Particulate matter sources, emissions, and control options—USA

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ABSTRACT

A new national standard for particulate matter (PM) in the ambient air is being developed in the United States (U.S.). Whereas the current standard applies to PM less than 10 micrometers (m) in aerodynamic diameter (PM_{10}), the standard being developed would also address particles smaller than 2.5 m ($PM_{2.5}$). To meet either the current standard or a new one, sources of both primary and secondary particles need to be inventoried and controlled. (Control of 'primary' particles, which are emitted directly into the air, involves emissions prevention or collection at the source. Control of 'secondary' particles, which are formed in the atmosphere, requires reducing emissions of precursor constituents such as sulfur oxides, nitrogen oxides, and ammonia.) This paper summarizes the current knowledge on sources of primary particles in the U.S., and options for control of their emissions. Research needs and plans are briefly addressed.

1. BACKGROUND

Because epidemiological associations have been found at particle concentrations below the existing ambient air quality standard, a new national standard is being developed in the U.S. (USEPA, 1996d). Whereas the current standard applies to PM less than 10 m in aerodynamic diameter (PM_{10}), the standard being developed would also address particles smaller than 2.5 m ($PM_{2.5}$). The current PM_{10} standard considers only size and mass concentration; chemical composition and toxicity of the particles are not addressed. The proposed $PM_{2.5}$ standard is also based solely on size and mass concentration. Promulgation of the new standard is scheduled for June 1997. To meet either the current standard or a new one, sources of both primary and secondary particles need to be controlled. Control of 'primary' particles, which are emitted directly into the air, involves emissions prevention or collection at the source. Control of 'secondary' particles, which are formed in the atmosphere, requires reducing emissions of precursor constituents such as condensible organic compounds, sulfur oxides, nitrogen oxides, and ammonia.

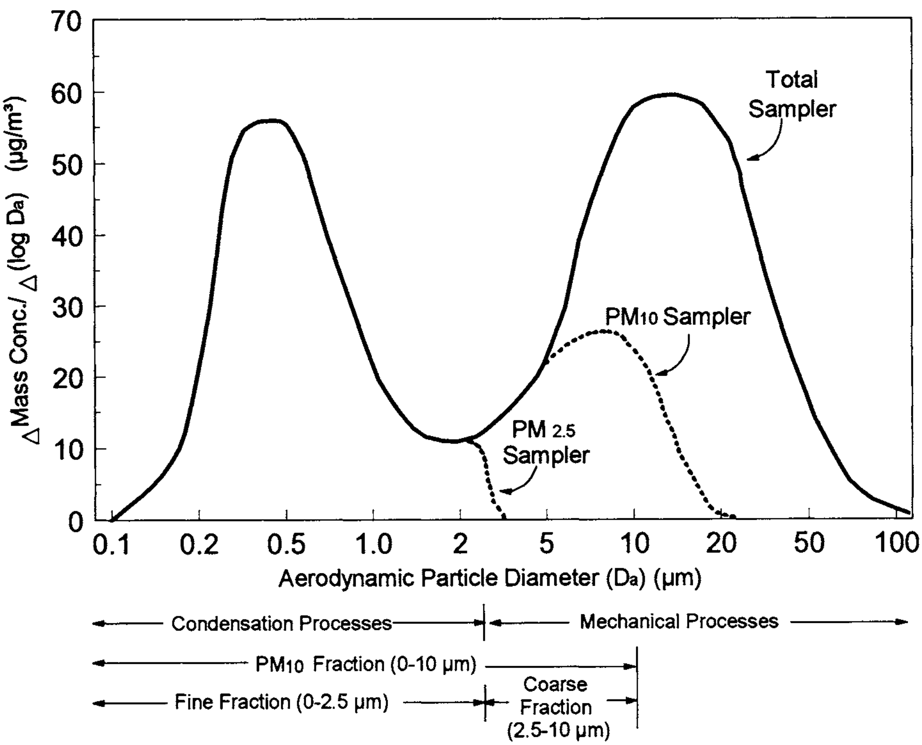

Figure 1. PM_{10} and $PM_{2.5}$ related to a typical size distribution for ambient particles. (Adapted from USEPA, 1996b)

Figure 1 shows a typical size distribution of particles in the ambient air, and how PM_{10} and $PM_{2.5}$ relate to the total distribution. This figure indicates that particles are generally distributed bimodally by size in the atmosphere, with the minimum of the distribution between 1 and 3 μm aerodynamic particle diameter. Particles in the fine mode are created as primary particles either by combustion or other high-temperature processes, or as secondary particles in the atmosphere. Most of the mass of particles in the coarse mode comes from materials that have been ground down by mechanical processes, although fine-mode particles can also become attached (USEPA, 1996b).

Ambient PM is a complex mixture of sizes and types of particles that originate from many sources. The size, chemical composition, and source of particles may all play a role in human exposures, and health effects resulting from those exposures. Little detailed information is available on the specific chemical makeup of ambient particles, especially the metal speciation and semivolatile organic components of fine particles. Chemical characterization of PM from a broad range of locations with a variety of source types is needed to better determine the composition, range of transport, and variability of airborne particles.

2. SOURCES OF FINE PM

The types of sources that contribute to the two modes in the distribution shown in Figure 1 are generally known. Particles in the finer mode include primary particles from high-temperature metallurgical and combustion processes, secondary particles from atmospheric reactions, and an unknown (but theoretically small) amount of fine particles that have been deposited and resuspended by wind or human activities. Particles in the coarser mode include coarse windblown and road dust, pollens and spores, and some industrial particles. PM_{10} samples are generally dominated, on a mass basis, by coarse-mode particles. $PM_{2.5}$ samples can also have substantial amounts, on a mass basis, of coarse-mode particles from the left-hand tail of the coarse-mode distribution; this may be especially likely in areas with significant sources of resuspended dust.

Since the epidemiological studies of PM health effects are based on geographically dispersed (but mostly urban) locations with numerous and varied emission sources, the major sources of the particles are difficult to ascertain. Researchers most familiar with these studies have speculated that two types of sources may be particularly important: primary emissions from combustion sources, and secondary particles formed in the atmosphere. Further hypotheses have pointed toward acidic particles, particles containing sulfates, and particles containing transition metals; all such particles would implicate combustion sources. See Vedal, 1997 and Wilson and Spengler, 1996 for recent reviews.

There is a lot of uncertainty with respect to 'fugitive' particles. Their emission rates are poorly quantified, but potentially large compared to emission rates of combustion and other industrial sources. Table 1 summarizes the U.S. national inventory for particle emissions from general industrial, combustion, and fugitive sources. Note that approximately 70% of the total primary $PM_{2.5}$ is from fugitive sources. This is presumably because the national total PM_{10} emissions are so large, and the mass in the left-hand tail of the coarse mode (see Figure 1) is large compared to the mass of particles being created in the fine mode. Even though the estimates for $PM_{2.5}$ fugitive emissions are being re- evaluated and may be revised downward somewhat, they will still constitute a large percentage of the national total.

Emissions data summarized in Table 1 illustrate the large potential contribution of sulfur oxides [SO_x – principally sulfur dioxide (SO_2)] and nitrogen oxides [NO_x – principally nitrogen dioxide (NO_2)] gaseous precursors to ambient PM concentrations. If their combined 40 million tons per year were completely converted to sulfate and nitrate particles, their emissions would be equivalent to about 60 million tons per year of primary $PM_{2.5}$. However, ambient $PM_{2.5}$ in the U.S. is typically 30 to 50% sulfate and much less nitrate, except in southern California where nitrate may be as much as 45% in some winter periods (USEPA, 1996b). This implies a conversion to $PM_{2.5}$ of somewhat less than 10 million tons per year, or somewhat less than 20% of the gaseous precursor emissions.

Figure 2 shows geographically the areas that do not meet the current PM_{10} standard of 150 $\mu g/m^3$ (24-h average) and 50 &g/m^3 (annual average). It also indicates the source types that are thought to be the major causes of non-attainment with the standard. The number of additional areas of the country that will not meet a new $PM_{2.5}$ standard, and the additional sources needing control, will depend largely on the concentration values and averaging periods that are selected (Paisie, et al., 1997).

There is further uncertainty about exposures to airborne particles inside buildings. Considering the high fraction of time that people (especially those who may be most health-susceptible) spend indoors, there is a need to quantify the three major sources of indoor exposure: particles from outdoors, direct emissions from indoor sources, and resuspended particles from indoor activities (a type of fugitive source). Table 2 summarizes the various types of sources of particles found in buildings. Cigarette smoking, cooking, and penetration of outdoor particles through the building envelope have all been reported to make significant contributions to indoor concentrations of fine particles, but a substantial portion is due to unexplained indoor sources (Wallace, 1996).

Table 1

Summary of U.S. national inventory of particle and precursor emissions

		National Emissions (10^3 Mg/year)			
		$PM_{2.5}$	PM_{10}	SO_x	NO_x
Industrial Processing		600	800	200	1000
Combustion Sources					
industrial & commercial		300	500	19000	11000
residential		600	600	10	70
vehicular		500	600	800	10000
open burning		1100	1300	7	200
Fugitive Sources					
roads		3000	18000		
agricultural production		2000	11000		
construction		2000	8000		
Totals		10000	40000	20000	20000

Adapted from USEPA, 1996a.

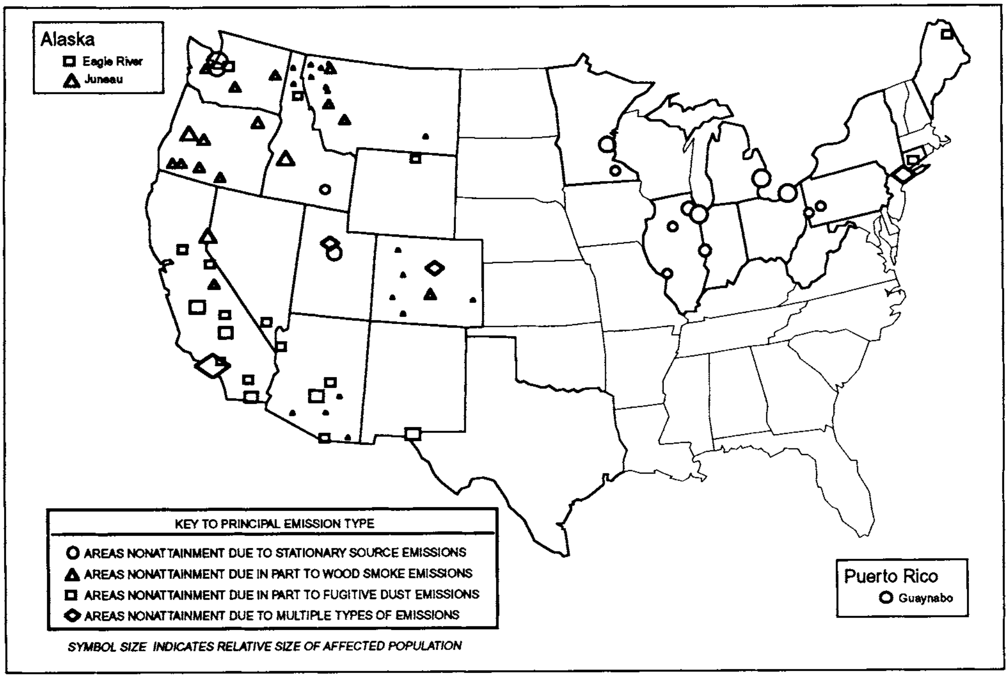

Figure 2. Areas designated non-attainment for PM_{10} particles, by emission type. (Adapted from USEPA, 1996c)

Table 2

Sources of exposure to indoor particles

Outdoor Origin	Indoor Origin
Outdoor air (through infiltration, ventilation)	Smoking, Cooking
Tracked-in soil	Space heating (esp. kerosene, wood)
Carried-in dusts	Ventilation systems, Humidifiers
- from industrial workplaces	Office machines
- from other places	Dust mites, Pets
	Diseased people (bacteria, viruses)
	Personal activities (e.g., hobbies)
	Maintenance/renovation activities
Resuspension of Particles of Either Outdoor or Indoor Origin	
Cleaning activities	
Maintenance/renovation activities	
People movements	

3. EMISSION RATES AND COMPOSITIONS OF PRIMARY PARTICLES

Table 3 presents an overview of emissions of primary particles in the U.S. Note that the four largest source categories are 'fugitive' types. As noted previously, the field data that are the basis of these estimates are being reevaluated. Cascade impactors used in these field studies may have experienced significant 'bounce' of larger particles onto impaction plates designed to collect smaller particles. The $PM_{2.5}$ fraction of PM_{10} may therefore be overstated somewhat. Even after adjustment of the data, however, fugitive sources are likely to remain significant, at least on a national basis, for $PM_{2.5}$.

As noted in Table 1, combustion sources comprise the next largest grouping. However, the constituents of combustion PM may be more toxic on average than the constituents of fugitive PM. Furthermore, combustion sources are generally closer to the population, and may lead to higher exposures per unit of emission.

Table 3
Overview of current knowledge on U. S. emissions of fine particles. (Numbers in regular type are typical values, selected from the referenced literature; *entries in italics are estimates or judgements by the author*)

Source Type [refs. for emissions data]	Constituents of Concern	Total U.S. Emission Rate (10^3 tons/yr)		Approx. U.S. Population in Close Proximity (millions)
		$PM_{2.5}$	PM_{10}	
Roads [a,b,c]	Fine silica and other crustal elements plus re-entrained carbon, asbestos, and metal compounds	3300*	18,000	*250*
Agricultural Production (incl. erosion) [a,b,c]	Fine silica and other crustal elements	2000*	11,100	*Mostly in rural areas*
Construction Activities [a,b,c]	Fine silica and other crustal elements plus industrial reentrainment of carbon, asbestos and metal compounds	1700*	8,500	*Mostly in urban areas*
Open Burning (incl. wildfires, agric. burning, etc.) [a,b,c]	Products of uncontrolled combustion	1130*	1320	*Mostly in rural areas*
Residential Wood Combustion [a,b,c]	Polycyclic Organic Matter (POM)	550	~600	*Mostly in rural and suburban areas*
Diesel Engine Combustion [a,b,c]	Products of incomplete combustion, PM precursor (NO_x)	450	500	*Mostly in urban areas*
Mineral Products Production [a,b,c]	Fine silica and other crustal elements	100	200	*Near urban areas*
Pulverized Coal Boilers [a,d]	Ar, Cr, Hg, Mn, Ni, Pb, Sb, Se, V, Cl, and PM precursors (SO_x, NO_x)	*Unknown*	160	*Utility: mostly rural* *Industrial: mostly near urban areas*

Heavy Fuel Oil Combustion [a,e]	Cr, Fe, Ni, Pb, V, POM, Cl, PM precursors (SO_x, NO_x)	*~30*	30	*Mostly in urban areas*
Residential Fuel Oil Combustion [a]	POM, PM precursor (NO_x)	*~20*	20	*Mostly in urban areas*
Waste Incineration [a,f,g,h]	As, Be, Cr, Cd, Hg, Ni, Pb, PCDD/F, PCB's	*Unknown*	~45	*Mostly near urban areas*
Metal Smelting and Refining [a,i]	Cd, Cr, Pb, Zn, SO_x	*Unknown*	400	*Mostly in rural areas*
Automobiles [j]	V, carbon, organics, PM precursor (NO_x)	*~20*	*~20*	*Mostly near urban areas*
Outdoor Air Introduced into the Indoor Environment	Fine and coarse particles	Unknown	Unknown	*250*
Tracked-in Dust	Pb, other heavy metals, pesticides	Unknown	Unknown	*250*
Indoor Activities (that generate or resuspend particles)	Metals, pesticides, combustion aerosol organics	Unknown	Unknown	*250*

* Estimates of fine particle emissions from these "fugitive" sources, although large compared to other sources in this table, are very uncertain and need to be confirmed.

References for Table 3

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c. Dunkins and Cowherd (1992).
d. Davison, et al. (1974), Kaakinen, et al. Klein, et al. (1975), White, et al. (1984), Markowski and Filby (1985), Kauppinen and Pakkanen (1990), Andren, et al. (1975), Billings and Watson (1972).
e. Bulewicz, et al (1974), Haynes, et al. (1978), Feldman (1982), Chung and Lai (1992)
f. Mumford, et al. (1986), Trichon and Feldman (1989), Trichon and Feldman (1991).
g. Lisk (1988), Greenberg, et al. (1978).
h. Shen (1979).
i. Harrison and Williams (1983).
j. Average of 10 mg/mile; 2×10^{12} vehicle miles traveled.

4. CONTROL OPTIONS

Managing the health risks of exposures to fine particles requires knowledge of the sources and types of particles that are most likely to cause health risks, and knowledge of the performance and costs of risk reduction technologies. Data on the chemical, physical, and toxicological characteristics of the emissions are needed to determine controllability and risk reduction potential.

Since many of the estimates of $PM_{2.5}$ emissions are estimated from data on PM_{10}, there is great uncertainty in the fine particle emissions inventory. In addition, there is a general lack of data on the chemical composition of fine particle emissions. The need for emission characterization is greatest for those sources with constituents (such as metals and acidic components) that are candidates for causal mechanism studies of respiratory health effects, and for those sources having the largest mass contributions to $PM_{2.5}$ in the environment.

As Table 4 implies, there are few data on the effectiveness and costs of emissions prevention, emissions reduction, or exposure reduction technologies for fine particles (i.e., $PM_{2.5}$). Reliable data on emission prevention for either industrial or indoor sources are nearly non-existent. Most of the available data on cost-effectiveness of emission controls are for industrial sources of total PM (and at best, for PM_{10}). Although limited data are available on the efficiency and cost of air cleaning to remove particles from indoor air, there are virtually no data on the effectiveness of air cleaning in reducing exposures to fine particles. Since indoor concentrations of particles approach outdoor concentrations when outdoor concentrations are high, or are about twice outdoor concentrations when outdoor concentrations are low (e.g., Spengler et al., 1981; Sheldon et al., 1989), and since people spend roughly an order of magnitude more time indoors than outdoors, it will be important to develop controls for indoor exposures.

Table 4

Overview of current knowledge on control of fine particles. (Values for efficiencies and costs are estimates or judgements by the author)

Source Type	Primary Control Options, Efficiencies for PM_{10}	Approximate Costs of PM_{10} Controls
Roads	Vacuum sweeping (0-50%) Water flushing & sweeping (0-96%) Paving and roadside improvements Covering trucks Speed and traffic reduction	Dependent on type of control, time of event, frequency of event/yr, and volume of traffic. Very limited published data.
Agricultural Production (incl. erosion)	Low tillage, punch planting, crop strips, vegetative cover, windbreaks Chemical stabilizers, irrigation	Dependent on crop type and regional weather conditions. Little data.
Construction Activities	Wet suppression of unpaved areas, material storage, handling and transfer operations Wind fences for windblown dust	Dependent on type of control, time of event, land area of event, and activity level of equipment. Very limited published data.
Open Burning (incl. wildfires, agric. burning)	Low wind speed and appropriate wind direction	Unknown.
Residential Wood Combustion	Replace with cleaner burning stoves or furnaces	~$1000 per replaced stove or furnace.
Diesel Engine Combustion	Combustion modification Improved fuel characteristics Particle traps	Very limited published data.
Mineral Products Production	Enclosing crushing, transfer areas Water spray suppression Chemical stabilization of unpaved traffic areas	Dependent on type of control and activity level of equipment. Little data.

Pulverized Coal Boilers	ESPs, Fabric Filters	Capital cost \$50-100/kW Annual cost 2-5 mills/kWh Total installed cost \$25-50 per m^3/h
Heavy Fuel Oil Combustion	Cyclones, ESPs	Unkown.
Residential Fuel Oil Combustion	Proper maintenance, modern furnaces	Unknown.
Waste Incineration	Fabric filters, ESP's, venturi scrubbers	Total installed cost \$15-30 per m^3/h.
Metal Smelting and Refining	ESPs, cyclones	Total installed cost \$15-30 per m^3/h.
Outdoor Air Introduced into the Indoor Environment	Air cleaners for ventilation air (30 - 98%)* Whole-building air cleaners (30 - 98%)* In-room air cleaners (30 - 98%)*	Capital cost \$3-10 per m^3/hr of outdoor air treated. Capital cost \$1-10 per m^3/hr of indoor air treated. \$200 to 800 per room.
Tracked-in Dust	Cleaning (e.g., vacuuming) Whole-building air cleaners (30 - 98%)* In-room air cleaners (30 - 98%)*	No published analyses. Capital cost \$1-10 per m^3/hr of indoor air treated. \$200 to 800 per room.
Indoor Activities (that generate or resuspend particles)	Source control, including maintenance Whole-building air cleaners (30 - 98%)* In-room air cleaners (30 - 98%)*	Highly variable; no published analyses. Capital cost \$1-10 per m^3/h of indoor air treated. \$200 to 800 per room.

Range of single-pass efficiency for removing particles. The effectiveness of air cleaners in reducing exposures to indoor particles is very dependent on installation and operating conditions, and is generally less than the single-pass efficiency.

5. SUMMARY

Growing concerns about the health risks of fine particles have led to the proposal of a revised ambient air quality standard for PM smaller than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) in the U.S. The existing data base on national emissions shows that sources of both primary and secondary particles make significant contributions. On a nationwide basis, the greatest mass of primary and precursor emissions come from combustion, fugitive, and industrial sources, in that order. In specific localities, any of these source categories can be dominant.

It is clear that the new emphasis on fine particles will focus risk management research on large sources and combustion sources of various types. The role of fugitive sources in creating exposures to fine particles is less clear, and field studies to obtain better emissions data are needed. The importance of exposures inside buildings is also unclear; the penetration of outdoor particles into buildings of various types and the role of indoor sources of particles need further study. Current knowledge of the costs and effectiveness of control options for PM_{10} is limited and poorly documented. Carefully documented data on control options for $PM_{2.5}$ are nearly nonexistent.

A risk management research program is now in the early stages of developing new information to address these questions:

- What sources contribute to the fine particle exposures that are of greatest concern to the health research and regulatory community?
- What are the emission rates and physical and chemical characteristics of particles from these sources?
- What are the most cost-effective prevention and control options for these sources (e.g., process changes, upgrades of existing controls, application of new technology)?

Research results will enable the U.S. Environmental Protection Agency to assist state and local regulatory agencies in the development of cost-effective local prevention and control strategies for reducing exposures to fine particles. These strategies are likely to be based on a combination of industrial process changes, improved operation of existing particle control devices, installation of new control equipment on selected sources, increased control of gaseous precursors to ambient fine particles, and control of particles in buildings.

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