

The Size of Suspended Particulate Matter in Air

Size distributions of ambient aerosols must be studied in order to determine their effects on the environment.

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Suspended particulate matter in air is recognized as a major pollutant that can have a pronounced effect on our health and well-being (1). The concentration, composition, and particle size of suspended particulate matter at a given site are determined by such factors as meteorological and other properties of the atmosphere, topographical influences, emission sources, and by particle parameters such as density, shape, and hygroscopicity. Ambient aerosols are generated from the incomplete combustion of fuels, incineration, industrial activity, photochemical and condensation processes, and from erosion and weathering. Once in the aerosol state, suspended particles undergo a removal process from the atmosphere by sedimentation, washout, diffusion to surfaces, and coagulation mechanisms (2).

The concentration of aerosols in air is measured routinely at sites throughout the United States, Great Britain, and other countries by various air-sampling devices. In the United States, particulate concentration is determined gravimetrically over a 24-hour period at about 200 sites with high-volume (Hi-Vol) air samplers (3). At sites in Great Britain and other European countries, smoke concentration is determined by measuring photometrically the degree of staining produced by the aerosol collected on

a paper filter (4). In the United States and Great Britain, gross particulate samples are commonly analyzed chemically for metals and water- and benzene-soluble components.

One of the most important properties of suspended particulate matter is the particle size distribution. Because the degree to which suspended particles penetrate the respiratory system is a direct function of particle size, the size distribution of the particles must be determined before their hazardous effects on health can be assessed (5). For example, a major fraction of particles that are 0.5 micrometer or less in diameter are deposited in the pulmonary portion of the respiratory system (1). Furthermore, the sizes and the constituents of suspended particles affect visibility (6), particle-particle and particle-gas interactions, soiling, deterioration of materials, and a wide range of other atmospheric, meteorologic, and geophysical phenomena including precipitation formation and the scattering of solar radiation back into space.

Until recently, very few measurements had been made of the particle size distribution of suspended particulate matter in ambient air. Particle-counting devices based on light scattering, electronic, or electrical measurements are of limited use in monitoring air pollution. Such devices suffer the disadvantages of high cost, a lower size resolution limit of about 0.2 μm , and complexity of operation, and the data ob-

tained from them are difficult to interpret. Similarly, available mechanical fractionating devices either require excessively long sampling periods for sufficient material to be collected for gravimetric or chemical analysis, or they provide too poor or too limited resolution of sizes. In addition, the mechanical size-classifiers often have such disadvantages as collection surfaces that are not amenable to gravimetric or chemical analysis, and operational difficulties.

A sampler described by Lee and Flesch (7, 8) overcomes most of these disadvantages and is now used in the National Air Surveillance Network (NASN). This sampler, designated the NASN cascade impactor, was adapted from an Andersen sampler (9), a commercially available device that has been used in some limited studies of air pollution (10, 11). The NASN cascade impactor can fractionate particles ranging from about 0.5 to 3.5 μm in diameter and can collect a sufficient amount of particulate matter on each of several stages over a 24-hour period to permit particle size distributions to be determined gravimetrically. Studies conducted in the United States and other countries with this instrument have provided data on the size distribution of aerosols in a variety of urban and non-urban atmospheres, inside buildings, and by roadways and tunnels.

In this article I describe briefly how particle size distributions are determined gravimetrically, then summarize the results of studies that demonstrate some of the effects of seasonal and meteorological changes, emission sources, indoor-outdoor relationships, and other factors on the size distribution of suspended particulate matter.

Characterization of Particle Size Distributions

The NASN cascade impactor is an intermittent sampler that operates continuously for periods of 24 hours. Particles are separated according to size by utilizing the differences in their aerodynamic dimensions. Five stages are used in series (see Fig. 1). Each stage has a different air inlet geometry

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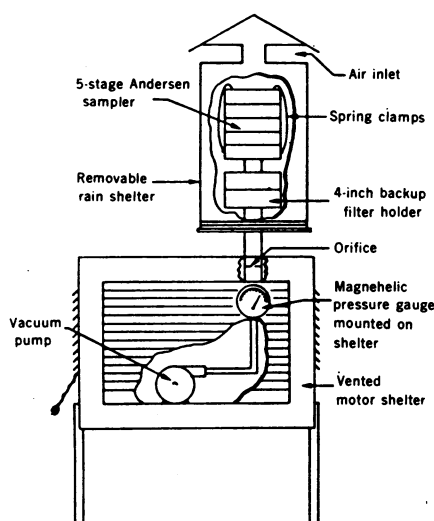


Fig. 1. Diagram of the modified Andersen impactor sampler and shelter. [From *Research and Development*, copyright by Technical Publishing Co., June 1972]

so that air velocities increase progressively with descending stages. Particulates with a large aerodynamic mass are impacted on the upper stages where

the air velocities are low, whereas particles with small aerodynamic mass are impacted on the lower stages where the air velocities are high, often in the sonic range.

The NASN cascade impactor was adapted from a six-stage Andersen sampler (9) which ordinarily operates at 1 cubic foot per minute (equivalent to 2.83×10^4 cubic centimeters per minute). The sixth stage of the Andersen sampler was removed, and a membrane filter 10.16 cm in diameter was placed downstream to collect the small unimpacted particles; the sampler could then be operated at a flow rate of 0.14 to 0.17 m³/min (5 to 6 ft³/min). The backup filter holder can accommodate glass fiber or membrane filters 10.16 cm in diameter. Both the impactor and the backup filter are housed in a removable rainshield to provide protection from precipitation and large debris. The rate of air flow is determined by measuring the pressure-drop across an orifice, approximately 0.64 cm in diameter, with a magnehelic or other pressure gauge having a capacity of 127 cm of water.

Table 1. Data obtained from the NASN impactor network during 1970, showing quarterly and annual size distribution of particulate matter suspended in air.

Quarter	Samples (No.)	Average concentration ($\mu\text{g}/\text{m}^3$)	Average MMD (μm)	Average geometric S.D.	Average mass of particulate (%)	
					$\leq 1 \mu\text{m}$	$\leq 2 \mu\text{m}$
Chicago						
1	4	97.8	2.31	10.41	37	48
2	6	82.4	0.51	8.16	63	74
3	7	98.3	0.62	5.88	61	75
4	4	63.0	0.66	8.00	58	71
Year	21	86.5	0.76	8.18	55	68
Cincinnati						
1	1	61.9	0.37	5.71	72	84
2	6	77.5	0.54	6.47	63	76
3	7	88.9	0.77	5.15	57	72
4	4	48.6	1.01	4.32	50	68
Year	18	74.3	0.70	5.49	59	74
Denver						
1	4	51.4	0.41	7.99	67	78
2	5	51.4	0.19	10.22	76	85
3	7	59.1	0.34	9.50	69	79
4	5	80.7	1.02	10.65	50	62
Year	21	59.7	0.40	10.50	65	75
Philadelphia						
1	2	60.4	0.31	6.02	74	85
2	6	50.9	0.26	11.21	71	80
3	7	66.1	0.62	3.91	64	81
4	5	56.8	0.55	6.34	63	76
Year	20	58.5	0.47	5.65	67	80
St. Louis						
1	5	81.2	0.97	6.61	51	65
2	5	73.7	0.53	10.33	61	72
3	9	76.5	0.89	5.69	53	68
4	3	44.5	1.02	5.34	50	66
Year	22	73.1	0.83	6.80	54	68
Washington, D.C.						
1	5	53.0	0.47	5.98	67	79
2	6	55.3	0.26	8.80	73	83
3	6	73.5	0.51	3.95	69	84
4	6	41.1	0.73	4.11	59	76
Year	23	56.3	0.46	5.22	68	81

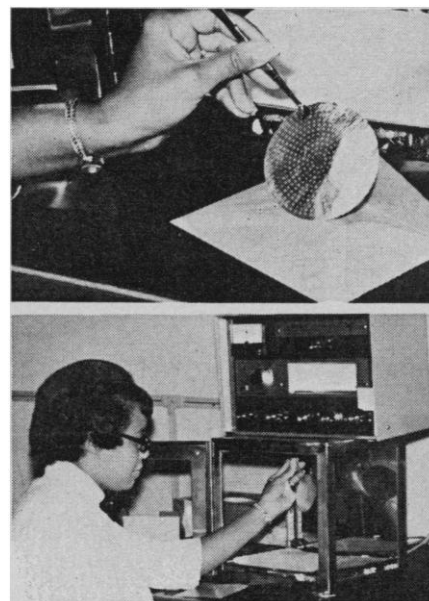


Fig. 2. Microweighing apparatus. [From *Research and Development*, June 1972]

A calibration of the pressure-drop across the orifice as a function of the flow rate is made by placing a dry test meter at the inlet end of the sampler and varying the air flow with an air inlet on the vacuum pump.

Samples of particulates from the five impactor stages are collected on aluminum foil disks 8.26 cm in diameter. These foil disks are conditioned in a temperature- and humidity-controlled room for 24 hours and weighed to the nearest microgram on a Kahn G2 microanalytical electrobalance. The samples are weighed in a chamber by suspending the aluminum foil disk from a wire attached to the microbalance as shown in Fig. 2. Both the electrobalance and the collection surfaces are housed in a room maintained at 50 percent relative humidity and a temperature of 22°C. Membrane filters (10.16 cm in diameter, 3.0 μm pore size) with a nylon mesh reinforcement, are used to collect unimpacted particles; they are conditioned and weighed in the same way as the aluminum disks.

The impactor is prepared for sample collection by placing a weighed aluminum disk on top of each stainless steel collection plate and a conditioned and weighed membrane filter in the backup filter holder. All the collection surfaces are handled with forceps. The sampler is operated for a 24-hour period, and the average flow rate is calculated from the rates of air flow at the beginning and at the end of the period. After samplings, the aluminum collection disks and the membrane filter are folded in half with the sample side touching,

placed in glazed paper envelopes, and returned to the laboratory where they are reconditioned in the temperature- and humidity-controlled room for 24 hours before they are reweighed.

The sizes of the particles separated by each stage of the sampler, or the "effective cutoff diameter" (ECD) for each stage, was determined (8) from the theory of Mercer (12) and Ranz and Wong (13). Fractionation is achieved between about 0.5 and 3.5 μm diameter expressed as equivalent spheres of unit density. The results obtained with concurrently operated NASN impactors come within about 15 percent of each other.

The cumulative distribution of particle sizes is prepared by plotting the logarithm of the ECD for each stage on the Y-axis as a function of the cumulative mass (percent) less than or equal to that at each stage on the X-axis; a standard least-squares linear regression is the fit to the data. Since the particle size of all suspended particles follows approximately normal logarithmic distribution, the mass median diameter (MMD) is found at the 50 percent mass cumulative point, and the geometric standard deviation (S.D.), which is a measure of the particle dispersion, is estimated from the ratio of the 84 percentile to the 50 percentile.

The size distribution curves can also be used to determine the percentages of particulate mass equal to or below various diameters. The actual concentration of particles less than or equal to a selected size can then be determined by multiplying the percentage of the cumulative mass for that fraction by the total aerosol concentration.

U.S. Network of Cascade Impactors

In January 1970, the NASN established a cascade impactor network of six stations: Chicago, Cincinnati, Philadelphia, Denver, St. Louis, and Washington, D.C. An NASN impactor was installed at the Continuous Air Monitoring Project (CAMP) station in each city (14) and was operated for a 24-hour period once every 2 weeks according to a random schedule. The purpose of this network was to characterize the particle size distribution of ambient aerosols in major urban areas of the United States and to obtain data which could supplement the information from other types of air surveillance networks (3). The impactor network was expanded to ten stations in 1971 by the

addition of two background stations at Grand Canyon, Colorado, and Cape Hatteras, North Carolina, and two more urban stations at Steubenville, Ohio, and Seattle, Washington. A network of 50 stations is planned to be in operation by 1975.

The data obtained from the impactor network in 1970 revealed two important characteristics of ambient aerosols that could not have been found by measuring total concentrations: suspended particulates in air are (i) pre-

dominantly smaller than 1 μm , and (ii) well described by a log-normal function as indicated by the good linear fit. Table 1 shows that except for the single valid sample collected during the first quarter in Cincinnati, the average MMD was lowest at all sites during the second quarter. The highest average MMD varied with season and with site. The lowest average annual MMD was found in Denver; the highest in St. Louis.

The concentration of suspended par-

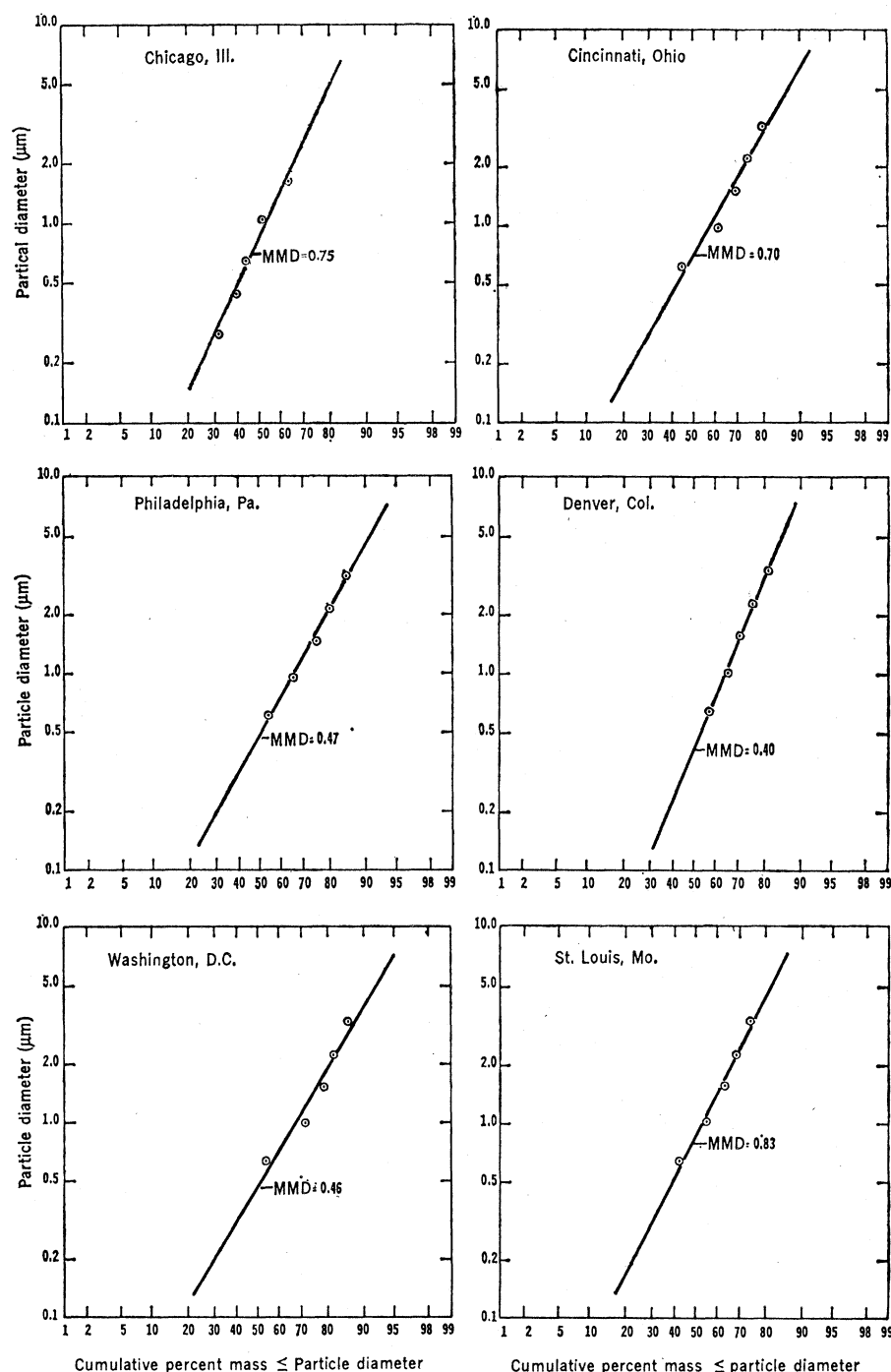


Fig. 3. Annual composited particle size distributions for 1970: Chicago, Cincinnati, Philadelphia, Denver, Washington, D.C., and St. Louis. [From *Research and Development*, June 1972]

Table 2. Data obtained with NASN impactors located at three different sites in Great Britain.

Month	Samples (No.)	Average concentration ($\mu\text{g}/\text{m}^3$)	Average MMD (μm)	Average geometric S.D.	Average mass of particulate (%)	
					$\leq 1\ \mu\text{m}$	$\leq 2\ \mu\text{m}$
<i>London</i>						
February	7	89.6	0.43	8.04	66	77
March	8	88.6	0.34	8.05	70	80
April	6	95.3	0.40	9.37	66	77
May	7	110.6	0.66	7.70	58	71
<i>Kew Observatory</i>						
February	8	53.6	0.46	6.04	67	80
March	7	57.4	0.27	6.70	75	85
April	9	64.6	0.29	4.84	79	89
May	9	72.9	0.43	6.12	68	81
<i>Eskdalemuir Observatory</i>						
January	3	51.3	0.30	5.21	77	87
February	8	36.5	0.06	15.63	84	90
March	8	34.8	0.08	12.49	84	90
April	9	36.2	0.09	9.68	86	92
May	7	56.3	0.31	6.33	74	84

ticulate matter equal to or below various size parameters can be determined from Fig. 3. For example, an average of 55 percent of the particles measured in Chicago in 1970 were less than or equal to $1 \mu\text{m}$ diameter. Since the average concentration was $86.5 \mu\text{g}$ per cubic meter for the total aerosol, $47.6 \mu\text{g}/\text{m}^3$ was less than or equal to $1 \mu\text{m}$ diameter, that is, 55 percent of $86.5 \mu\text{g}/\text{m}^3$.

The geometric S.D., roughly equivalent to the slope of the distribution, provides an indication of the particle dispersion or range of particle sizes in an aerosol. The broadest particle size distribution, gauged from the average annual geometric S.D. of 10.50, was observed at Denver; the narrowest distribution, shown by an average annual geometric S.D. of 5.22, occurred at

Washington (see Table 1). Except for Chicago, the dispersion patterns could be explained, at least in part, by seasonal factors. That is, during the first winter quarter, the narrow distribution of particles reflected the homogeneity of aerosols emitted predominantly from heating sources; in the second quarter, broadening of the distribution reflects the reduction of heating activity, a reduction that removes a fraction of the large aerosol particles and gives, effectively, a greater heterogeneity of aerosol particles; during the third quarter, the particle size distribution again becomes more narrowly dispersed with the influx of large dust and debris particles re-entrained from the ground during dry, windy conditions.

Data from Other Countries

Great Britain. In 1970, NASN cascade impactors were used as part of a project by the Environmental Protection Agency (EPA) to compare particulate monitoring methods used in the United States with those commonly used in Europe (15). Impactor samples were collected at three sites in Great Britain over a 4-month period. These sites were London, a major urban area; Kew Observatory, a suburb of London; and Eskdalemuir Observatory, a site having a clean-air background, in southern Scotland. As with aerosols found in the United States, the particle size distributions at the British sites were approximately log-normal as shown in Fig. 4; at all three sampling sites suspended particulate matter was also predominantly less than $1 \mu\text{m}$ in diameter (Table 2).

The largest average particle sizes occurred in London, whereas the smallest average particle sizes were found at Eskdalemuir. At Kew and Eskdalemuir, the particle size distribution curves shifted progressively from larger to smaller average particle diameters from January through April, reflecting the decreasing quantities of large particulates emitted during the incomplete combustion of fuels. During May, however, the average particle size at all three sites increased sharply, apparently reflecting the influence from tree pollen (16). At the London site the average particle size remained relatively constant throughout the months of February, March, and April, indicating that the particulate aerosol was not influenced, to any great degree, by large particulates emitted from the incomplete combustion of fuels.

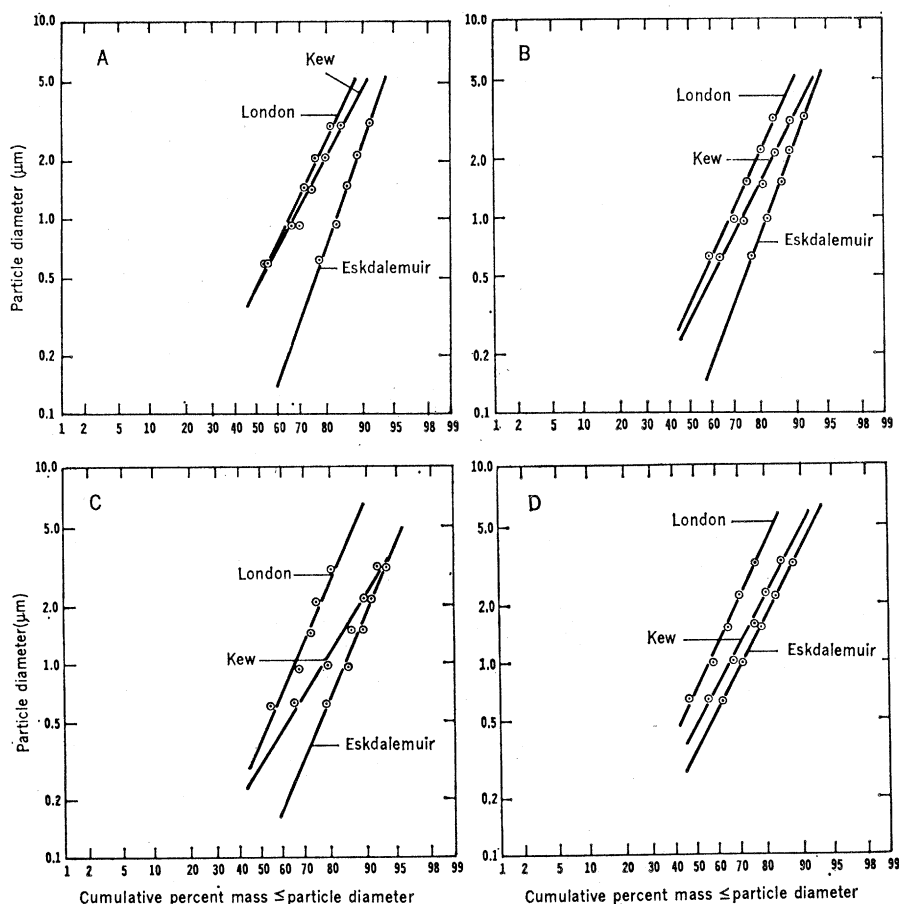


Fig. 4. Average monthly particle size distributions for London, Kew, and Eskdalemuir. (A) February, (B) March, (C) April, and (D) May, 1970. [From *Atmospheric Environment*, 6, 593 (1972)]

These data are reasonable because Eskdalemuir Observatory, a site with no nearby sources of particulate emission, exhibited the smallest particles. Particles less than $1\text{ }\mu\text{m}$ in diameter generally remain in the atmosphere for long periods of time before settling out and can, therefore, be transported long distances from their sources by meteorological and diffusional forces. In contrast, the larger particle sizes measured in London, and to some extent at Kew Observatory, indicate the presence of particulate emission sources within the immediate vicinity; the larger particles emitted from these sources did not settle out to any appreciable extent before the air was sampled.

Turkey. During a recent study in Ankara, Turkey (17), we made a number of size distribution measurements with an NASN cascade impactor. The population in Ankara relies heavily on low quality, highly volatile lignite coal for fuel. About 612,000 metric tons of lignite coal having low heat value are combusted annually and account for an estimated 65 to 70 percent of the particulate emissions, primarily from residential heating (18). Ankara, with a population of slightly over 1 million inhabitants, is surrounded by hills about 1100 meters high; the center of the city has an altitude of only about 850 meters. The ditch-like geographical configuration of the city, in conjunction with little wind motion (average wind speed is only 2 to 3 meters per second), contributes to the formation of frequent temperature inversions which prevent the dispersal of suspended particulate matter and other pollutants.

Ten samples were collected over 24-hour periods during April and May 1971. An impactor was placed on the second floor of the School of Hygiene located near the center of the city. Table 3 shows that the MMD of particles in Ankara air ranged from 1.21 to $2.19\text{ }\mu\text{m}$, with an average MMD of $1.79\text{ }\mu\text{m}$ for ten samples. Suspended particulate matter in Ankara air is therefore distributed in sizes predominantly greater than $1\text{ }\mu\text{m}$. In contrast, samples collected from April through June 1970 in Cincinnati and Philadelphia had average MMD's of 0.54 and $0.26\text{ }\mu\text{m}$, respectively (8); seven samples collected in London during May 1970 had an average MMD of $0.66\text{ }\mu\text{m}$ (15). In Fig. 5, the Ankara aerosol is compared with the size distribution of samples collected during an equivalent season in other cities. As shown above, these aerosols are typical of those found

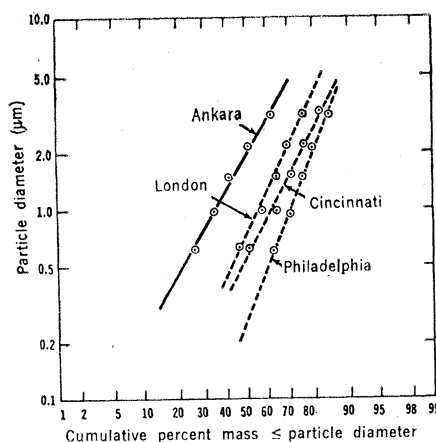


Fig. 5. Composite particle-size distribution curves: Ankara, ten samples collected from 26 April to 5 May 1971; London, seven samples collected during May 1970; Cincinnati, six samples collected from April to June 1970; Philadelphia, seven samples collected from April to June 1970. [From *Environmental Science Technology* 6, 929 (1972)].

throughout the United States and Great Britain and illustrate the predominantly submicrometer particle size at sites other than Ankara.

The range of particle sizes in Ankara air, as measured by the geometric S.D., is broad; similar dispersions are found in urban air of the United States and Great Britain. This broad particle dispersion may reflect (i) the age of the emission aerosol, or (ii) the coagulative activities which yield large particles by growth processes from smaller particles. With emission aerosols in Ankara, newly formed particles in a large size range may be injected into the air from the incomplete combustion of lignite fuel, whereas the smaller particles with a lower settling velocity remain suspended for a comparatively longer time. On the other hand, the almost continuous inversion conditions which exist in Ankara may produce the broad range of particulate sizes by growth processes.

Comparisons of the particle size distribution of suspended particulate matter in Ankara with other urban areas reveal the uniqueness of the aerosol in Ankara air. The large average particle size may be characteristic of aerosols emitted from the combustion of poor-quality lignite fuels. Although the aerosol concentration in Ankara is higher than the concentrations at sites in the United States and Great Britain, the proportion of particles that can be deposited in the pulmonary portion of the respiratory system (particles less than $0.5\text{ }\mu\text{m}$ in diameter) is less. For example, of the sites sampled, Ankara exhibited the largest average aerosol concentration of $115.9\text{ }\mu\text{g}/\text{m}^3$; however, only $25.4\text{ }\mu\text{g}/\text{m}^3$ was less than or equal to $0.5\text{ }\mu\text{m}$ diameter compared to $45.5\text{ }\mu\text{g}/\text{m}^3$ for London, $37.3\text{ }\mu\text{g}/\text{m}^3$ for Cincinnati, and $30.4\text{ }\mu\text{g}/\text{m}^3$ for Philadelphia during a similar period.

Aerosol Growth during Temperature Inversions

Measurements of particle size distribution taken during periods of temperature inversions in Cincinnati are shown in Fig. 6. Compared with data from samples collected during similar noninversion periods the previous year, these results indicate that aerosols grow markedly in size during stagnation conditions. Table 4 shows that the MMD's for samples collected during inversion periods were 1.32 and $1.99\text{ }\mu\text{m}$ as compared to 0.44 and $0.48\text{ }\mu\text{m}$ during the previous year, and $0.70\text{ }\mu\text{m}$ for the 1970 average MMD (8). The larger-size aerosols undoubtedly account for the visibility reduction or "haziness" during inversion conditions because the large particles scatter more light than the predominantly submicrometer-sized aerosols ordinarily present in urban air.

The lower geometric standard deviation

Table 3. Particle size distribution parameters measured in Ankara air. Samples were obtained on ten consecutive days from 26 April to 5 May 1971.

Day	Concentration ($\mu\text{g}/\text{m}^3$)	MMD (μm)	Geometric S.D.	Particulate mass (%)	
				$\leq 1\text{ }\mu\text{m}$	$\leq 2\text{ }\mu\text{m}$
1	104.2	1.44	7.00	43	57
2	119.2	1.64	5.79	39	55
3	107.9	1.21	6.38	46	61
4	104.8	2.09	5.24	33	49
5	68.4	1.33	6.32	44	59
6	90.8	1.82	5.14	36	53
7	121.3	1.99	5.21	34	51
8	132.8	1.79	4.19	35	54
9	163.0	2.19	4.61	31	48
10	146.5	2.03	4.25	32	50
Composite	115.9	1.79	5.20	37	53

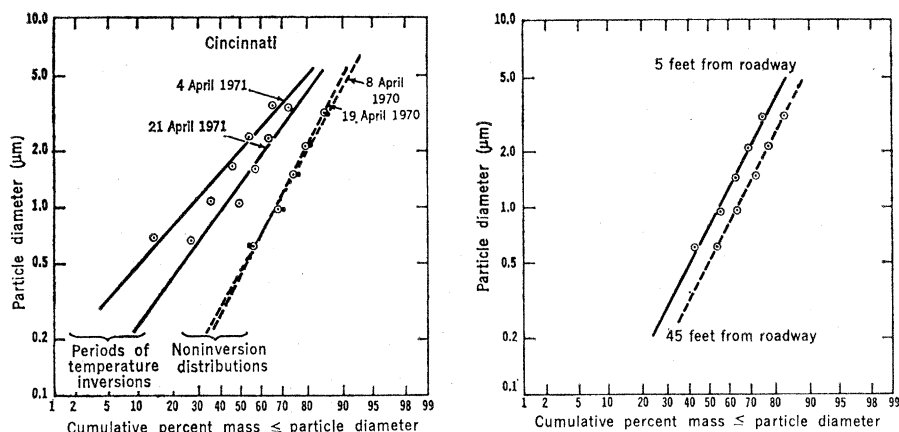


Fig. 6 (left). Particle-size distribution curves during temperature inversion and non-inversion conditions at Cincinnati CAMP station. Fig. 7 (right). Composite particle-size distribution curves at distances of 5 feet and 45 feet from roadway. Samples collected for 24-hour periods concurrently at Franklin D. Roosevelt Drive, New York City, on 2, 12, and 13 December 1969.

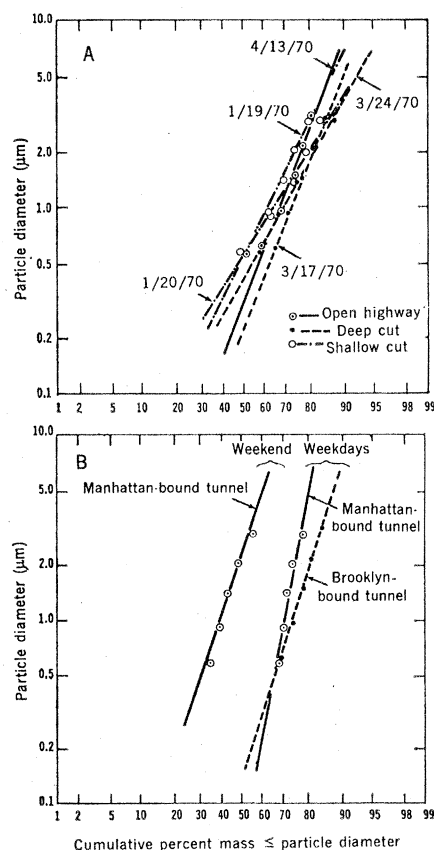
tions during inversion conditions show that the inversion aerosol is concentrated in a narrower range of sizes than the noninversion aerosol. This narrower distribution suggests that aerosol coagulation and condensation growth processes during stagnation periods affect submicrometer-sized aerosols to a greater degree than aerosols with larger-sized particles. It is interesting that the size parameters during stagnation periods appear to be unrelated to the concentration of the aerosol as shown in Table 4. It is reasonable to expect particle growth to be limited, however, so that above a certain size sedimentation becomes a major factor in limiting the aerosol concentration.

Aerosols and Highways

That air turbulence is produced by rapidly moving vehicles along a highway is well known. Although large dust particles and debris are reentrained from the roadway surface into the traffic air space, previous measurements of particulate matter found in automobile exhaust (11) have shown that the aerosol consists predominantly of particles less than 1 μm in diameter. The two divergent aerosol sources—that is, particle reentrainment from the roadway and vehicular-emitted particulates—are important because they affect the actual particle size distribution of highway aerosols. The dangers of inhalation of roadway aerosols to a driver with his windows open, for example, or to a traffic patrolman, may depend on the source of the aerosol.

As part of a study to define the concentration of selected pollutants as

a function of highway design, particle size measurements were made at a point 5 feet from a roadway and, simultaneously, 45 feet away (1 foot is equivalent to 0.3 meter). Figure 7 presents composite size distribution curves for measurements made alongside Franklin D. Roosevelt Drive in New York City. The average MMD 5 feet away was 0.72 μm, whereas 45 feet away the MMD was 0.45 μm. These distributions confirm that aerosols immediately adjacent to the highway have a larger



average size and are consequently less likely to be deposited in the respiratory system than aerosols found further away from the air turbulence area. As shown in Table 5, there was little difference in the range or dispersion of particle sizes between the two sampling points. It is surprising that particle size distributions in samples obtained near roadways are not very different from those obtained from urban sites when the samplers have been located in downtown areas but not immediately adjacent to highways.

In a previous study (19) measurements of particle size distribution were made at various types of highways in conjunction with measurements of total suspended particulate matter, carbon monoxide, hydrocarbons, and traffic parameters. The purpose of the study was to determine the influence of highway configuration on vehicle-emitted pollutants in an effort to obtain data that might serve as guidelines to highway planners. Thus NASN impactors were placed about 45 feet away from the roadway of four six-lane highways where the daily average vehicle count ranged markedly from site to site. The highways were located in New York City and included the Bruckner Expressway, an open highway with 55,000 vehicles per day; Cross Bronx Expressway, a deep cut roadway with 118,000 vehicles per day; Brooklyn-Queens Expressway, a shallow cut with 11,500 vehicles per day; and the Franklin D. Roosevelt Drive, a partially covered highway with 97,000 vehicles per day. The average traffic speeds were 48 miles per hour (1 mile is equivalent to 1.6 kilometers) at the Bruckner and Cross Bronx Expressways, 38 miles per hour at the Brooklyn-Queens Expressway, and 35 miles per hour at Franklin D. Roosevelt Drive.

The data for the four highway configurations (45 feet from the roadway) presented in Figs. 7 and 8 and Table 5

Fig. 8. Particle size distributions adjacent to various highway configurations, determined in 1970. (A) Samples collected over a period of 24 hours, 45 feet from New York City roadways. Open highway, Bruckner Expressway; deep cut, Cross Bronx Expressway at Joseph Avenue; and shallow cut, Brooklyn-Queens Expressway at Hicks Street. (B) Data for a tunnel. Samples collected at Brooklyn-Battery Tunnel over a period of 72 hours, 4 to 7 January; Manhattan-bound tunnel at weekend over a period of 70 hours, 9 to 12 January; and Manhattan-bound tunnel on a weekday over a period of 24 hours, 6 January.

show a high degree of similarity. Although there were some differences in the particulate concentrations, the aerosol particles were predominantly less than 1 μm in size with MMD's ranging from 0.21 to 0.72 μm . The particle size distribution curves in Fig. 8 show remarkable similarity at sites which are highly divergent in configuration, traffic volume, and vehicular velocity. As with ambient aerosols, the particle dispersion for highway-associated aerosols is variable with the geometric S.D. ranging from 4.72 to 12.93.

The data collected at the four highways show that aerosols away from the roadway turbulence area are not appreciably influenced by highway configuration or traffic parameters. Residents adjacent to highways, but away from the turbulence area, may, therefore, be exposed to automotive particulate emissions including lead, nitrate, halide, and hydrocarbon aerosols (11) which are of a size that can become deposited in the pulmonary portion of the respiratory system despite the configuration of the highway.

Aerosols inside tunnels present another interesting picture. Impactors were placed midway inside the tubes of the Brooklyn Battery Tunnel, where weekday traffic averaged about 25,000 vehicles per day traveling at an average speed of 25 miles per hour. During these weekday periods the average MMD's were in the range of 0.5 to 0.11 μm (Table 5). During a weekend period, however, the MMD value increased to 1.95 μm (Fig. 8B).

Most of these differences may be explained by variations in the environment of the tunnel, the ventilation effects produced by the tunnel exhaust system, and the higher velocity of the traffic during weekends. During weekdays, the major source of aerosol is from the vehicles themselves; few or no particles arise from external tunnel sources because of the low traffic velocity (25 miles per hour) and the confines of the tunnel walls. The tunnel aerosol can, therefore, be expected to reflect the nature of the aerosol emitted as automobile exhaust, that is, the lower submicrometer-size range (11). On the other hand, weekday traffic moves at a higher velocity and introduces a greater share of large roadway debris from outside the tunnel by virtue of a "piston effect," that is, the partial vacuum produced behind a rapidly moving vehicle in conjunction with the air pushed forward and out of the other end of the tunnel.

Indoor-Outdoor Relationships

Most of us spend the majority of our time indoors in our homes and places of business. Although it is important to characterize aerosols in outdoor air in order to assess inhalation hazards and other effects, it is even more important from the standpoint of our health to characterize aerosols in indoor air. A large part of particulate matter found indoors is generated within buildings through heating, cooking,

smoking of tobacco, and abrasion of surfaces. There is evidence, however, that particulate matter suspended in indoor air is related to outside aerosols (20).

As part of a study of air pollution in various types of buildings (21), samples were collected simultaneously inside and outside buildings with the NASN cascade impactor. The buildings were broadly classified into three categories: non-air-conditioned public buildings located in a downtown area;

Table 4. Data obtained during inversion and noninversion conditions in Cincinnati, Ohio.

Date	Concentration ($\mu\text{g}/\text{m}^3$)	MMD (μm)	Geometric S.D.	Particulate mass (%)	
				$\leq 1\ \mu\text{m}$	$\leq 2\ \mu\text{m}$
<i>Inversion</i>					
4 April	33.9	1.32	3.79	42	63
21 April	137.4	1.99	3.00	27	51
<i>Noninversion</i>					
8 April*	83.7	0.44	6.79	67	79
19 April	75.3	0.48	5.83	66	79

* Samples collected in 1970; all other samples were collected in 1971.

Table 5. Data obtained with NASN impactors located near highways in New York City.

Date	Concentration ($\mu\text{g}/\text{m}^3$)	MMD (μm)	Geometric S.D.	Particulate mass (%)	
				$\leq 1 \mu\text{m}$	$\leq 2 \mu\text{m}$
Brooklyn Battery Tunnel					
9–12 January	142	1.95	19.54	42	51
8–9 January	224	0.05		72	76
4–7 January	31.8	0.11	18.25	77	78
Bruckner Expressway					
13–14 April	86.3	0.28	12.93	69	78
Cross Bronx Expressway					
17–18 March	129	0.21	11.62	74	82
24–25 March	86.4	0.39	5.55	71	83
Brooklyn–Queens Expressway					
19–20 January	71.6	0.49	6.89	65	77
20–21 January	77.3	0.56	4.72	65	80
Franklin D. Roosevelt Drive*					
2–3 December	88.2	0.72	6.46	57	71
13 December	72.4	0.45	6.41	67	79

* Samples obtained in 1969; all other samples obtained in 1970.

Table 6. Buildings from which indoor samples of air were obtained by means of NASN impactors operated for 72-hour periods at each site during the fall of 1969 and winter of 1970.

Site	Description
<i>Public buildings</i>	
Hartford Public Library	Masonry construction; air-rights built over four-lane highway; not air conditioned; samples taken on main floor
Hartford City Hall	Masonry construction; adjacent to heavily traveled street; not air conditioned; samples taken on main floor
<i>Office buildings</i>	
250 Constitution Place	Six-story air-conditioned structure built over a five-level underground parking garage; samples taken on third floor
100 Constitution Place	16-story air conditioned structure not built over parking garage; samples taken on second floor
<i>Private homes</i>	
25 Blinn Street	Old duplex structure near I-84; not air conditioned; no gas stove; occupied by nonsmoking working couple; samples taken on first floor
60 Carrol Road	New colonial home near I-84; not air conditioned; no gas stove; occupied by nonsmoking active family; samples taken on first floor

modern air-conditioned office buildings in a downtown area; and private homes (see Table 6). The particle size distributions shown in Fig. 9 differed sharply between indoor and outdoor aerosols. Although all the distributions were log-normal, the particle sizes were smaller and the degree of particle dispersion was markedly less inside than outside the buildings. Table 7 shows that with

the exception of the sample obtained on 3 March at the Blinn Street site, the aerosol concentration was less, and the MMD and geometric S.D. were smaller inside than outside. The sharp differences in particle size between indoor and outdoor aerosols at the library and City Hall reflect the effect of turbulence. The aerosol outside these buildings is probably composed of

numerous large reentrained debris particles, whereas the larger particles of the indoor aerosol have undergone sedimentation in the comparatively undisturbed air. Similar, although less pronounced, differences were observed in the indoor and outdoor aerosols of the air-conditioned office buildings and, to some extent, the private homes. The range of particle sizes estimated from the geometric S.D. supports this explanation. Table 7 shows that outdoor aerosols are broadly dispersed, while indoor aerosols are narrowly dispersed and reflect the removal of large particles by filtration, sedimentation, and impaction mechanisms. It is generally accepted that particles approximately $0.5 \mu\text{m}$ in diameter or less are deposited in the pulmonary portion of the respiratory system (1). In the data reported here, 22 to 62 percent of the outdoor aerosol was less than or equal to $0.5 \mu\text{m}$ diameter compared to 48 to 75 percent of the indoor aerosol. These findings show that when the concentrations of indoor and outdoor aerosols are about the same, the indoor aerosol presents a greater inhalation hazard. For example 38 percent of the outdoor aerosol collected at 100 Constitution Place was less than or equal to $0.5 \mu\text{m}$ diameter compared to 75 percent for the indoor aerosol. Since the concentrations of particles were almost equal at this site, that is, $39.9 \mu\text{g}/\text{m}^3$ outside and $36.3 \mu\text{g}/\text{m}^3$ inside, the aerosol concentration less than or equal to $0.5 \mu\text{m}$ diameter was $15.2 \mu\text{g}/\text{m}^3$ outside but $27.2 \mu\text{g}/\text{m}^3$ inside. At most sites, however, the lower concentration of particles inside can reduce the hazards of inhalation. For example, 52 percent of the indoor aerosol at 250 Constitution Place was less than or equal to $0.5 \mu\text{m}$ diameter compared to only 34 percent outside. This represented a concentration of $19.4 \mu\text{g}/\text{m}^3$ inside (52 percent of $37.3 \mu\text{g}/\text{m}^3$) which was not much higher than the outside concentration of $17.0 \mu\text{g}/\text{m}^3$ (34 percent of $49.9 \mu\text{g}/\text{m}^3$). Many more studies of this type should be conducted, as well as studies of the chemical composition of the fractions of various sizes.

Conclusions

By means of the NASN cascade impactor the particle sizes of ambient aerosols can be measured by fractionating the particles according to their aerodynamic dimensions, and the frac-

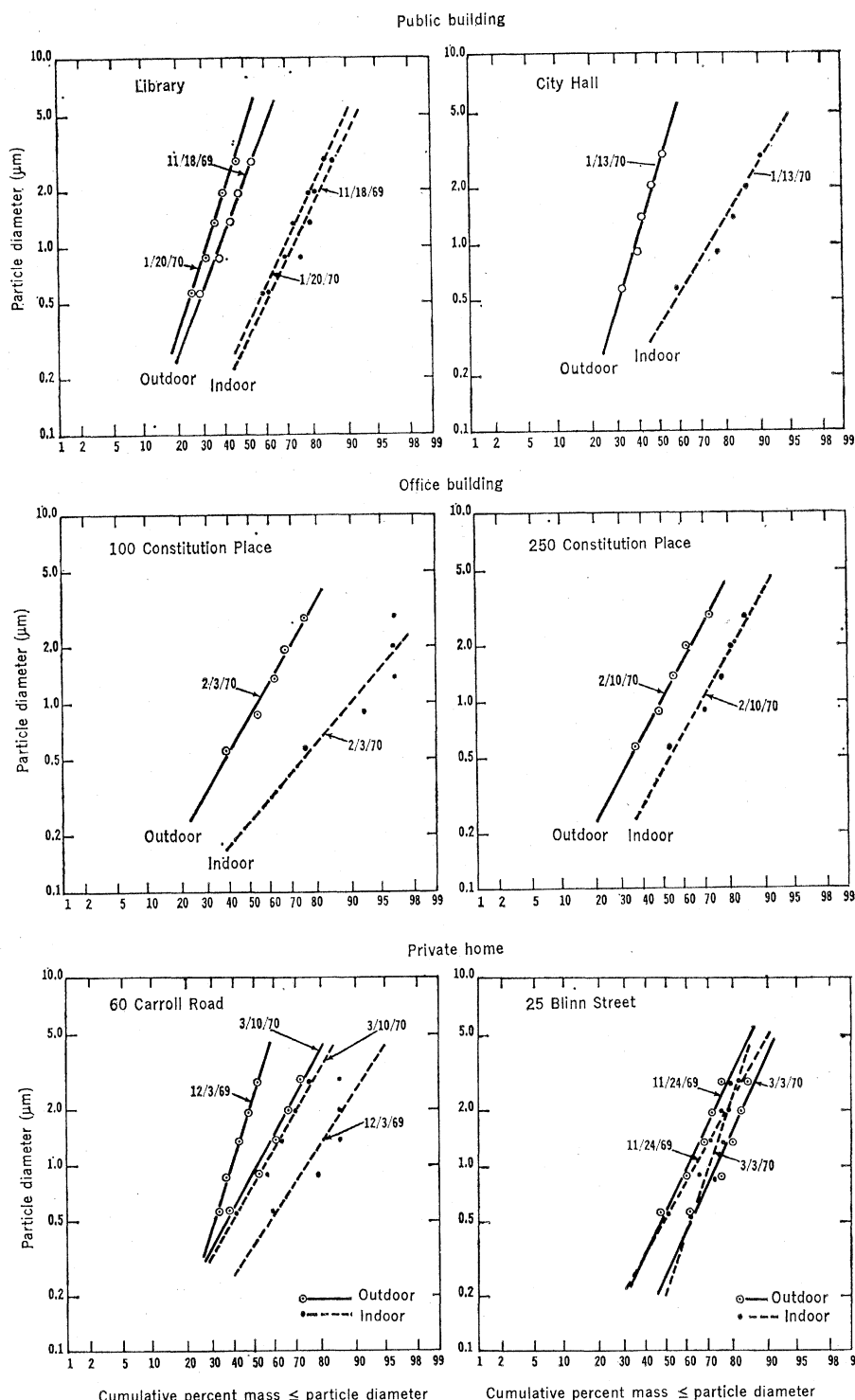


Fig. 9. Particle size distributions found inside and outside buildings in Hartford, Connecticut.

tions can be quantified gravimetrically. Data obtained with this instrument show that particulate matter suspended in urban air in the United States and Great Britain is remarkably uniform in distribution and that the particles are predominantly less than 1 μm in diameter. In Ankara, Turkey, the high proportion of particles larger than 1 μm in diameter are associated with the incomplete combustion of lignite; particulate concentrations in Ankara, however, are considerably higher than those measured elsewhere. The growth of particles in air during temperature inversions occurs in Cincinnati; the greater proportion of large particles found there during inversion periods can account for the reduction in visibility. Aerosols in the vicinity of highways are composed of particles larger than those found some distance away because of the reentrainment of debris by the traffic-induced turbulence. On the other hand, highway configuration and traffic volume have little effect on the size distribution of aerosols in samples collected away from the turbulence area of the roadway. Aerosols measured inside buildings are predominantly smaller in size and exhibit a narrower range of sizes than aerosols outside buildings.

Much more work must be done before we can determine the extent to which our health may be affected by particles suspended in the air we breathe. Instruments are required that permit more effective size fractionation and that have higher sampling rates than the NASN cascade impactor. A four-stage impactor that collects samples at 20 cubic feet per minute and can be installed on a conventional high-volume sampler has recently become available and may permit greater resolution of particle sizes (22). To control pollution, we must know more about the chemical composition of the various fractions of aerosols and about the size distributions of aerosols in and near specific emission sources. We know practically nothing about such aerosols at present. Similarly, very little is known about condensation nuclei, that is, particles 0.01 to 0.001 μm in diameter. Because control devices remove particles in size ranges well above 0.01 μm , we ought to know the long-term trends of condensation nuclei in order to assess their potential effects on weather and climate. The effects on our health of suspended particulate matter inside houses and buildings where we spend most of our time must also be more clearly defined.

Complete and reliable measurements of particle sizes could lead to better methods of pollution control that would improve the quality of our lives.

References and Notes

1. U.S. Department of Health Education, and Welfare, *Air Quality Criteria for Particulate Matter*, Pub. No. AP-49, Washington, D.C. (1969).
2. M. Corn, in *Air Pollution*, A. C. Stern, Ed. (Academic Press, New York, ed. 2, 1968), vol. 2, p. 57. *Air Pollution* consists of three volumes containing resource information on air pollution.
3. G. B. Morgan, G. Ozolins, E. C. Tabor, *Science* **170**, 289 (1970).
4. M. Clifton, *Proc. Roy. Soc. Med.* **57**, 615 (1964).
5. P. E. Morrow, *Amer. Ind. Hyg. Ass. J.* **25**, 213 (1964).
6. W. E. K. Middleton, *Vision Through the Atmosphere* (Univ. of Toronto Press, Toronto, 1952).
7. R. E. Lee, Jr., and J. P. Flesch, "A gravimetric method for determining the size distribution of particulates suspended in air," paper presented at the annual meeting of the Air Pollution Control Association, New York, N.Y., 22-26 June 1969.
8. R. E. Lee, Jr., and S. Goranson, "The NASN cascade impactor network: part I, the size distribution of suspended particulate matter in air," paper presented at the national meeting of the American Chemical Society, Washington, D.C., 12-17 September 1971.
9. A. A. Andersen, *Amer. Ind. Hyg. Ass. J.* **27**, 160 (1966).
10. J. Wagman, R. E. Lee, Jr., C. H. Axt, *Atmos. Environ.* **1**, 479 (1967); R. E. Lee, Jr., R. K. Patterson, J. Wagman, *Environ. Sci. Technol.* **2**, 288 (1968); R. E. Lee, Jr., and R. K. Patterson, *Atmos. Environ.* **3**, 249 (1969).
11. R. E. Lee, Jr., R. K. Patterson, W. L. Crider, J. Wagman, *Atmos. Environ.* **5**, 225 (1971).
12. T. T. Mercer, *Health Phys.* **10**, 873 (1964).
13. W. E. Ranz and J. B. Wong, *Ind. Hyg. Occup. Med.* **5**, 464 (1952).
14. G. A. Jutze and E. C. Tabor, *J. Air Pollut. Control Ass.* **13**, 278 (1963).
15. R. E. Lee, Jr., J. S. Caldwell, G. B. Morgan, *Atmos. Environ.* **6**, 593 (1972).
16. D. L. R. Bailey, personal communication.
17. Committee on the Challenges of Modern Society, "Assessment of air quality in Ankara, Turkey," in *Guidelines to Assessment of Air Quality*, North Atlantic Treaty Organization, Publ. No. 6 (Environmental Protection Agency, Rockville, Md., 1971); R. E. Lee, Jr., and C. F. Smith, *Environ. Sci. Technol.* **6**, 929 (1972).
18. Based on information provided by the Engineering Investigation Group (MAG) of TBTA, a research institute in Ankara.
19. "Final report on the study of air pollution aspects of various roadway configurations," submitted to the New York City Department of Air Resources, 1971; L. H. Dworetzky, W. H. Giles, C. Simon, R. E. Lee, Jr., "Report on urban expressway air pollution at ten sites in New York City," paper presented at Symposium on Relation of Land Use and Transportation Planning to Air Quality Management, Rutgers Univ., New Brunswick, N.J., 13 October 1971.
20. V. J. Schaefer, V. A. Mohnen, V. R. Veirs, *Science* **175**, 173 (1972).
21. J. Yocom, W. L. Clink, W. A. Cote, "A study of indoor-outdoor air pollutant relationships," final report prepared by the Travelers Research Corporation for the U.S. Environmental Protection Agency under contract CPA 22-69-44, May 1970. I derived the various particle size distribution relationships from data contained in this report.
22. The NASN cascade impactor was obtained from the Research Appliance Company, Allison Park, Pa. The Hi-Vol sampling head is available from 2000, Inc., 2000 Sullivan Road, College Park, Ga. I thank A. Cohen for providing the data shown in Fig. 6.

Table 7. Particle size parameters for air samples collected inside and outside buildings.

Outside or inside	Date	Concen- tration ($\mu\text{g}/\text{m}^3$)	MMD (μm)	Geo- metric S.D.	Particulate mass (%)	
					$\leq 1 \mu\text{m}$	$\leq 2 \mu\text{m}$
Library						
Out	18 November 1969	38.4	2.14	12.6	39	49
In	18 November 1969	30.5	0.26	7.46	75	85
Out	20 January 1970	138	3.91	19.5	33	42
In	20 January 1970	50.9	0.33	8.04	71	81
City Hall						
Out	13 January 1970	66.6	2.14	26.2	41	50
In	13 January 1970	48.5	0.34	4.37	77	89
100 Constitution Place						
Out	3 February 1970	39.9	0.79	5.42	56	71
In	3 February 1970	36.3	0.22	3.07	91	98
250 Constitution Place						
Out	10 February 1970	49.9	1.02	5.66	50	65
In	10 February 1970	37.3	0.42	5.43	70	82
25 Blinn Street						
Out	24 November 1969	41.9	0.54	7.71	62	74
In	24 November 1969	24.6	0.50	5.68	66	79
Out	3 March 1970	18.2	0.24	8.43	75	84
In	3 March 1970	27.4	0.19	16.8	72	80
60 Carrol Road						
Out	3 December 1969	30.5	2.15	20.7	41	50
In	3 December 1969	26.6	0.36	4.30	76	88
Out	10 March 1970	64.9	0.87	5.84	54	69
In	10 March 1970	60.0	0.77	5.46	56	72