>(1)</sup> Fine particles have very long life times in the atmosphere, and can therefore be transported long distances before removal by dry deposition or precipitation.<sup>(6)</sup> Particles in the  $0.1$  to  $1\text{ }\mu\text{m}$  size range also scatter more light per unit mass than larger particles and thus control visibility reduction.<sup>(1)</sup> Significant increases in fine particle loading could have climatological consequences.<sup>(7)</sup> The greater surface area associated with fine particles provides sites to which more toxic chemical compounds can be attached or formed. Most importantly, these particles, because of their aerodynamic size, penetrate deeply into the respiratory system and impose a probable danger to public health by their own intrinsic toxicity, or by acting as a transport mechanism for more toxic substances.<sup>(1)</sup> The effective size is apparently important even for particles generally considered to be in the respirable range. Toxicological studies have found that sulfuric acid aerosols of  $0.7\text{ }\mu\text{m}$  size produced a four fold greater response than  $2.5\text{ }\mu\text{m}$  particles of the same compound.<sup>(8)</sup>

The chemical properties of small particles have been found to be at least as important as their physical size in defining the fine particulate problem. Considering the differing origins and compositions of fine and coarse particles, illustrated in Figure 1 and discussed above, chemical characteristics in some respects determine the ultimate particle size distribution. In addition, medical authorities believe that the chemical nature can influence the respiratory penetration of particles, their solubility and retention in the lungs, and their resultant biological toxicity.<sup>(1,9)</sup> Chemical composition also determines the extent to which atmospheric fine particles can cause material damage, crop losses, and ecological disruption during removal by precipitation or deposition.<sup>(10)</sup>

TABLE I. Results of Ambient Monitoring Studies<sup>(14),(15)</sup>

City	Percent Fine Particulate Mass Secondarily Formed <sup>a</sup>	Fine Particulate Mass as Percent of TSP
Pasadena, California	75	60
Pomona, California	80	40
New York City, N. Y. (Welfare Island)	60	50
Columbus, Ohio	70	NA
Secaucus, N. J. <sup>b</sup>	80	60
Denver, Colorado	60 - 75	15 - 25

<sup>a</sup>Sulfates, nitrates, ammonia, and organic compounds (organic material assumed 1/2 primary, 1/2 secondary).

<sup>b</sup>Moderate pollution conditions.

TABLE II. Regional Variation Major Chemical Classes Typically Occurring as Fine Particulate (Annual Arithmetic Average Based on 1966-1968 NASN Data)<sup>(18)</sup>

Region	SO <sub>4</sub> <sup>=a</sup> ( $\mu\text{g}/\text{m}^3$ )	BSO <sup>b</sup> ( $\mu\text{g}/\text{m}^3$ )	NO <sub>3</sub> <sup>-a</sup> ( $\mu\text{g}/\text{m}^3$ )	Pb ( $\mu\text{g}/\text{m}^3$ )	Total ( $\mu\text{g}/\text{m}^3$ )
East <sup>c</sup>	18.9	7.8	2.1	1.2	30
Mideast <sup>d</sup>	14.6	7.3	2.9	1.3	26
South	10.0	7.8	2.5	1.0	21
Midwest <sup>e</sup>	5.9	5.3	1.8	1.0	14
Mountain	3.4	4.7	1.3	1.3	10
Southwest	4.4	5.5	2.1	1.0	13
West Coast	9.4	8.9	3.8	1.9	24

<sup>a</sup>Measured NH<sub>4</sub><sup>+</sup> partitioned to SO<sub>4</sub><sup>=</sup>, NO<sub>3</sub><sup>-</sup>

<sup>b</sup>Benzene soluble organics, about 1/2 to 2/3 of total organics

<sup>c</sup>Providence, R. I. to Washington, D. C.

<sup>d</sup>Ohio, Michigan, Indiana, Illinois

<sup>e</sup>Minnesota, Iowa, Nebraska, Missouri

Recent epidemiological and toxicological studies appear to support the relative importance of certain chemical categories of fine particulate matter. Preliminary data from the Community Health and Environmental Surveillance System (CHESS) studies indicates that the respirable fraction of particulate matter (generally equated to fine particulate matter) has not shown as pronounced an association with adverse health effects, as have some specific fine particulates such as nitrates and sulfates. (11,12)

Chemical composition of particles has usually been assumed to be of lesser importance than size in causing visibility deterioration. However, several recent studies have indicated that chemical properties of fine particulate matter are also important in determining visibility reduction. Studies in the Los Angeles basin found that sulfates accounted for two to three times the visibility reduction for a given mass than other fine particulate components. (13) Hygroscopic and/or deliquescent fine particles can absorb water and increase in size at higher humidities, resulting in an aerosol visibility reduction much greater than might be expected on a mass basis. (6) At lower humidities, chemical composition can influence the ability of a particle to reduce visibility by affecting its light absorptive properties.

Studies which examined both size and chemical composition of atmospheric particulates are too limited in number and time to provide adequate characterization and source assessment for most urban areas. However, the results of short-term studies, some of which are summarized in Table I, provide some insight into the principal sources of fine particulates. Some 60 to 80 percent of fine particulate mass can be accounted for by the aforementioned gas to particle transformations which occur in the accumulation mode. (15) Particles formed in this manner have often been termed secondary particles; material which leaves a source in particulate form is termed primary or directly emitted particulate. As indicated in Figure 1, secondary fine particles have been found to consist almost entirely of sulfate, nitrate, ammonium, and organic compounds. These groups were found to comprise 50 percent of the total suspended particulate levels in the Los Angeles Basin. (16) The principal directly emitted fine particulate components in most urban areas are lead-related compounds from automotive sources, which can account for as much as 10% of the TSP in some cities. (17) The sum of all other directly emitted metals typically accounts for only about five percent of the fine particulate mass. (18)

Using National Air Surveillance Network (NASN) hi-vol measurements of the major chemical components of fine particulates as a surrogate, it was found that the chemical composition and concentration of fine particulates varies considerably from one geographic area to another (Table II). (18) It can be seen that the water soluble sulfates are the principal fine particulate component in the eastern United States, with organics and nitrates relatively more important in western areas. It is important to note that benzene soluble organic (BSO) measurements probably underestimate total organic concentration since benzene is not a good solvent for some of the more polar oxygenated organic compounds which may be present. (14) In addition, recent data suggests that particulate nitrate results may be unduly high due to collection of gaseous nitric acid vapor. (19) Nevertheless, the general observation that marked geographical variations exist in the chemical composition of fine particles is still valid. Since



health and welfare effects associated with these chemical components vary, it is likely that different areas of the country may have similar fine particulate concentrations, but with decidedly dissimilar impacts.

### Control Strategy Implications

Available information on health and welfare effects does not permit the establishment of adequate dose/response functions for fine particles. Given the variability in fine particulate composition, it may not be possible to develop generally applicable dose/response relationships for undifferentiated fine particles. However, preliminary research indicates that some adverse effects may be associated with certain fine particulate classes even when current TSP standards are met.<sup>(12)</sup> It is probable that control programs aimed at the specific classes of fine particles will be needed. Although developments of such programs will require much additional research, some assessment of their direction is possible at this time.

Current limited information summarized in the previous section suggests essentially three approaches to controlling fine particulates, either as a single group or as chemical subgroups: (1) Control of emissions of gaseous precursors to fine particulates which account for the majority of fine particulate mass; (2) Control of the mechanisms which promote transformation of gases to particles; (3) Control of direct emissions of fine particles.

Since important pollutants in the fine particulate category are emitted directly (e.g., lead) and formed in the atmosphere (e.g., sulfates), it is likely that effective control strategies will require integration of all three approaches. In view of the high proportions of gas derived aerosols, conventional particulate control plans stressing direct emissions control alone do not appear likely to be sufficient to provide adequate protection against fine particulate effects. Since sulfates, nitrates, and organics appear to be the major fine particulate components, the control of fine particulates may require more stringent control of sulfur dioxide (SO<sub>2</sub>), nitric oxide (NO), and hydrocarbons (HC) emissions. It is of note that only certain chemical fractions of total HC emissions are thought to be of major significance in forming organic aerosols.<sup>(20)</sup>

The above substances are converted to particles by a number of incompletely understood mechanisms which involve atmospheric variables such as levels of other pollutants, humidity, sunlight intensity, and temperature. Control of certain pollutants may slow these reactions and reduce the amount of gases which form particulates. For example, aerosol formation appears to be accelerated by photochemical activity.<sup>(16)</sup> Hence, reductions in photochemical oxidant levels would indirectly influence fine particulate levels by reducing the amount of gases which are transformed into aerosols. Since reactive HC emission reduction is the principal method of oxidant control, organic aerosols would be both indirectly and directly affected by oxidant reduction programs.<sup>(13)</sup> Although some benefit can be expected from controlling pollutants which influence aerosol formation mechanisms, many other important variables, such as humidity, cannot be controlled. Thus, control of mechanisms which form particles in the atmosphere is likely to be a supplement to control of direct precursor gases, rather than a complete strategy. Additional understanding of aerosol formation mechanisms is needed.

Although it cannot be considered as the likely principal control approach, control of directly emitted fine particulates is of continuing importance. As previously discussed, direct emissions appear to account for 20 to 40 percent of the fine particulate mass. These emissions could dominate small particle loading in the vicinity of strong sources. Many directly emitted compounds are both toxic and tend to volatilize in high temperature processes, recondensing as fine particles before or during emission.<sup>(21)</sup> Lead, arsenic, selenium, and benzo(a)pyrene are examples of such toxic materials, and are typically found in the fine particle mode. Although these substances are not generally found in high concentrations, levels around certain stationary sources may be of concern. In addition, toxicologically innocuous directly emitted particles such as elemental carbon or iron oxide may provide reaction sites for and/or catalyze aerosol formation processes.<sup>(22)</sup> Improved characterization of both direct particulate emissions as well as the ambient aerosols is needed throughout the nation.

Besides accounting for complex gas/particle transformations and direct emission characteristics, control strategies may have to consider the impact of long range transport of significant quantities of fine particulates and their precursors. A growing body of data suggests that this phenomenon influences ambient concentrations of oxidants<sup>(23)</sup> and sulfates.<sup>(24)</sup> In the case of sulfates, it has been noted that geographical and temporal correlations exist between SO<sub>2</sub> emissions increases in the northeast quadrant of the U. S. and observed or suspected increases in regionally high urban and non-urban sulfate levels, acidic precipitation, and visibility deterioration.<sup>(25)</sup> Future fine particulate control programs may have to be based on broader considerations than the effects of local emissions on local air quality. Methods must be developed to permit evaluation of region wide source/receptor relationships as mediated by fine particulate transformation/transport/removal processes.

#### Fine Particulate Control Strategies

Original plans for a fine particulate air quality standard were based on the belief that this approach could protect against adverse effects related to particle size. This approach was also recognized as a possible improvement over the ability of existing TSP standards to safeguard public health and welfare on a national basis. Ambient concentrations of TSP in some regions can be greatly influenced by larger particles, such as wind-blown road dust, which may not be of major interest in protecting public health and visibility.<sup>(26)</sup> Hence, TSP may be a less adequate indicator of health risk from particulates than are fine particles, although few epidemiological studies have attempted to relate ambient levels of general fine particles to health effects.

In recent years, however, it has become more apparent that a general fine particulate standard may not go far enough toward improving current standards. As noted earlier, the fine particulate fraction is made up of a variety of compounds with varying toxicity. Two of the major components, sulfates and nitrates, have shown an association with health effects. Some fine particulates, such as arsenic and polycyclic organic mater, may be toxic or carcinogenic even in relatively small amounts.

Given that the chemical composition varies from region to region, attainment of a given level of fine particles would not guarantee that all fine particulate components of interest would be reduced to safe levels, unless a national fine particulate standard was set at an unrealistically low level or supplemented by standards for specific toxic components. It would appear that preceeding directly to control of fine particulate chemical components while maintaining current TSP standards may well represent the most effective means to adequately safeguard public health and welfare.

Although no decisions can be made concerning the ultimate form of future control programs, work is proceeding towards regulation of specific fine particulate compounds. EPA has estimated that it will take several years to develop information needed to regulate sulfates, a major fine particulate component, in many areas. Less is known about nitrate and organic aerosols.

Since it will take years to initiate regulations for the major fine particulate components, it is important to provide some assessment of the potential impacts of current air pollution control programs in the light of current knowledge on fine particles. Based on available information, current regulatory programs for SO<sub>2</sub>, NO<sub>2</sub>, HC, TSP, and photochemical oxidants will have an impact on fine particulate levels. These regulatory programs include ambient air quality standards, state implementation plan emission limits, new source performance standards for selected stationary source categories, and emissions standards for mobile sources. Table III presents Nationwide Emissions Projections which include the effect of these programs and anticipated growth.

Table III - Projected Natiowide Emissions<sup>a</sup> of Fine Particulate Related Pollutants under Current Regulatory Plans (10<sup>6</sup> tons/yr)

	<u>Particulates</u>	<u>SO<sub>2</sub></u>	<u>NO<sub>x</sub></u>	<u>Hydrocarbons</u>
1972	23.2	32.6	22.2	33.7
1975	18.1	33.2	24.5	30.7
1980	18.8	34.0	26.2	31.0
1985	19.7	34.8	26.4	33.8
1990	20.9	38.8	28.6	38.5

<sup>a</sup>Projections were obtained by normalizing published estimates<sup>(27,28,29,23)</sup> to a common nationwide emissions inventory<sup>(30)</sup> for base year 1972. Adjustments on some source categories were made to reflect recent information.

Although a linear relationship between reductions in emissions and ambient fine particulate concentrations is unlikely, these projections do provide some insight into future trends in ambient levels of fine particulates. With the possible exception of nitrogen oxides, it is apparent that major emission increases in fine particulate related pollutants are not anticipated in the next 10 years. The pattern of increasing emissions of these pollutants which held through the 1960's appears to have been slowed or stopped. Although trends data are limited, it is likely that associated apparent increases in non-urban sulfate levels,<sup>(25)</sup> acid precipitation,<sup>(31)</sup> and regional visibility degradation,<sup>(10)</sup> which also occurred during the 1960's, have been limited as well. Current regulatory programs should permit adequate time for research efforts to document effects levels and source/receptor relationships, and to develop improved control technologies for fine particles in the next few years without marked increases in ambient concentrations of fine particulates.

### Conclusions

A National Ambient Air Quality Standard for fine particulate matter as a group can no longer be regarded as the certain choice for a long range strategy to control fine particulates. Recent evidence suggests an approach which places greater emphasis on the control of selected categories of fine particulate matter, especially sulfates and nitrates, with continued reliance on the current TSP standard for overall control of particulates. This strategy provides continuity in existing particulate control programs, and has the potential for more effectively achieving necessary health and welfare benefits by placing priority on the control of the more harmful components of fine particulate matter. A general fine particulate standard would not necessarily ensure against effects from specific chemical compounds and would not obviate the need for separate regulatory programs for such toxic particulates as lead and other trace elements, acid aerosols such as sulfates and nitrates, and carcinogens such as certain polycyclic organics.

However, control of fine particulates may become increasingly important for achieving the TSP standard in many metropolitan areas. Furthermore, much of the general fine particulate research is useful in developing criteria for regulation of specific toxic particulates. Therefore, work necessary to develop a general fine particulate standard should continue, but it should not be given as high a priority as research directed specifically to developing control programs for known toxic particulates. In the meantime, implementation of existing air pollution regulations can limit increases in emissions of fine particles and their precursors.

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