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The Trend of Suspended Particulates in Urban Air 1957-1964

U.S. DEPARTMENT OF HEALTH, EDUCATION AND WELFARE

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THE TREND OF SUSPENDED PARTICULATES IN URBAN AIR: 1957 - 1964

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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
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CONTENTS

ABSTRACT

The trend toward lower average concentrations of suspended particulate pollution in the urban air in the United States from 1957 through 1963, indicated by measurements of the National Air Sampling Network, is investigated from three aspects: (1) statistical significance, (2) the validity of the sampling and analytical methods, and (3) relationship to environmental changes in the vicinity of the sampling stations. The evidence indicates that the decline is real and that the methods employed can reliably detect substantive changes in the concentration of suspended particulate pollution. Information on changes in the environments of a representative group of stations revealed that where significant decreases in suspended particulates occurred, they were often logically related to specific changes in source emissions. Thus the indicated decline in the suspended particulate portion of urban air pollution is verified and substantially accounted for. A statistically significant increase in the annual mean occurred in 1964; if continued, such an increase would suggest the possibility of cyclic influences.

INTRODUCTION

Since 1957, samples of suspended particulate matter have been collected for the National Air Sampling Network (NASN) for a period of at least 1 year at stations in more than 300 cities in the United States. One reason for collecting these data is to assess any trends in the particulate portion of urban air pollution.

In the most recent summary of NASN data,* a downward trend of suspended particulates was reported for the period 1957-1963 (Figure 1). The causes of such a trend are of obvious importance in understanding air pollution. It should be noted, however, that although the particulates are an important and usually conspicuous portion of urban air pollution they represent only about 1%, by weight, of the six major gaseous pollutants measured by the Continuous Air Monitoring Program.† Two of these gaseous pollutants, nitrogen dioxide and sulfur dioxide, have been measured at several NASN stations. The gas concentrations measured by the NASN (beginning in 1960) and by CAMP (beginning in 1962) have not followed the trend shown by the particulates, but have remained essentially the same or have increased slightly. Therefore, the decline in the annual average of urban suspended particulates cannot be interpreted as an indication that the general problem of atmospheric contamination is subsiding.

Results of the search for the reasons behind the decline in the particulate portion of urban air pollution are discussed in the following pages under three broad headings: (1) statistical evaluation of the data, (2) scrutiny of the sampling and analytical procedures, and (3) survey of selected station environments.

STATISTICAL EVALUATION

One immediately recognized factor that influences the annual mean of suspended particulates for the entire Network is that many stations do not sample each year. Some stations sample on alternate years. Some stations have been discontinued and many have been added since 1957 as the scope of the NASN sampling program expanded. Thus the annual mean of suspended particulates for the Network

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* "Air Pollution Measurements of the NASN, Analysis of Suspended Particulates, 1963," U.S. Department of Health, Education, and Welfare, Public Health Service, Division of Air Pollution, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, 1965.

† Measurements of sulfur dioxide, nitric oxide, nitrogen dioxide, total hydrocarbon, carbon monoxide, and total oxidant are summarized in "Air Pollution in Six Major U. S. Cities as Measured by the Continuous Air Monitoring Program (CAMP)," D. A. Lynn and T. B. McMullen, U. S. Department of Health, Education, and Welfare, Public Health Serivce, Division of Air Pollution, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, 1965.

is based on a slightly different set of stations each year. Although the list of participating stations changes somewhat from year to year, there is within the Network a group of stations where samples have been collected, with few exceptions, each year since 1957. These 'every-year' stations make up a fixed group whose annual mean is unaffected by the additions and deletions that can influence the annual mean of the full Network. The trend in annual mean for the 'every-year' group parallels and strongly influences the trend in annual mean for the entire Network (Figure 1). The mean for the 'every-year' group is higher because this group is disproportionately composed of larger communities. Since this 'every-year' group lends itself more readily to statistical analysis than the full Network, the investigation concentrates on these stations.

The values for the annual mean of the 'every-year' group in Table 1 show that an appreciable decrease occurred over the period 1957-1963, with major decreases in 1959 and 1961. The annual mean shows an increase in 1964; continuation of such an increase would raise the possibility that the year-to-year variation may be due in part to some cyclic influence. Statistical analysis of the data summarized in Table 1 shows that a change of at least 3.6 percent can be considered significant.* On this basis, the decreases that occurred in 1959 and in 1961 are significant, as is the increase in 1964.

Table 1. ANNUAL GEOMETRIC MEAN SAMPLE CONCENTRATION OF SUSPENDED PARTICULATES at 'EVERY-YEAR' STATIONS

	1957	1958	1959	1960	1961	1962	1963	1964
'Every-year' [a] Station Mean	119.3	117.6	112.0	109.9	101.1	100.9	100.2	106.3
Change from Previous Year, %		-1.4	-4.8	-1.9	-8.0	-0.2	-0.7	+6.0

[a] A complete list of the 'every-year' stations is given in Appendix A.

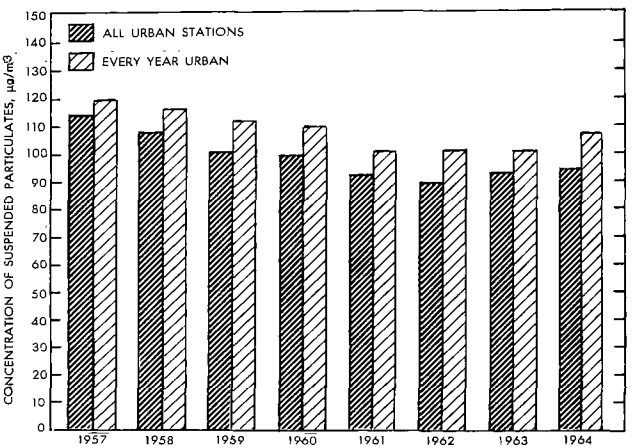

Figure 1. Annual geometric mean concentrations of suspended particulates at urban stations.

* Calculated through the Student's "t" parameter, using an average of 1500 samples per year and geometric standard deviation of 1.8, at the p=0.05 level of significance.

Additional evidence for the trend to lower annual mean concentrations of suspended particulates for the group of 64 'every-year' stations is found in the grouping of the maximum and minimum annual means for the individual stations during the period 1957 to 1963. If conditions in the station environments had remained essentially unchanged during the period, fluctuations in the annual means would be primarily the result of random sampling variations. If that were so, then the maximum or the minimum annual mean for each station should have an equal chance of occurring in any year and the occurrences of the maximum annual means and minimum annual means for the group as a whole would be expected to be distributed more or less equally throughout the 7-year period. They are not. Table 2 illustrates the bunching of maxima in the early years and the abrupt increase in the number of minima from 1961 on. The occurrences of maxima and minima depart from a random distribution (equal number each year) with a chance of less than 5 in 1000 that this abnormal distribution could be due merely to random variability in the data.

Table 2. DISTRIBUTION OF MAXIMUM AND MINIMUM ANNUAL MEAN CONCENTRATIONS OF SUSPENDED PARTICULATES AT 'EVERY-YEAR' STATIONS

Year	Number of maxima	Number of minima
1957	17 1/2[a]	2
1958	16	6 1/2
1959	9 1/2	3 1/2
1960	8	6
1961	4	16 1/2
1962	5 1/2	16
1963	3 1/2	13 1/2

[a] If the same annual mean occurred in 2 years, each was given a score of 1/2.

Since the total decline occurred in more than 1 year, it is necessary to compare a period before the drop with a period after the drop to define the total magnitude of the change. Since the annual mean concentration changed little from 1957 to 1958, the values for these 2 years prior to the beginning of the decline have been combined for comparison with the combined means for 2 years after the decline, 1962 and 1963, when the annual mean again was relatively constant. The combined mean for all 'every-year' stations for the first 2 years is 118 $\mu g/m^3$; for the last 2 years, 101 $\mu g/m^3$. This is a net change of 17 $\mu g/m^3$ or 14.4 percent. Statistically, the change is highly significant.* Comparison of the changes in mean levels for the individual 'every-year' stations (Table 3) shows that levels for 44 stations decreased between the period 1957-8 and 1962-3. Only seven stations reported a net increase.

* There is less than 1 chance in 1000 that the difference in levels is due only to normal variability in the data.

Table 3. DISTRIBUTION OF PERCENT CHANGE IN MEAN CONCENTRATION OF SUSPENDED PARTICULATES AT 'EVERY-YEAR' STATIONS FROM 1957-1958 to 1962-1963

Percent change in 2-year mean	No. of stations
Total positive	7
+16% to +25%	2
+ 6% to +15%	5
± 5% (i.e., no change)	13
6% to -15%	17
-16% to -25%	11
-26% to -35%	12
-36% to -45%	4
Total negative	44
All stations	64

From the results of these analyses it can be concluded that decreases in annual mean concentrations occurred at enough of the 'every-year' stations to produce a significant drop in the combined annual mean for the entire 'every-year' group.

Approximately half of the total change is concentrated in 1961, and since the investigation was concentrated on the 1961 drop, the remainder of this report will present information pertaining to that period. Such a change could originate from two possible sources: (1) internal, as a result of some change in operation or measurement techniques, and (2) external, as a result of some real change in the environments of some or all of the 64 urban areas. These possibilities are explored in detail in the following sections.

SAMPLING AND ANALYTICAL PROCEDURES*

Suspended particulate samples are obtained with a high-volume air sampler, which draws air at a rate of about 60 cubic feet per minute through an 8- by 10-inch glass-fiber filter capable of removing practically 100 percent of all particulates of 0.3 micron diameter or greater. During each sampling period the sampler is operated for 24 hours.

The prescribed procedures for handling and measurement of the samples have remained essentially unchanged since the organization of the Network in its present form in 1957. The possibility of a bias

* Sampling and analytical procedures of the NASN are described in detail in the previously cited 5-year summary.

resulting from some unintentional alteration of procedure must be considered, however. If such an internal change had occurred, the values from all stations would have been affected to the same extent, resulting in a simple transposition of the distribution of annual change in means. The distributions of the year-to-year changes in percent of the previous year's concentration are shown in Table 4.

Table 4. DISTRIBUTION OF 'EVERY-YEAR' STATIONS BY PERCENT CHANGE IN CONCENTRATION OF SUSPENDED PARTICULATES FROM PREVIOUS YEAR.

Percent change in annual mean particulate concentration	1957 to 1958	1958 to 1959	1959 to 1960	1960 to 1961	1961 to 1962	1962 to 1963
Total positive	16	20	13	15	23	24
+46% to +55%					1	1
+36% to +45%	1	1	1		3	
+26% to +35%	2	3	2	1	2	2
+16% to +25%	3	6	2	3	5	9
+ 6% to +15%	10	10	8	11	12	12
± 5% (i.e., no change)	12	14	24	17	19	18
6% to -15%	13	10	13	13	11	11
-16% to -25%	9	6	7	14	9	8
-26% to -35%	2	5	3	4	2	3
-36% to -45%				1		
-46% to -55%						
Total negative	24	21	23	32	22	22
No data	12	9	4	—	—	—
All stations	64	64	64	64	64	64

The distribution of changes from 1960 to 1961 is distinctly different from that of other years but is skewed, not simply transposed. This indicates that the annual mean for each station was not shifted uniformly by some procedural bias. Such a bias, if it had occurred, would be expected to affect the results for nonurban 'every-year' stations as well. There is no evidence of such a change. The annual mean concentration of suspended particulates at 20 nonurban 'every-year' stations* has remained essentially unchanged at 24 ± 1 $\mu g/m^3$ for the period 1958 through 1963.

If an error had been introduced in assessing the gross weight of suspended particulates, a conflicting upward trend might occur in the proportions of constituent pollutants such as organic matter, sulfates, nitrates, and metals. Data for the benzene-soluble organic matter are more comprehensive than for any of the other particulate fractions (Table 5).

* The nonurban 'every-year' stations are listed in Appendix B.

Table 5. ANNUAL MEAN CONCENTRATION OF BENZENE-SOLUBLE ORGANIC MATTER AT 'EVERY-YEAR' STATIONS

	1957	1958	1959	1960	1961	1962	1963
Concentration, $\mu g/m^3$	7.14	9.79	8.44	8.29	7.42	7.44	7.13
Fraction of gross particulates, %	5.98	8.32	7.53	7.55	7.35	7.38	7.00

After an unexplained jump in concentration between 1957 and 1958, the mean level of the benzene-soluble organic fraction for these 64 stations has been trending downward. The proportion of organics in the gross particulates shows no conflicting discontinuity in 1959 or 1961.

Data on the other measured fractions — nitrates, sulfates, and metals — generally lack the continuity from year to year that would permit averaging the means for a group of stations. Therefore, seven stations have been selected for which data on nitrates, sulfates, and iron are most complete; the relative concentrations of these constituents are summarized in Table 6 as percentages of the annual mean suspended particulates. No contradictory trends in constituent particulates are indicated among the few stations listed for which the data are adequate to provide some perspective. The annual mean concentrations of suspended particulates for the seven stations are listed in Table 7.

Table 6. ANNUAL FRACTION OF CONSTITUENT POLLUTANTS IN TOTAL SUSPENDED PARTICULATES FOR SELECTED CITIES

		Percent of total suspended particulates					
		1957	1958	1959	1960	1961	1962
Baltimore, Md.	Nitrate	1.5	1.7	0.7	1.3	1.1	1.2
	Sulfate	8.7	11.9	12.1	10.5	13.4	12.6
	Iron		1.4			2.0	1.1
Boston, Mass.	Nitrate		1.0			0.6	0.3
	Sulfate		10.0			12.4	11.2
	Iron		1.2				
Chattanooga, Tenn.	Nitrate		0.9	0.7		0.5	
	Sulfate		5.7	5.3		5.1	
	Iron					1.8	
Indianapolis, Ind.	Nitrate	1.4	1.4			1.0	1.3
	Sulfate	8.3	7.9			8.4	8.3
	Iron		1.2			4.2	0.9
Los Angeles, Cal.	Nitrate	3.1	3.8	3.3		4.3	
	Sulfate	6.9	6.8	6.8		8.1	
	Iron		1.1		1.8	1.5	2.5
Philadelphia, Pa.	Nitrate		1.5	0.8		1.4	1.3
	Sulfate		10.0	12.2		16.4	12.3
	Iron		1.5		2.7	1.9	2.1
Pittsburgh, Pa.	Nitrate		1.5	1.0	1.6		1.6
	Sulfate		9.1	8.5	9.2		10.6
	Iron				3.3		

Table 7. ANNUAL MEAN CONCENTRATIONS OF SUSPENDED PARTICULATES FOR CITIES LISTED IN TABLE 6

	Geometric mean concentration, $\mu g/m^3$					
	1957	1958	1959	1960	1961	1962
Baltimore, Md.	141	118	142	127	132	127
Boston, Mass.	130	145	127	130	125	131
Chattanooga, Tenn.		215	188	171	190	146
Indianapolis, Ind.	145	165	188	171	149	134
Los Angeles, Cal.	201	206	158	143	154	139
Philadelphia, Pa.	196	157	159	147	160	151
Pittsburgh, Pa.	176	155	218	143	126	153
Combined geometric mean	163	163	166	137	137	140

To pursue the possibility of a procedural bias further, measurements on NASN samples collected in Baltimore and Philadelphia were compared with independently measured samples collected with identical high-volume samplers, owned by the city, at the same locations on the same dates. It should be mentioned that the NASN 24-hour sampling periods often do not exactly coincide with the 24-hour sampling periods of the city-owned samplers at these two stations. The NASN starting times are scheduled randomly throughout the normal working hours, while the city-owned sampler in Philadelphia is routinely started at 10:00 A.M. In Baltimore, the city-owned sampler was started on the NASN schedule until the end of 1961. During 1962 and 1963 that sampler was routinely started between 8:30 and 9:00 A.M.

Differences in concentrations measured on NASN samplers and city-owned samplers in Baltimore and Philadelphia are analyzed in Tables 8 and 9, respectively.

The correlations between the paired samples from Baltimore are good, ranging from 0.7 to better than 0.95. There is no break in correlation in 1961, but the differences in averages in 1962 and 1963 are large enough to be of concern. This period coincides with the period of time that the city-owned sampler was operated on a fixed time schedule in contrast to the random NASN starting times used prior to 1962. The difference in starting times, which causes a disparity of up to 7 hours between the two sampling periods, would be expected to increase the variation among sample differences, but it would not be expected to bias sample differences more in one direction than another. No known changes in calibration or position of the samplers, or analysis of the samples has occurred that might produce the observed differences in 1962 and 1963.

The paired samples collected in Philadelphia correlate well. The coefficient is 0.85 or better for all years except 1962, when it dropped to 0.64. On the basis of only 25 observations this departure is not statistically significant. The largest difference between annual

Table 8. SUMMARY OF DIFFERENCES[a] BETWEEN PAIRED SAMPLES IN BALTIMORE, MD.

	1958	1959	1960	1961	1962	1963
January			-14	+50	-113	+ 3
			+ 3		-49	
		+24				
February			-14	-22	-96	-35
			0	+101		+ 1
March		+29	- 4	-22	-73	-108
			+35	7	-59	+40
April		+31	+10	-15	-70	-24
		+26		-18	-72	
		+15		-24		
May	-14	-81	+ 8	-27		-35
				-17		+81
June		+23	2	-12	+17	1
			2	4		-20
						-39
July	+33	+63	+ 1	+ 5	8	9
	-45	+23	+16	-25		+ 3
						4
August	+24		-61	-31	+ 5	-59
	+ 1		+ 3			-53
September	-17		+ 2	-28	+28	+18
	-20		4	-46	+ 9	
				-21		
October	-12		3	-16	+ 8	-36
	8		+47		-10	+ 9
November	-56		-12	-34	-19	-42
			+ 7			-47
December			+95	3	3	+ 5
			+29		+47	-59
Number of paired samples	10	9	22	21	17	23
Correlation coefficient	0.806	0.714	0.965	0.907	0.837	0.876
NASN avg, $\mu g/m^3$	110.4	153.0	146.6	151.3	135.0	141.4
City avg, $\mu g/m^3$	121.8	136.0	140.2	160.7	162.0	159.3
Avg diff[a] (d), $\mu g/m^3$	-11.4	+17.0	+6.4	-9.4	-27.0[b]	-17.9[b]
S_d	27.1	39.2	28.6	31.4	46.7	38.7

[a]NASN value minus city value, $\mu g/m^3$.

[b]Statistically significant.

averages was only 7.3 $\mu g/m^3$ in 1960. In 1959 the difference of 4.9 $\mu g/m^3$ was statistically significant, but only because the variability in the differences between paired samples was extremely low in that year. The difference is less than 3 percent of the average concentrations and not of concern from a practical standpoint.

On the basis of the preceding considerations, [i. e., (1) correlation with independent sampling in Baltimore and Philadelphia, (2) the unchanging nonurban mean, and (3) the relatively steady proportion of the organic constituent since 1959 and the limited information indicating

Table 9. SUMMARY OF DIFFERENCES[a] BETWEEN PAIRED SAMPLES IN PHILADELPHIA, PA.

	1958	1959	1960	1961	1962	1963
January	5	3	0	-13	-10	8
		3		-37	+ 2	
February	6	2	+ 4	-32	+10	-11
	-16	2	5	-78	+83	-14
		7	3			+14
March	+23	6	0	-66	5	+ 4
				-33		+18
				-15		
April	3	6	0	3	-113	1
	0	3	0	+11	-18	6
				4	5	
May	0	2	-55	-13	+63	+14
	+21	7	-39	-34	+23	-23
		7				
June	2	8	+ 2	-30	3	5
	5	-18	5	+21	+ 5	+11
July	1	6	-44	+14	+22	=16
	0	-12	+29	-23	+19	-24
	6					
August	2	6	-83	-38	+22	+ 8
	2		-11	-12	+58	+10
			+ 9		+ 3	2
September		5	9	+50	-12	+ 7
		+ 2	+ 4	+19	-22	
		0		+25		
October	-10	6	+33	+66	+54	+18
	-28		+46	+87	+18	+82
						-54
November	-14	4	-10	+48	-25	-46
	2	0	-14	-44	-17	-43
	2	0				
December	4	-12	-20	+10	-39	+ 8
	6	0	5	+ 2	+25	2
Number of paired samples	22	25	24	27	25	25
Correlation coefficient	0.991	0.998	0.928	0.853	0.644	0.873
NASN avg, $\mu g/m^3$	173.0	178.8	161.1	171.5	157.6	156.2
City avg, $\mu g/m^3$	176.2	183.7	168.4	176.0	152.1	158.6
Avg diff[a] (d), $\mu g/m^3$	-3.2	-4.9[b]	-7.3	-4.5	+5.5	-2.4
S_d	10.4	4.3	27.4	38.6	38.6	26.4

[a]NASN value minus city value, $\mu g/m^3$.

[b]Statistically significant.

comparable constancy in the proportions of nitrates, sulfates, and iron], it is concluded that the sampling and analytical procedures have been, and continue to be, reliable and that the indicated drop in particulate concentrations in 1961 is not spurious.

STATION ENVIRONMENTS

Having established the statistical significance of the indicated drop in suspended particulates and having discounted the likelihood that the drop is due to changes in analytical procedure, we sought for explanations in terms of the environments of the sampling stations. No geographical pattern is apparent among the stations that reported distinct drops in level in 1961; the stations are scattered among all regions of the country except the central east coast.* Nevertheless, local meteorology comes to mind as a likely factor that could exert a strong influence. A basic change in the meteorology of a number of cities, or even a change in the typical weather on randomly selected sampling days, could affect the particulate levels. L. E. Niemeyer, Meteorological Section, Laboratory of Engineering and Physical Sciences, evaluated meteorological factors for the 10 NASN stations† for which data showed the largest variations in particulate concentrations between the years 1959 and 1961. He found that most of the sample values in the upper and lower 10 percent of the concentration range were associated with meteorological conditions that hindered or enhanced the dispersion of pollution. The annual station means were then recalculated, excluding these extreme values. Mr. Niemeyer reported (complete text is given in Appendix C), ". . . it is felt that by the elimination of the extreme samples in which meteorological factors have a definite bearing, the remaining samples would more nearly approximate the true annual mean air quality under 'average' conditions." He found, however, that the generally decreasing trend through 1961 was still apparent in the adjusted data of the 10 stations selected.

All reasonable precautions are taken to eliminate bias from the selection of sampling dates; the distribution of daily weather conditions cannot be anticipated, however, and it is conceivable that the sampling days at a given station could coincide with an abnormal number of good dispersion days in a given year, resulting in an erroneous indication of reduced pollution levels. Mr. Niemeyer comments, ". . . if by chance the NASN had sampled more frequently on days favoring (good) dispersion in 1961, studies of the frequency of occurrence of meteorological elements such as the average wind speed would have indicated a significant difference in the observed frequencies." The implication is that no such disparities were discovered. On the basis of these investigations it was concluded that neither large-scale nor local meteorological factors exerted a major influence on the decline in mean particulate concentrations in these 10 cities.

As evidence that the 26 randomly selected NASN sampling dates do provide a reliable estimate of the average annual concentration of suspended particulates in spite of uncontrollable variables such as

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* Region III: Kentucky, Maryland, North Carolina, Virginia, West Virginia.

† Atlanta, Ga.; Chicago, Ill.; Houston, Tex.; Los Angeles, Calif.; Nashville, Tenn.; New Orleans, La.; Philadelphia, Pa.; Pittsburgh, Pa.; St. Louis, Mo.; Salt Lake City, Utah.

meteorology, Table 10 lists the annual means predicted by the NASN and the actual means determined by almost daily sampling with the city-owned sampler in Philadelphia.

Table 10. COMPARISON OF ESTIMATED MEANS FROM RANDOM SAMPLES WITH TRUE MEAN OF DAILY SAMPLES IN PHILADELPHIA, PA.

	City samples		NASN samples		
	N	$\tilde{\mu}$	N	$\bar{x}$	± 90% conf. limits
1960	342	157 $\mu g/m^3$	25	147	± 20 $\mu g/m^3$
1961	361	155 $\mu g/m^3$	26	160	± 20 $\mu g/m^3$
1962	358	145 $\mu g/m^3$	25	151	± 15 $\mu g/m^3$

The decision was made to conduct a telephone survey of the 64 'every-year' stations, in search of environmental changes that would correlate with observed changes in particulate concentrations. Each conversation was initiated with a neutral inquiry regarding any significant changes in the neighborhood of the sampling site, or in the city as a whole, that might have affected the levels of suspended particulate. This query was intended to elicit responses that were not biased in favor of only those events that would be expected to contribute to the observed trend in concentrations. It was determined, however, that certain specific subjects should be covered in each conversation; therefore if they did not arise spontaneously in response to the general inquiry, the following specific questions were posed:

(1) "Have there been any major demolition or construction projects, possibly associated with urban renewal or expressway projects in the community, during this 7-year period?"

Urban renewal, city core rehabilitation, and similar projects are considered potential sources of particulates, especially where demolition debris is burned on site. The influence of such projects on particulate concentrations depends in part on the rapidity with which the several phases follow each other. If the acquisition of property precedes the actual demolition by many months, the vacated plants and residences no longer contribute their accustomed share of the pollution burden, and concentrations may temporarily go down. The extent to which the demolition phase raises the observed level again can depend on the proportions of masonry and frame construction, the presence or absence of ordinances regulating open burning and control of dust at demolition sites, and the degree to which these ordinances are enforced. Finally, the observed influence depends on the locations of the project area and the sampling site relative to the prevailing wind direction.

(2) "Has there been any significant change in the proportions of coal, oil, and natural gas consumed in the community, and have there been any conversions among major individual fuel users in the vicinity of the sampling site?"

In the United States as a whole a slow trend away from coal for industrial and for space-heating purposes occurred during the 7-year period being considered here, 1957 to 1963. In spite of this trend, the total consumption of coal has remained close to 400 million tons as a result of increasing consumption by the electric utilities (Figure 2).

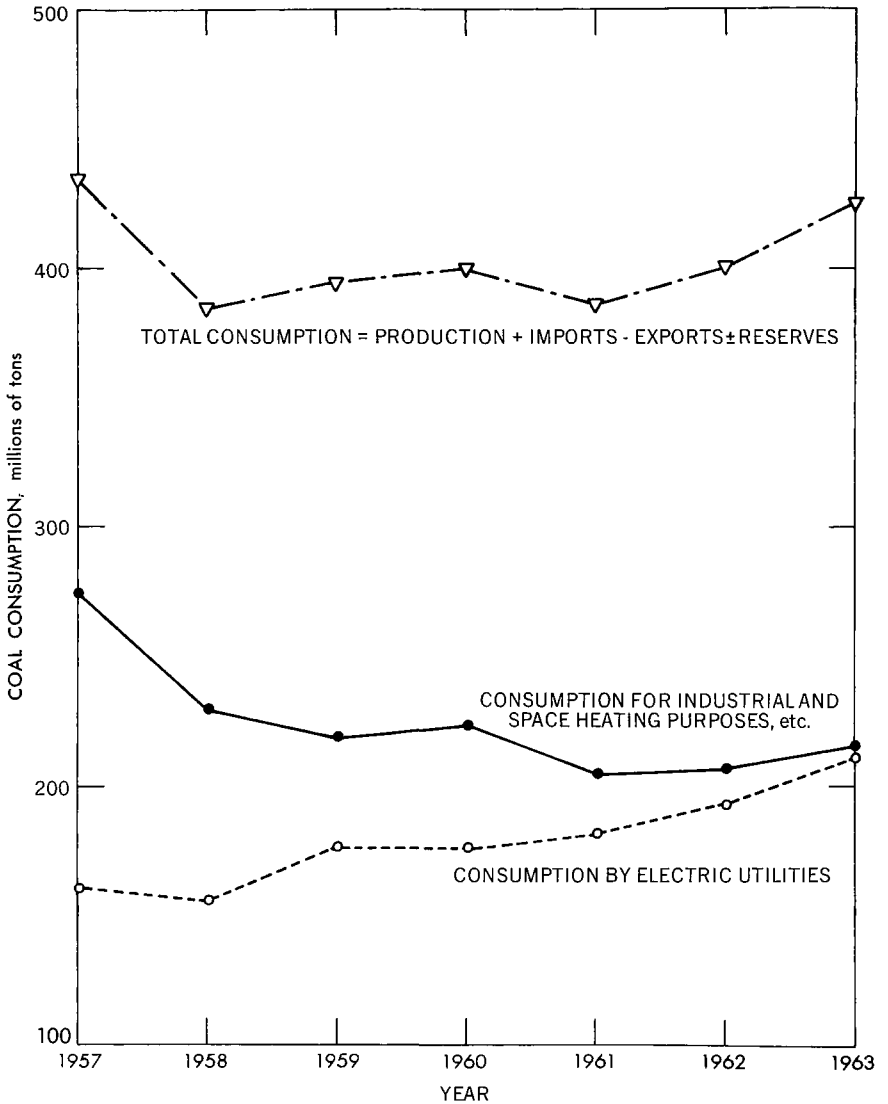

Figure 2. Consumption of bituminous, lignite, and anthracite coal in the United States.

In 1957, some 37 percent of the coal used went to the electric utilities. By 1963 the share consumed by electric utilities had risen to 50 percent.* The significance of this is that an increasing proportion of this country's annual coal production is being burned in large, relatively efficiently operated electric power plants that are often equipped with effective fly ash collectors and, for an equivalent amount of fuel, emit less particulate pollution than the smaller units in which the coal was formerly burned. Furthermore, the location of an electric utility is often more remote from the urban center than the former users. As a result, somewhat less of the potential particulate pollution inherent in those 400 million tons of coal is being dispersed in urban atmospheres. In communities where several large industries and/or numerous homeowners have switched from coal to gas, a distinct downturn in particulate concentrations could result.

(3) "Have there been any significant changes in sources in the vicinity of the sampler through installation of control equipment, start up of new units, shutdown of old units, or changed methods of operation that would be expected to influence particulate levels?"

Many cities have some form of smoke control ordinance that gives their administrators some power to require the abatement of sources, both large and small, that individually or collectively emit objectionable amounts of particulate pollution. In many people's minds air pollution is identified chiefly with this visible smoke and tangible grit. Therefore, in those communities that have achieved progress against air pollution, it has been accomplished largely on that fraction of total air pollution that the NASN sampler measures, the suspended particulates. These local control agencies frequently have the power of review and approval over new installations of boiler and process equipment; thus they exert an influence on decisions in the early planning stages leading to the installation of "cleaner" operating equipment in new or expanded industrial facilities. Sometimes sources are eliminated as the consequence of the urban renewal or expressway projects previously discussed.

In addition to these specific questions concerning the sampler's environment, the location of the station was checked, the dates of any changes in location confirmed, and the possible effects on particulate concentrations as a result of such changes assessed. A request was also made for data on any supplementary air quality measurements taken locally, such as settled dust, AISI spot tapes, etc. The survey questionnaire is reproduced in Appendix D.

On the basis of responses to the telephone survey, the stations were grouped into categories according to the most prominent changing factor, if any, in their environments during the 1960-1961 period. A decrease in the mean level of suspended particulates was observed for

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*"1963 Minerals Year Book, Vol. II-Fuels" U.S. Dept. of the Interior, Bureau of Mines.

42 of the 64 'every-year' stations. These stations were grouped according to whether the percent change in annual mean had (a) greater than 95 percent probability of being significant, (b) between 90 and 95 percent probability of being significant, (c) less than 90 percent probability of being a significant change.

Of the total of the negative changes, almost one third (30.1%) is accounted for by 23 stations at which the percent change has less than 90 percent probability of being significant (Table 11a), and this portion is attributed to normal random variation. The percent change at nine stations is in a borderline area with 90 to 95 percent probability of significance. These stations contributed 27.6 percent to the total of the negative changes. The remaining ten stations, for which the reported decreases have better than 95 percent probability of being significant, contributed a disproportionate 42.3 percent to the total of the negative changes. These groups are further classified in Table 11a according to the categories of stations near which changes are known to have occurred that conceivably could have contributed to the observed decrease, and those near which no changes are known to have occurred. Of the 23 stations at which the percent change is felt to be not significant, only three were in environments where possible influencing changes had occurred and the contribution of these three to the total was less than 5 percent. In the 90 to 95 percent probability group, three stations, near which possible influencing changes occurred, account for more than a third of this group's contribution to the total. In the third group, where the decreases have better than 95 percent probability of being significant, plausibly related changes were reported in the vicinity of five stations, which together account for half of the contribution from this group. For the 19 stations at which the probability of significant change is more than 90 percent, known changes in contributing influences occurred in the vicinity of 8 stations, which together account for one third of the combined contributions from the 42 stations reporting decreases.

Table 11a. SUMMARY OF 'EVERY-YEAR' STATIONS REPORTING DECREASES in 1961 and POSSIBLE RELATED CHANGES in ENVIRONMENTAL INFLUENCES

Environmental influences	Level of significance		Total of significant contributions	Not significant $p > 0.10$
	$p < 0.05$	$0.05 < p < 0.10$		
Known changes-subtotal	22.4%(5)[a]	10.3%(3)	32.7%(8)	4.7%(3)
Cessation of demolition and/or construction	3.7%(1)	3.2%(1)	6.9%(2)	4.0%(2)
Control and/or elimination of sources	14.9%(3)	7.1%(2)	22.0%(5)	0.7%(1)
Fuel conversion	3.8%(1)		3.8%(1)	
No known changes-subtotal	19.9%(5)	17.3%(6)	37.2%(11)	25.4%(20)
TOTAL			69.9%(19)	30.1%(23)

[a]Values in parentheses indicate number of stations involved.

In contrast, of the 20 stations that reported an increase in annual mean concentration of suspended particulate samples in 1961 over 1960, only one has an increase large enough to be in the borderline area in which the percent change has 90 to 95 percent probability of being significant (Table 11b). No changing influences that might have caused an increase at that station were known. Demolition or construction was reported in the vicinity of two stations, but since the increases in particulates at these two stations were not significant, no relationship can be hypothesized.

Table 11b. SUMMARY OF 'EVERY-YEAR' STATIONS REPORTING INCREASES in 1961 and POSSIBLE RELATED CHANGES in ENVIRONMENTAL INFLUENCES

Environmental influences	Level of significance		Total of significant contributions	Not significant
	$p < 0.05$	$0.05 < p < 0.10$		$p > 0.10$
Known changes-subtotal				29.1%(4)[a]
Demolition and/or construction				16.9%(2)
Control and/or elimination of sources				12.2%(2)
Fuel conversions				
No known changes-subtotal		6.3%(1)	6.3%(1)	65.6%(15)
TOTAL			6.3%(1)	93.7%(19)

[a]Values in parentheses indicate number of stations involved.

Levels increased at two stations located in cities where it was reported that control or elimination of sources was being effected, but again the increases are not statistically significant and may be attributed to the variability inherent in the data.

The preceding analysis and discussion of the changes in mean concentration of suspended particulate samples at the individual 'every-year' stations indicates that there was good reason for the significant net decrease in the combined mean of the 'every-year' group in 1961. Half of the significant negative contribution can be related to the probable influence of changing factors in the neighborhood of eight sampling stations. At four other stations, although the decrease appeared significant, the improvement was only temporary, since increases of comparable magnitude occurred in their annual means in the next year or two.

Three examples illustrate some of the environmental factors that appear to have influenced the decreasing particulate concentrations as measured by an NASN sampler.

Boise, Idaho. The annual mean of the suspended particulate samples at the Boise station averaged 109 $\mu g/m^3$ between 1957 and 1960. In 1960 a gas pipeline to the city was completed and the conversion of many commerical, coal-burning units to gas began immediately.

From 1961 through 1963 the annual mean has averaged 82 $\mu g/m^3$, with the possible indication of a slow downward trend consistent with increasing conversion of fuel-burning units to gas.

Rochester, N. Y. The annual mean concentration of suspended particulates in Rochester rose from 107 $\mu g/m^3$ in 1958 to 141 $\mu g/m^3$ in 1960, apparently in direct response to the demolition, clearing, and grading activities associated with the construction of an expressway directly adjacent to the building on which the sampler is located. In 1961, as the focus of these activities moved on, the annual mean dropped to 106 $\mu g/m^3$. Results for 1962 and 1963 indicate that the annual mean is now stabilized at around 85 $\mu g/m^3$. Not only has the construction activity moved out of the sphere of influence, but those neighborhood sources in the path of the expressway have been eliminated. Cessation of demolition and construction was judged to be the major factor contributing to the reduction of the 1961 annual mean at the Rochester station, but with the passage of this transient perturbation the equilibrium concentration now seems to be some 15 percent lower than before. If the analysis is correct, the strip of expressway traffic contributes less in the way of suspended particulates to the neighborhood than did the replaced strip of small industries and residential heating units.

Des Moines, Iowa. The following description is taken from the excellent written report by Sherry Robinson, Chief of Environmental Sanitation for the City of Des Moines. The annual mean particulate concentration averaged about 155 $\mu g/m^3$ from 1957 through 1960. In 1961, the annual mean dropped to 119 $\mu g/m^3$ and available data indicate a slow, continuing decline in 1962 and 1963. Coincident with the drop in 1961, the following events took place:

1. The burning of 300 to 400 tires daily about 1/2 mile southwest of the sampler location was discontinued.

2. Electrostatic precipitators were installed at a cement plant 3-1/2 miles southwest, up the Racoon River. The precipitators retrieve some 40 tons of cement dust daily.

3. The demolition of some 30 buildings in the downtown area, within four to eight blocks of the sampler site, was completed in 1961.

4. In an industrial area from two to ten blocks south of the sampler site numerous coal-burning units have been converted to gas in the last 3 years (1961-1963). This continuing shift in the proportions of major fuels consumed in the area may be responsible for the gradual decline, which appears to be continuing in 1962 and 1963.

5. In an area north of the sampling site, total demolition and clearance in 1961 produced considerable dust but also resulted in the elimination of many antiquated home-heating units, which had burned coal.

The net effect of these events in Des Moines was to lower the concentrations of suspended particulates measured at the NASN sampler

site. For the purpose of assigning Des Moines to a category in Table 11a the principal cause was judged to be the control of the two sources listed in items 1 and 2.

These three examples are among the more definitive; specific events are logically related to the observed changes in particulate concentrations. These situations show that the high-volume air sampler is responsive to changes in those factors in the environment that are related to particulate pollution. The detailed appraisal of circumstances in Des Moines emphasizes the hazards inherent in ascribing an observed change in particulates to a single cause.

In one instance, the changes described in the city conflicted with the observed change in annual concentration measured at the NASN station. Although the increase was not statistically significant because of the wide variability in sample values, the mean concentration in 1961 at the Charleston, W. Va., station was nevertheless some 27 percent higher than the 1960 value. The increase was not sustained, however. In 1962 the mean concentration dropped again by a comparable amount.

In some cases where environmental changes were reported but a negligible change in particulate concentration was measured, and in some cases where, conversely, a significant change in particulates occurred for no known reason, a more detailed survey and analysis of the area encompassed by the sampler's sphere of perception would probably reveal a consistent explanation. Some contributing factors may not be recognized or fully appreciated because individually they seem inconsequential, yet in combination they may produce a measurable net effect.

SUMMARY

It has been established that the total change in the mean sample concentration for the 'every-year' group of NASN stations over the period 1957 through 1963 is statistically significant and can be related in part to specific changes in individual or collective particulate sources. The major decreases in the annual mean occurred in 1959 and 1961. The increase in 1964 is also significant and, if continued, will be suggestive of a cyclic fluctuation. These changes in mean concentration are statistically significant since they are greater than the criterion of the 'least-significant-difference' which is equal to 3.6 percent ($p = 0.05$). Conceivably, as the temporary perturbations in the environment resulting from expressway and urban renewal projects taper off and as the possibilities for fuel conversions are exhausted, the proliferation of sources attending the underlying growth of many urban habitats will reassert its influence and start the concentrations of particulate pollution climbing again.

APPENDIX A: 'EVERY-YEAR' NASN STATIONS and SUMMARY OF RESPONSES TO TELEPHONE INQUIRY ABOUT CHANGING ENVIRONMENTAL INFLUENCES

	Changing Environmental Influences				
	Demolition and const.	Cessation of dem. and const.	Source abatement	Fuel conversion	No known changes
Birmingham, Ala.					X
Phoenix, Ariz.					X
Little Rock, Ark.					X
Los Angeles, Cal.					X
San Diego, Cal.					X
San Francisco, Cal.			X		
Denver, Colo.					X
Hartford, Conn.					X
New Haven, Conn.					X
Wilmington, Del.					X
Washington, D. C.		X			
Tampa, Fla.					X
Atlanta, Ga.					X
Boise, Ida.				X	
Chicago, Ill.			X		
E. Chicago, Ind.					X
Indianapolis, Ind.					X
Des Moines, Ia.			X		
Wichita, Kan.					X
New Orleans, La.					X
Portland, Me.					X
Baltimore, Md.					X
Boston, Mass.					X
Worcester, Mass.			X		
Detroit, Mich.		X			

Appendix A: (Continued)

	Changing Environmental Influences				
	Demolition and const.	Cessation of dem. and const.	Source abatement	Fuel conversion	No known changes
Minneapolis, Minn.			X		
Jackson, Miss.					X
Kansas City, Mo.					X
St. Louis, Mo.		X			
Helena, Mont.					X
Omaha, Neb.					X
Las Vegas, Nev.	X				
Newark, N. J.					X
Albuquerque, N. M.					X
Binghamton, N. Y.					X
New York City, N. Y.					X
Rochester, N. Y.		X			
Syracuse, N. Y.					X
Utica, N. Y.					X
Charlotte, N. C.					X
Bismarck, N. D.					X
Cincinnati, Ohio					X
Cleveland, Ohio					X
Columbus, Ohio					X
Dayton, Ohio					X
Youngstown, Ohio					X
Portland, Ore.	X				
Philadelphia, Pa.					X
Pittsburgh, Pa.					X
Providence, R. I.					X
Columbia, S. C.			X		
Sioux Falls, S. D.					X
Chattanooga, Tenn.					X

GPO 821—754—4

Appendix A: (Continued)

	Changing Environmental Influences				
	Demolition and const.	Cessation of dem. and const.	Source abatement	Fuel conversion	No known changes
Nashville, Tenn.					X
Dallas, Tex.					X
Houston, Tex.					X
San Antonio, Tex.			X		
Salt Lake City, U.					X
Burlington, Vt.					X
Norfolk, Va.					X
Seattle, Wash.					X
Charleston, W. Va.			X		
Milwaukee, Wisc.					X
Cheyenne, Wyo.					X

APPENDIX B: NONURBAN 'EVERY-YEAR' STATIONS

Grand Canyon Pk., Ariz.
Montgomery Co., Ark.
Montezuma Co., Colo.
Florida Keys, Fla.
Butte Co., Idaho
Parke Co., Ind.
Delaware Co., Iowa
Acadia Nat. Pk., Me.
Calvert Co., Md.
Jackson Co., Miss.
Shannon Co., Mo.
Glacier Nat. Pk., Mont.
Thomas Co., Neb.
White Pine Co., Nev.
Cape Hatteras, N. C.
Ward Co., N. D.
Clarion Co., Pa.
Washington Co., R. I.
Richland Co., S. C.
Shenandoah Nat. Pk., Va.

APPENDIX C: MEMORANDUM CONCERNING METEOROLOGICAL FACTORS

OPTIONAL FORM NO. 10 5010-107
MAY 1962 EDITION
GSA GEN. REG. NO. 27

UNITED STATES GOVERNMENT

Memorandum

TO Chief, LEPS DATE: August 13, 1964
Through: Chief, Meteorology Section, LEPS

FROM L. E. Niemeyer, Meteorology Section, LEPS

SUBJECT: NASN Values, 1960-1

Further investigation of the decrease in annual mean total particulate values of the every year urban stations of the NASN has not revealed any significant anomalies in weather patterns during the subject period. Of course, weather variations from "climatic normals" have occurred within the period in various sections of the nation, but whether or not the variations contributed to the decrease in mean particulate values in 1961 is difficult to determine.

When early pilot studies in this investigation did not reveal any marked change in meteorological parameters, we confined our efforts to the approach suggested in the referenced memorandum. Our thought was that if we could positively eliminate those cases from consideration which were directly attributable to meteorological influences, then those remaining might show whether or not there was an outside influence. In fact it was suspected that when the extreme cases were eliminated there would be no significant differences in the air quality means of the stations selected for investigation. Unfortunately this has not been the case.

Ten NASN stations were selected which showed the largest variation in air quality values between the years 1959 and 1961. After some pilot work, arbitrary limits were selected at the upper decile and the lower decile values for the individual stations and meteorological information was compiled for the NASN samples which fell into these deciles. Meteorological data for the selected deciles was divided into two classes. One class was defined using meteorological conditions favorable for dispersion of air pollution and the other class was defined for meteorological conditions which favored limited dispersion. After the data were so classified they were compared to the air quality samples. This comparison revealed that 86% of the samples were found in the proper category. That is if the meteorological conditions indicated that limited dispersion was favored then it was found that the air quality value fell in the upper decile. It is probable that some of the remaining 14% could be explained if wind direction were taken into account.

That is to say in some cases it is conceivable that a sample may be either high or low depending on the location of the sampler with respect to a near-by source and the given wind direction. Also one or two samples which fell into the selected decile categories were obviously erroneous.

See Note For example, the sample collected on the 13th of May 1960 at Detroit gives a value of 5.9 micrograms per cubic meter for the total particulate and 6.2 micrograms per cubic meter for the organic fraction!

Since an assignable cause was found for the samples in the upper and lower decile, we recomputed the annual arithmetic mean for these stations and found that the average range decreased from 52 micrograms per cubic meter to 18 micrograms per cubic meter. (Graphs attached.) It was also noted that the average range for individual stations was cut approximately in half while Pittsburgh's range dropped from 139 $\mu g/m^3$ to 6 $\mu g/m^3$. The generally decreasing trend through 1961 however, is still found in the adjusted data of the ten stations selected. It is my opinion, on the basis, of the work described above that the trend noted in the data is not due solely to meteorology and that some other factor has contributed. It is also felt that if by chance the NASN had sampled more frequently on days favoring limited dispersion in 1959 and on days favoring dispersion in 1961, studies of the frequency of occurrence of meteorological elements such as the average wind speed would have indicated a significant difference in the observed frequencies.

Further it is felt that by the elimination of extreme samples in which meteorological factors have a definite bearing on the concentrations measured the remaining samples would more nearly approximate the true annual mean air quality under "average" conditions.

It is my recommendation that the meteorological phase of the investigation to determine the cause for a decrease in the mean annual total particulate values of the NASN be concluded. While more exhaustive meteorological studies are possible, I do not think they would be sufficiently fruitful to warrant the manpower required to complete them.

The initiation of synoptic sampling by the NASN on the same day will be of great assistance to future meteorological investigations of this sort. It would also greatly assist future meteorological investigations if the NASN sampling day would be made to coincide with the calendar day since meteorological data are collected on this basis.

Note: Tabulation was misread. Listed value is 59 $\mu g/m^3$.

APPENDIX D: TELEPHONE SURVEY QUESTIONNAIRE

NASN 'Every Year' Station Survey Date: _____

City:_____ Phone:_____

Person called:_____ Title:_____

Person reached:_____

_____

_____

1. Demolition, construction, urban renewal or freeway projects in the vicinity of the sampler:_____

_____

_____

2. Change in fuel used, either city in general or specific buildings or plants in the immediate vicinity of the sampler:_____

_____

_____

3. Control, change in operation or shut down of large nearby sources:

_____

_____

4. Check Station Location:_____

_____

5. Supplementary data - settled dust, C.O.H (AISI), hi-vol:

_____

_____

BIBLIOGRAPHIC: McMullen, T. B. and R. Smith. The trend of suspended particulates in urban air: 1957-1964. PHS Publ. No. 999-AP-19. 1965. 27 pp.

ABSTRACT: The trend toward lower average concentrations of suspended particulate pollution in the urban air in the United States from 1957 through 1963, indicated by measurements of the National Air Sampling Network, is investigated from three aspects: (1) statistical significance, (2) the validity of the sampling and analytical methods, and (3) relationship to environmental changes in the vicinity of the sampling stations. The evidence indicates that the decline is real and that the methods employed can reliably detect substantive changes in the concentration of suspended particulate pollution. Information on changes in the environments of a representative group of stations revealed that where significant decreases in suspended particulates occurred, they were often logically related to specific changes in source emissions. Thus the indicated decline in the suspended particulate portion of urban air pollution is verified and substantially accounted for. A statistically significant increase in the annual mean occurred in 1964; if continued, such an increase would suggest the possibility of cyclic influences.

ACCESSION NO.

KEY WORDS:
Suspended Particulate Air Pollution
Trends
Environmental Changes
Case Histories

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