

Air



Digest of Ambient Particulate Analysis and Assessment Methods

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by

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ABBREVIATIONS FOR CHEMICAL ELEMENTS

Abbreviation	Chemical element	Abbreviation	Chemical element
Ac	actinium	Nd	neodymium
Al	aluminum	Ne	neon
Sb	antimony	Ni	nickel
Ar	argon	Nb	niobium
As	arsenic	N	nitrogen
At	astatine	Os	osmium
Ba	barium	O	oxygen
Be	beryllium	Pd	palladium
Bi	bismuth	P	phosphorus
B	boron	Pt	platinum
Br	bromine	Po	polonium
Cd	cadmium	K	potassium
Ca	calcium	Pr	praseodymium
C	carbon	Pm	promethium
Ce	cerium	Pa	protactinium
Cs	cesium	Ra	radium
Cl	chlorine	Rn	radon
Cr	chromium	Re	rehmium
Co	cobalt	Rh	rhodium
Cu	copper	Rb	rubidium
Dy	dysprosium	Ru	ruthenium
Er	erbium	Sm	samarium
Eu	europium	Sc	scandium
F	fluorine	Se	selenium
Fr	francium	Si	silicon
Gd	gadolinium	Ag	silver
Ga	gallium	Na	sodium
Ge	germanium	Sr	strontium
Au	gold	S	sulfur
Hf	hafnium	Ta	tantalum
He	helium	Tc	technetium
Ho	holmium	Te	tellurium
H	hydrogen	Tb	terbium
In	indium	Tl	thallium
I	iodine	Th	thorium
Ir	iridium	Tm	thulium
Fe	iron	Sn	tin
Kr	krypton	Ti	titanium
La	lanthanum	W	tungsten
Pb	lead	U	uranium
Li	lithium	V	vanadium
Lu	lutetium	Xe	xenon
Mg	magnesium	Yb	ytterbium
Mn	manganese	Y	yttrium
Hg	mercury	Zn	zinc
Mo	molybdenum	Zr	zirconium

1.0 INTRODUCTION

The U.S. Environmental Protection Agency (EPA), state and local air pollution control agencies, and independent research scientists have used a wide variety of techniques for analyzing and evaluating samples and data related to particulate matter present in urban and rural atmospheres. Although descriptions and examples of those techniques are available in the academic and professional literature, it is difficult for a potential user to learn about more than one technique at a time. As a result, he may apply one technique to a given problem, not knowing that another technique is more appropriate.

There is a need, then, for a compendium of techniques that have been or can be used to analyze a suspended particulate problem. Such a compendium should provide a brief and clear description of each available technique, provide references which will enable the user to obtain more information concerning the technique and its application to air pollution control problems, indicate the types of problems to which the technique can apply, explain ways in which the various techniques can interrelate, estimate the resources needed to implement each technique, and provide a good, clear example of how the technique has been applied in the literature. This is one set of purposes that will be met by this digest.

A second purpose of this digest is to provide the user with guidelines as to the use of these techniques in combination in order to resolve a specific type of problem. In order to do this, information is presented which summarizes and compares available techniques in terms of their problem applicability, resource requirements, and interrelationships.

Three general types of problems are addressed: characterizing the aerosol in terms of spatial extent, temporal patterns, and composition; qualitatively identifying the source categories (e.g., fuel combustion) that produce significant total suspended particulate (TSP); and quantitatively defining the TSP impact of specific sources (e.g., a large coal-fired power plant).

Resource requirements are assessed in terms of manpower, skill, computer access, and data. Manpower requirements are estimated for each individual technique and are assigned descriptors of low, moderate, and high. These estimates do not include the time required to produce the raw field or laboratory data. Rather they pertain only to the time required to manipulate, process, keypunch, and/or interpret the available data. Computer requirements are specified in terms of whether a computer is not necessary, optional, or necessary. The reader should be aware of the interdependence of these two categories. Skill requirements are described as being low, moderate, or high. Lastly, the types of data required to implement each technique are identified. It is assumed that the reader is aware of the different types of instrumentation or laboratory methods that will produce the data indicated.

Following the discussion of resources is a brief overview of the ways in which the subject technique relates to the other techniques presented in this digest. Three types of relationships are identified: combinable, meaning that the subject technique can be combined with another one to produce what is in effect a third technique; parallel, meaning the techniques provide two separate ways of answering the same question; or dependent, meaning the subject technique cannot be used without prior use of another one. In one way or another, each of the techniques discussed holds a parallel relationship with the others presented in this digest.

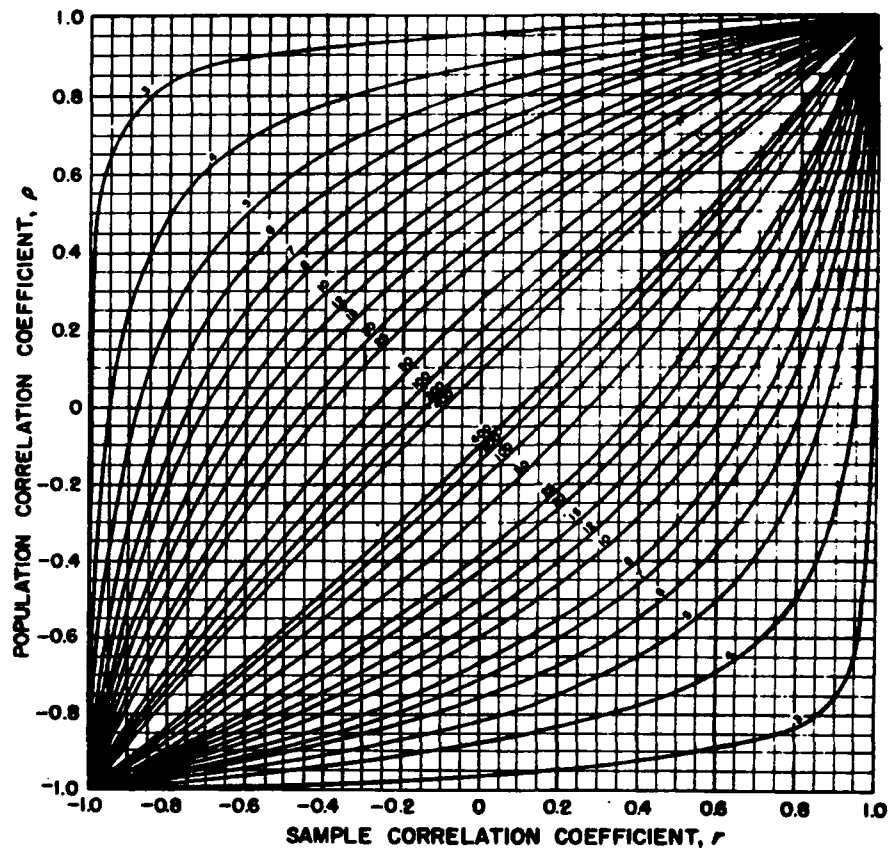
The following 26 individual techniques, disaggregated into six classes, are discussed in Chapters 2.0 through 7.0:

- Techniques assessing the temporal patterns of TSP concentrations
 - Long-term trends
 - Seasonal and monthly patterns
 - Weekday versus weekend and daily patterns
 - Diurnal variations
 - Emission pattern/air quality relationships
- Techniques assessing the spatial patterns of TSP concentrations
 - Concentration versus site type
 - Pollution, gradient, and dosage rose
 - Intersite correlations
 - Upwind/downwind
- Techniques assessing the effect of meteorological variables
 - Regression and correlation analyses
 - Decision tree
 - Precipitation
 - Wind speed
 - Trajectory analysis
- Emissions data analysis techniques
 - Emission inventorying
 - Microinventorying
 - Diffusion modeling
- Techniques which interpret chemical, elemental, and morphological data
 - Temporal, spatial, and meteorologically-affected patterns
 - Enrichment factor
 - Chemical element balance
 - Interspecies correlations
 - Pattern recognition
 - Factor analysis
 - Microscopy
- Techniques which interpret particle sizing data
 - Frequency distribution
 - Species-specific analysis

Although the techniques presented were selected following a comprehensive overview of much of the air pollution control literature, it should not be concluded that this digest is totally inclusive. To the contrary, it is stressed that there are other techniques which may be available (e.g., intervention analysis, population exposure analyses, Q-Q plots, box plots, and graphical enhancement of scatter plots). The authors have simply tried to identify those techniques which have been used most commonly or which hold good potential. Similarly, the reader

should not conclude that all possible interrelationships have been pursued. The reader is encouraged to use his imagination to create new and potentially rewarding combinations or interactions of techniques.

While imaginative combinations are urged, the user is also cautioned to be aware of two potential complications associated with the use of these techniques. First, these techniques are data manipulative and they assume that the data themselves are valid. The user should be aware that some concern has been expressed regarding the accuracy of the high volume sampler data itself. Factors such as static deposition, wind directionality, and artifact formation may tend to produce errors in measured concentrations and may cause the analyst to make erroneous conclusions concerning the nature and extent of his particulate problem. This subject is under active investigation by EPA. The user should also investigate the data base to assess the degree to which standard quality assurance procedures (such as those described in EPA's Guidelines for Development of a Quality Assurance Program series) and standard instrument siting procedures are followed.^{1,2} Second, many of the techniques involve the application of standard statistical measures such as linear regression, correlation analyses, analysis of variance, stepwise linear regression, multiple regression, and so on. The user should be alert to the fact that misuse of these techniques can lead to erroneous interpretation of available data. For example, one may find (as in some of the examples provided in the following chapters) that a data set has a correlation coefficient (r) of +0.8. To understand the statistical significance of this correlation, however, it is equally important to know the number of samples upon which that correlation is based. As Figure 1-1 indicates, a sample size of 5 indicates a true r of -0.20 to +0.96 at the 95 percent confidence level, whereas a sample size of 50 indicates a true r of +0.70 to +0.87. The user is urged to refer to standard statistical texts prior to applying these techniques.^{3,4,5,6,7}



Use explained in Reference 4. The numbers on the curves indicate sample size for the case of a two-variable linear regression.

Figure 1-1. Ninety-five percent confidence belts for correlation coefficient.

Source: Reference 4

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2.0 ANALYZING TEMPORAL PATTERNS OF SUSPENDED PARTICULATES

Analyzing temporal patterns of TSP concentrations is a simple and moderately effective means of initiating a comprehensive study of the causes, sources, and severity of a suspended particulate air pollution problem. This chapter discusses five techniques which have been commonly used to assess those trends and patterns:

- Long-term trends
- Seasonal and monthly patterns
- Weekday/weekend and daily patterns
- Diurnal patterns
- Emission pattern/air quality relationships

The analyst should exercise some caution when attempting to apply these techniques. He should, for example, ensure that the data are being compared for identical locations. Likewise he should ensure that equivalent measurement procedures, sampler configurations, and quality assurance checks have been employed.

Once he has determined that a trend analysis is indeed desirable, the analyst should consider which statistical method he will want to use. Some of the most commonly used methods, their nature, uses, and limitations, are summarized in Table 2-1.

2.1 LONG-TERM TRENDS

2.1.1 Description of Technique

This technique is retrospective in nature in that it describes the historical record of air quality measurements obtained from single sites or a regional area. It is best used to describe past data, but trend lines can be used (with some caution) to forecast future concentrations.

Table 2-1. STATISTICAL METHODS USEFUL IN ANALYZING TEMPORAL PATTERNS

Method	Description	Uses	Limitations
Graphical	Plot of TSP concentrations measured over a specified time period	Visual interpretation of data	May produce a plot difficult to interpret
Moving average	Obtained by replacing each value in a series of equally spaced data by the mean of itself and a standard number of values directly preceding it	Aids in determining more objective trend line by providing a smoother plot of the original data	
7 Whittaker-Henderson formula	Mathematical equation which yields a smoothed curve of TSP concentration vs time	Useful in determining an objective trend curve for any time period	Generally requires a computer
Daniel's test for trends using Spearman rank correlation	Comparison of a test statistic coefficient with normally distributed table values for significance; the data are ranked in ascending order when obtaining the test statistic	Useful in classifying a pattern as upward/downward and indicates consistency of pattern by the level of significance	Not very powerful when using a small sample size (<8 observations)

Table 2-1 (continued). STATISTICAL METHODS USEFUL IN ANALYZING TEMPORAL PATTERNS

Method	Description	Uses	Limitations
Parametric correlation	Comparison of a test statistic with percentile of Students' t statistics for significance	Useful in classifying a trend as upward, downward, or no change	Must assume data are of a normal or log-normal distribution
Regression analysis	Estimation of a linear or exponential model describing a set of data and an estimate of the level of significance	To produce estimates of the constant rate of absolute or percent change over time	Can produce a standard error of hypothesis testing
Chi-square analysis	Comparison of the percent of observation above a given level between two time periods; comparison is between the test statistic and the quantiles of a chi-square random distribution	Useful test for a change in extreme values or short-term statistics	Minimum of five observations for each time period above and below the comparison level; observations must be independent
Comparison of means using F-distribution	Testing of the hypothesis that there is no difference between observed means	Useful for distinguishing an actual difference between means and attaching a level of significance to results	Can produce a standard error of hypothesis testing

Source: References 3, 8, 9

Examination of long-term trends is most easily accomplished by using simple graphical methods. In doing so, a variety of averaging times may be used. Usually, plotting quarterly or annual means over time will be sufficient to depict the basic temporal pattern. Other parameters sometimes plotted include second high 24-h concentrations or the number of violations of the annual standard. A smoother plot of the basic data can be obtained by using some of the techniques listed in Table 2-1. The trend lines in Figure 2-1 were, for example, generated by the application of the Whittaker-Henderson formula.⁸

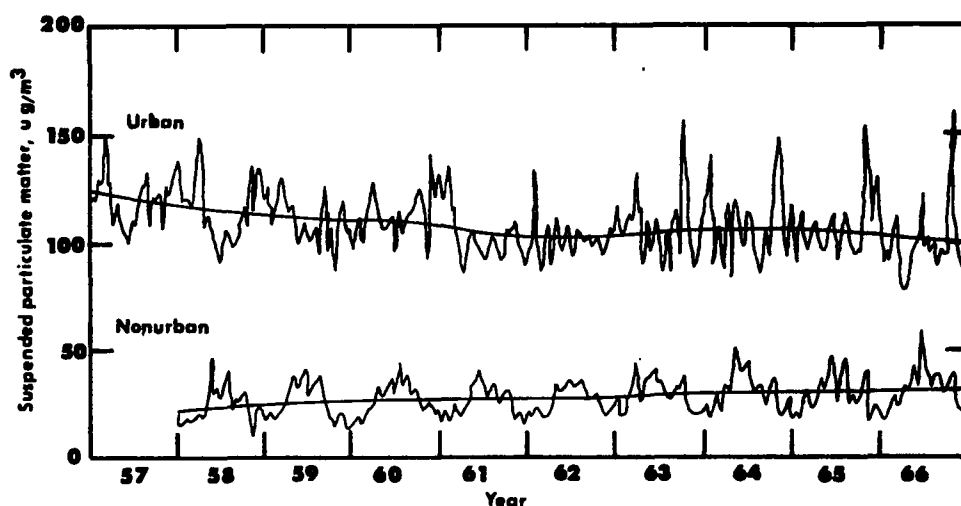


Figure 2-1. Long-term trends at composite sites.

Source: Reference 8

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Statistical techniques, such as those listed in Table 2-1, are desirable when a trend is not clearly distinguishable from a graphical plot of the data. They may be necessary to sort out the real change in air quality from random variability produced by such factors as wind speed or precipitation. Although statistical techniques are objective in the sense that they are reproducible, they are subject to the errors of hypothesis testing discussed in standard statistics texts.

It must be emphasized that an adequate data base must be available in order to perform any statistically valid trend analysis. For a three year running average, an absolute minimum

of five years' data is required, although a longer period of data acquisition (such as seven years) may be preferable.

2.1.2 Applicability of Technique

Long-term trend analysis characterizes the aerosol in terms of its variability over time. It can also permit the analyst to generate tentative propositions concerning the causes of the observed trends. A slow steady decrease may, for example, reflect the conversion of homes and commercial establishments from the use of coal to the use of natural gas as the primary heating fuel. A sharp increase or decrease in concentrations can be related to the start-up or shutdown of a source within the sampling area, if it coincides in time with such an event.

Care must be taken when proposing causes of long-term trends. Any proposition may be erroneous due to other random factors, such as meteorological interference, which can also affect the TSP concentration. This suggests that additional investigation of probable causes of trends should be performed. Used by itself, long-term trend analysis techniques yield little insight into the causes of a trend or the determination of the impact of minor changes in the amount or location of emissions.

2.1.3 Relationship to Other Techniques

Long-term trend analysis is a primary tool in the process of generally defining a particulate problem. Inferences as to the causes, effects, and scope of the problem can be made when spatial patterns and meteorological variations are also considered. Long-term trends by site type (e.g., urban versus rural) can assess the role of fugitive dust as the major contributing source category or can suggest the existence of other dominating source categories. Trends in intersite correlations can also be used to generate propositions concerning the nature of the dominant impacting sources.

A deviation from the historical trend can often be related to the meteorology of the area. Changes in precipitation, wind

speed, and/or wind direction from the historical average can significantly affect measured TSP concentrations for the period of the deviation.

Interpretation of physical, chemical, and elemental data can help identify major source categories as well as determine the impact of specific sources on the past air quality. Such data are rarely contained in the historical record, however, and cannot normally be included in the initial analysis of a particulate problem.

2.1.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Optional
Data	Minimum of five years historical TSP data

2.1.5 Example Application

An example can be drawn from a data summary generated for Seattle, Washington.¹⁰ Data had been acquired continuously at the Public Safety Building in Seattle since February 1965. The 12-month moving geometric mean plot in Figure 2-2 shows short-term fluctuation, but it also depicts a long-term downward trend which appears to level off just below the value of the annual standard. This long-term trend is even more evident in the 24- and 36-month moving geometric mean graphs. Assessment of a trend based on isolated 12-month segments of the 12-month moving geometric mean trace could easily be erroneous: the period from July to November 1974 indicates a sharp upward trend and just the opposite during the same period in 1975. From this analysis, it was concluded that ambient levels of suspended particles were decreasing in this major urban area.

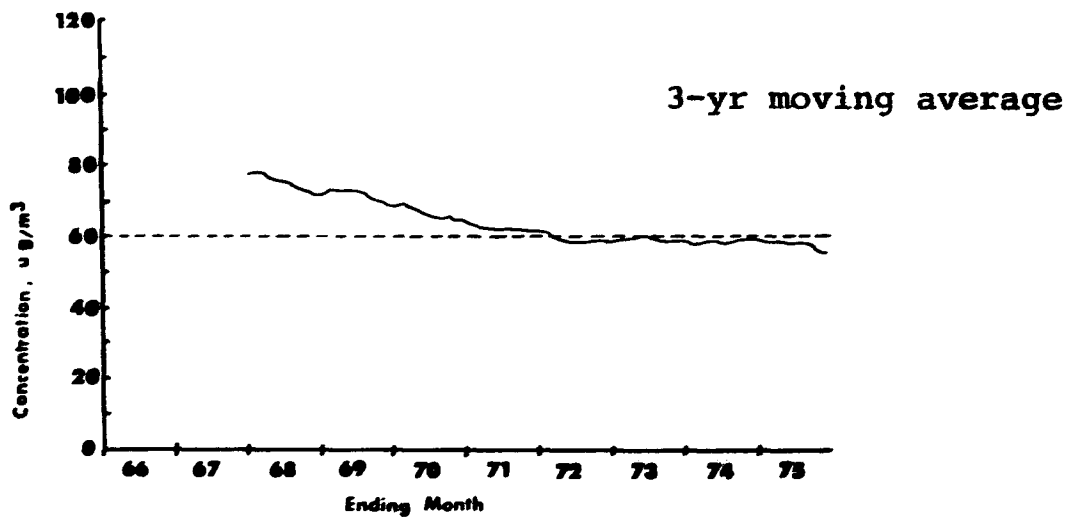
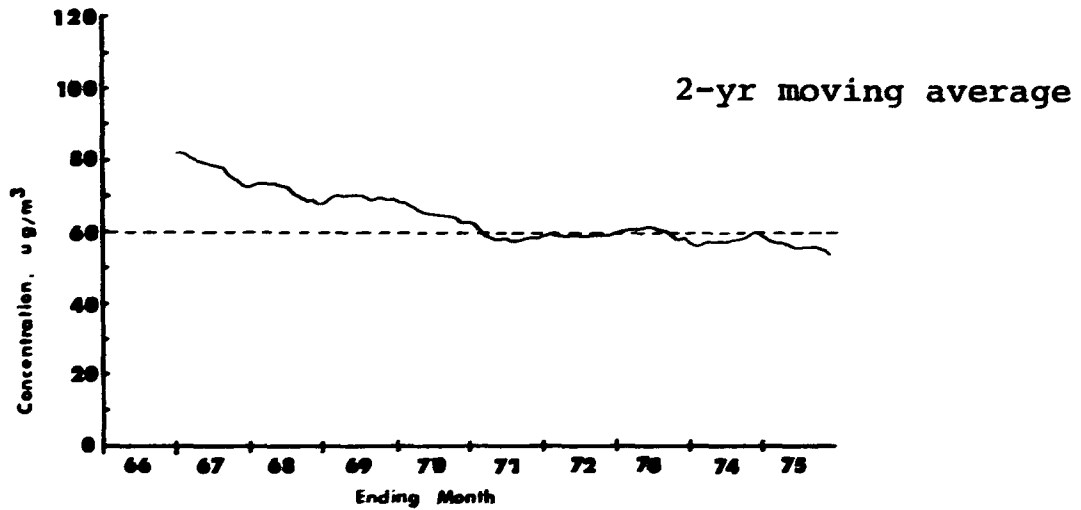
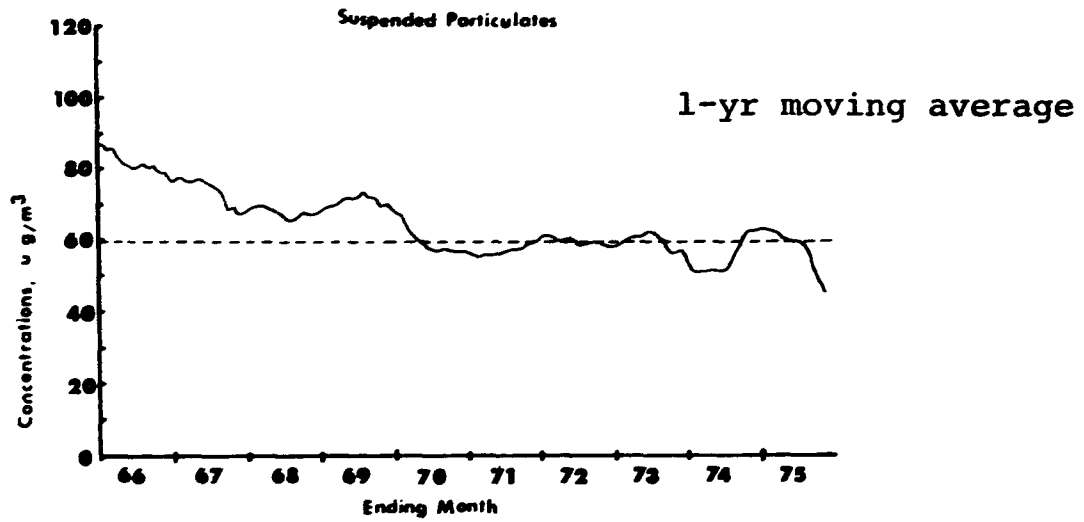


Figure 2-2. Long-term trends, Seattle, Washington.
Source: Reference 10

2.1.6 References

3, 8, 9, 10, 11, 12, 13, 14, 15

2.2 SEASONAL AND MONTHLY PATTERNS

2.2.1 Description of Technique

This technique is very similar to the long-term trend analysis described in Section 2.1.1. Usually, seasonal and monthly averages are plotted versus time over a period of several years. A quarterly or monthly moving average can also be used when curve smoothing of the raw data is desired. A smoother plot can also be obtained using quarterly or monthly moving averages or other techniques such as the Whittaker-Henderson formula. Figure 2-3, for example, is the same plot as Figure 2-1 with a different value for the constant "a" in the Whittaker-Henderson formula to approximate the seasonal trend curve.

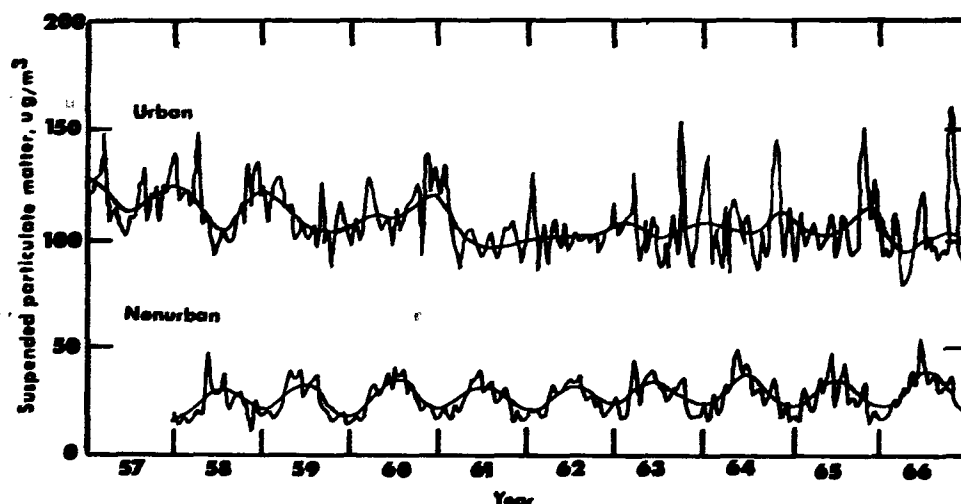


Figure 2-3. Seasonal patterns at composite sites.

Source: Reference 8

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Quarterly and monthly averages can also be computed for a number of years in combination. A minimum of three years' data is recommended if the data are to be used in this fashion. This will tighten the confidence limits around the computed means,

tend to factor out the effect of large-scale variations in meteorological conditions, and minimize the variations in industrial, fuel combustion, or other man-related emissions that have occurred.

Statistical techniques presented in Table 2-1 can also aid in the analysis.

2.2.2 Applicability of Technique

As in the case of long-term trends, this technique characterizes the aerosol in terms of its temporal fluctuation. Indirect identification of the major sources or source categories contributing to the measured TSP levels is also possible. As an example, consider the seasonal patterns depicted in Figure 2-3. The urban sites' tendency to produce peak values in the winter season may be largely attributable to the use of large amounts of fuel (other than gas) for space heating. The nonurban sites' tendency toward higher values in the summer may be closely related to fugitive and windblown dust from cyclical activities in rural areas.

Care must be taken when proposing causes of seasonal or monthly trends, however, since many other random factors can also affect the TSP concentrations. Precipitation, wind speeds, and wind direction can all play an important role in causing TSP concentrations. As with long-term trends, additional investigation of probable causes is often warranted.

2.2.3 Relationship to Other Techniques

Relatively little information is obtainable from a seasonal or monthly trend analysis when used alone. But this technique can suggest areas where further analyses are warranted. When used in conjunction with other techniques, more detailed analyses can be performed, such as identifying the location of the particulate problem, factors affecting TSP concentrations, and characteristic source categories contributing to those concentrations. Through the use of spatial pattern techniques (such as

comparing urban and nonurban sites), certain inferences can be made as to the role of fugitive dust sources. Seasonal pollution roses can be used to indicate possible contributing sources or to determine if the problem is due to more than one source. The scope of the problem can be refined through the use of intersite correlations over time. Given seasonal or monthly trends from several sites, intersite correlations can be performed to determine if the problem is regional or site specific for all seasons of the year.

The effects of meteorological conditions are easily assessed when used in conjunction with seasonal and monthly trends. Often, high correlation coefficients can be obtained when comparing wind speed, wind direction, or precipitation data to the seasonal trend. This comparison can suggest whether fugitive dust, industrial facilities, or other identifiable sources categories are culpable.

Interpretation of physical, chemical, and elemental data can aid in identifying contributing sources if the temporal and spatial patterns of these data coincide with the seasonal or monthly TSP patterns. An example of this is agricultural activity. If a tracer element related to agricultural operations is determined to have the same seasonal pattern as the TSP concentration, then the dominant role of that source category can be inferred.

2.2.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Optional
Data	Minimum of three years historical TSP data

2.2.5 Example Application

An example of seasonal variations and their interrelationship with other techniques is drawn from Phoenix, Arizona.¹⁶ This example consists of an evaluation of seasonal patterns of particulate levels and the associated seasonal meteorology affecting the levels.

Figure 2-4 presents the quarterly averages of concentrations of TSP at various high volume monitoring sites throughout the Phoenix area. It is suggested that the seasonal patterns and magnitudes fluctuate considerably from year to year due to variations in seasonal meteorology and statistical limitations stemming from the small number of samples per quarter. The authors concluded that:

- Concentrations of particulates did not appear to exhibit a consistent seasonal pattern at any of the stations
- For any given year, a similar seasonal pattern was observed at the different stations

The authors then investigated the relationship between these patterns and various meteorological parameters and found that:

- Measured levels of TSP were inversely proportional to rainfall frequency for any given season of the year
- Levels of TSP appeared to be less sensitive to rainfall frequency in the winter months than in the warmer summer months
- Levels of TSP were generally highest in the fourth and first quarters for a given number of rainfall days
- TSP concentrations were relatively the same in all but the second quarter when rainfall frequency increased
- TSP concentrations were inversely related to wind speed
- The most severe seasonal TSP concentrations were apparently caused by fugitive dust arising from human activities during periods when wind speeds were minimal

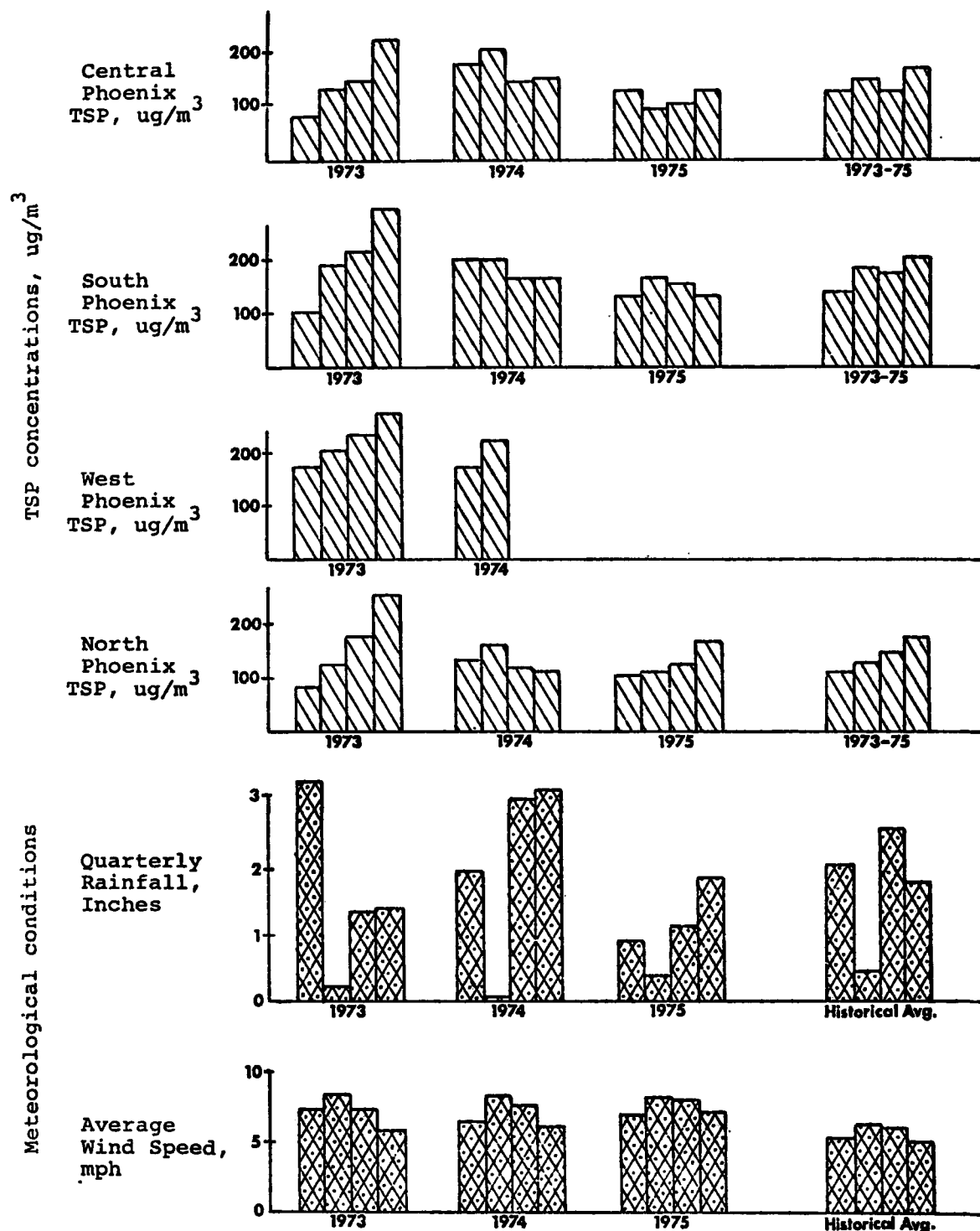


Figure 2-4. TSP quarterly averages, quarterly meteorology for the Phoenix area.
Source: Reference 16

2.2.6 References

8, 9, 10, 11, 12, 14, 15, 16, 17, 18

2.3 WEEKDAY/WEEKEND AND DAILY PATTERNS

2.3.1 Description of Technique

Historical data generally contain an equal number of samples collected for each day of the week. Averaging these data for each day or for weekdays/weekends reveals the average daily or weekday/weekend variations for a particular site or area. Since one year of historical data contains only a small number of samples for each day (typically nine per site), several years' data are normally necessary to derive any statistically valid inferences concerning sources or locations of emissions. The standard statistical techniques presented in Table 2-1 can be used to indicate significant differences and/or causes of the variations.

2.3.2 Applicability of Technique

Again, this technique characterizes the aerosol in terms of its temporal fluctuation. However, given an adequate number of data points, hypotheses indicating the major contributing sources or source categories can be proposed. High correlation coefficients between daily TSP levels and traffic patterns can often be obtained, thereby suggesting traffic as the major source. With certain assumptions pertaining to background levels and relative contributions from different sources, the qualitative impact of a specific source or source category can be estimated. Variations in emission rates of contributing sources can also be suggested.

2.3.3 Relationship to Other Techniques

Several other techniques can be used with the daily average variation analysis to enhance the investigator's understanding of the nature of the measured particulate levels. The spatial pattern analyses can yield additional information useful in

determining the locations of sources. Concentration versus site type for each day of the week and intersite correlations can indicate whether the levels are region wide or site specific. They can also suggest whether the TSP levels are due to fugitive or windblown dust or due to man's activities (e.g., industrial processes), if emissions from such sources are shown to vary over time in a pattern similar to that of TSP levels. By using an accelerated sampling frequency instead of the standard one out of six days, pollution roses for each day of the week can be used to indicate the direction of the particulate emissions and possibly to aid in locating a source. Care should be taken to account for statistical limitations imposed by the number of data points used, however.

It is difficult to correlate average daily TSP levels with meteorological conditions, although this analysis may yield some useful information. An average daily pattern of rainfall, wind speed, or wind direction will most likely not be found.

Physical, chemical, and elemental data analysis can offer additional insight as to the sources of particulate matter. With such data, TSP levels can be attributed to traffic, fugitive dust, or industrial processes emitting unique tracer elements. The pattern recognition technique can be structured so as to use average daily values as a basic input parameter.

2.3.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Optional
Data	Data base adequate to attach statistical significance to results (minimum of three years' TSP data)

2.3.5 Example Application

An example of a daily variation analysis is taken from Philadelphia, Pennsylvania.¹² Daily TSP levels had been monitored at several Philadelphia sites for a number of years, thereby allowing for a statistically significant analysis of the variations during the weekly cycle of human activity. Normalized TSP values were plotted by day of week (see Figure 2-5), taking background concentrations into account. These normalized values were then compared with city-wide traffic volume by day of the week.

Figure 2-5 implies that fluctuations in daily TSP were closely related to daily changes in city-wide traffic volumes. This analysis tended to indicate a close correlation between TSP levels and traffic volumes. It was observed, though, that daily fluctuations of other emission sources could also be causing much of the noted correlation.

2.3.6 References

9, 11, 12, 14, 15, 16, 17

2.4 DIURNAL VARIATION

2.4.1 Description of Technique

Variations in TSP levels during the day in any given area can be determined using this analysis technique. The data required for such an analysis are not generally contained in the historical record and must therefore be generated as part of a special study. The data are usually generated in terms of one-, two-, or four-h high volume samples collected over several days. TSP concentrations versus time of day can then be plotted, and the diurnal pattern determined. Sampling can be done over any number of days, weeks, or months. For longer sampling durations, less statistical weight is attached to each individual data point and missing data do not greatly affect the significance of the analysis.

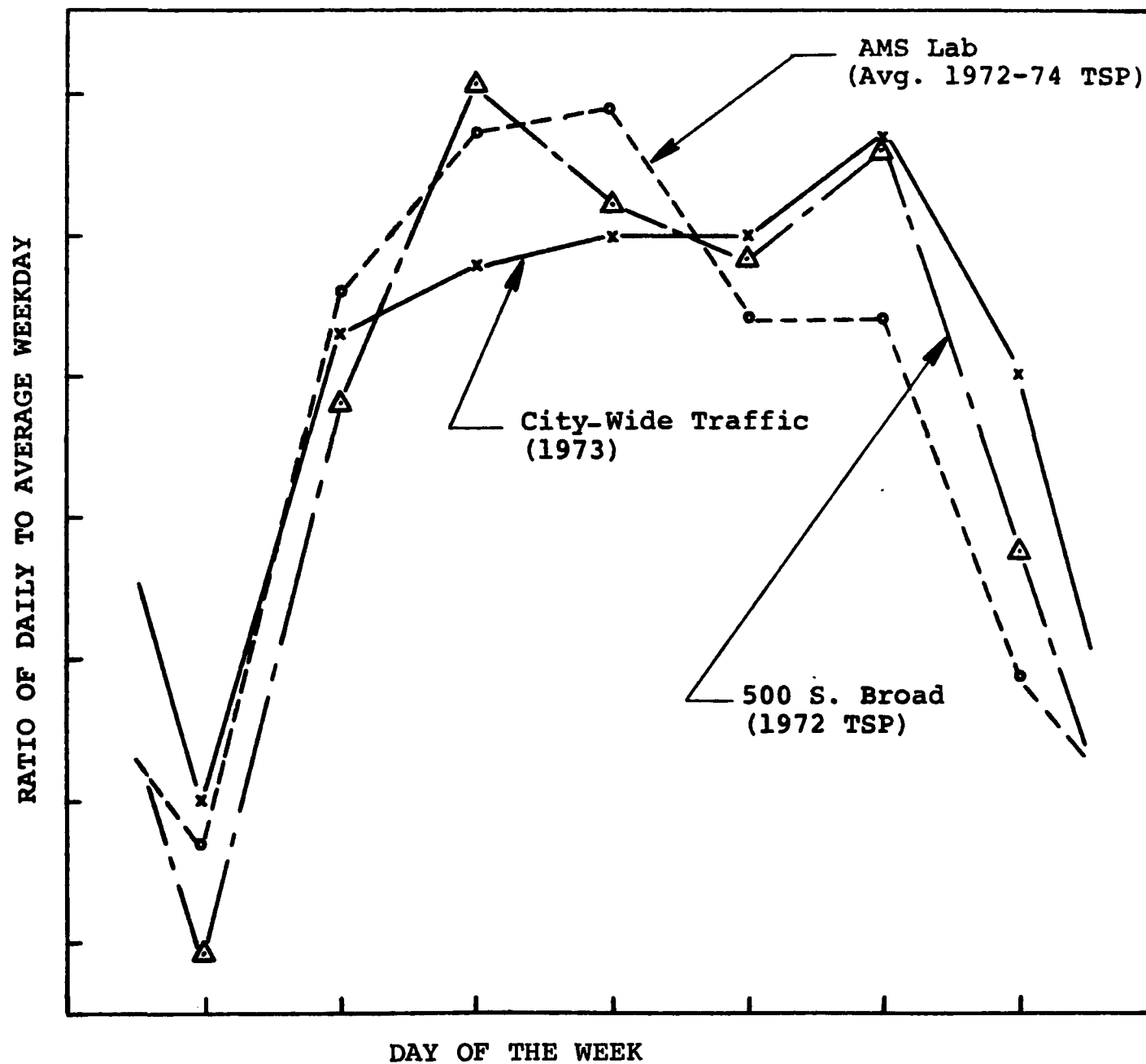


Figure 2-5. Daily variations in traffic and TSP levels.

Source: Reference 12

2.4.2 Applicability of Technique

By defining the typical variation in TSP concentrations during the day, this technique partially characterizes the aerosol. In addition, it is feasible to identify major source categories contributing to the observed concentrations if information is available on the emission patterns in the area. For example, reference 19 presents data relating to diurnal variations in TSP levels and compares these data to the diurnal traffic and power demand patterns. Through this comparison, it was suggested that the TSP levels closely followed the traffic and power demand patterns and were most likely affected by them. If there are no specific sources or source categories which display an unusual diurnal emission pattern, it is unlikely that this technique could be used to identify dominant sources.

2.4.3 Relationship to Other Techniques

The short time period under investigation in a diurnal variation analysis does not lend itself to use with many other techniques. General information is obtainable when this analysis is performed at several sites. Source categories contributing to the TSP levels can be suggested by locating samplers at different site types. Intersite correlations can be used to determine if there are nearby contributing sources and how dominating sources vary throughout the day. Wind speed, wind direction, and mixing height data can be used to assess their effects upon TSP levels throughout the sampling period.

By interpreting chemical, physical, and elemental data in the diurnal cycle, more specific information can be obtained, such as an identification of major contributing sources or source categories. Filter analysis techniques, such as microscopy and X-ray diffraction, can also be used to identify variations in impacting sources during each sampling period.

2.4.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Moderate ^a
Skill	Low
Computer	Not required
Data	One- to 4-h TSP averages of sufficient quantity to attach statistical significance to results

^a Manpower requirements are such due to special data and analysis requirements.

2.4.5 Example Application

An excellent example can be drawn from Philadelphia, Pennsylvania, where diurnal TSP levels were compared to diurnal traffic patterns.¹¹ It was hypothesized that the TSP levels measured at a particular intersection were traffic related and possibly were due to reentrainment of road dust. Four sampling periods were specified, and 13 periods were sampled, starting on a Friday night and continuing until the following Monday morning.

The hypothesis of reentrainment of the redistributed dried particles by vehicular traffic was supported by Figure 2-6, which presents the measured TSP concentrations and traffic volumes for each sampling period. Two samples were affected by sandblasting operations (displayed on the plot as the last two data points). Calculation of the linear regression correlation coefficients for the remaining 11 pairs yielded a value of +0.79, which is significant at the one percent level. Additional data revealed that a significant difference in TSP levels prior to, during, and after street flushing also existed, thus statistically supporting the hypothesis.

2.4.6 References

9, 11, 17, 19

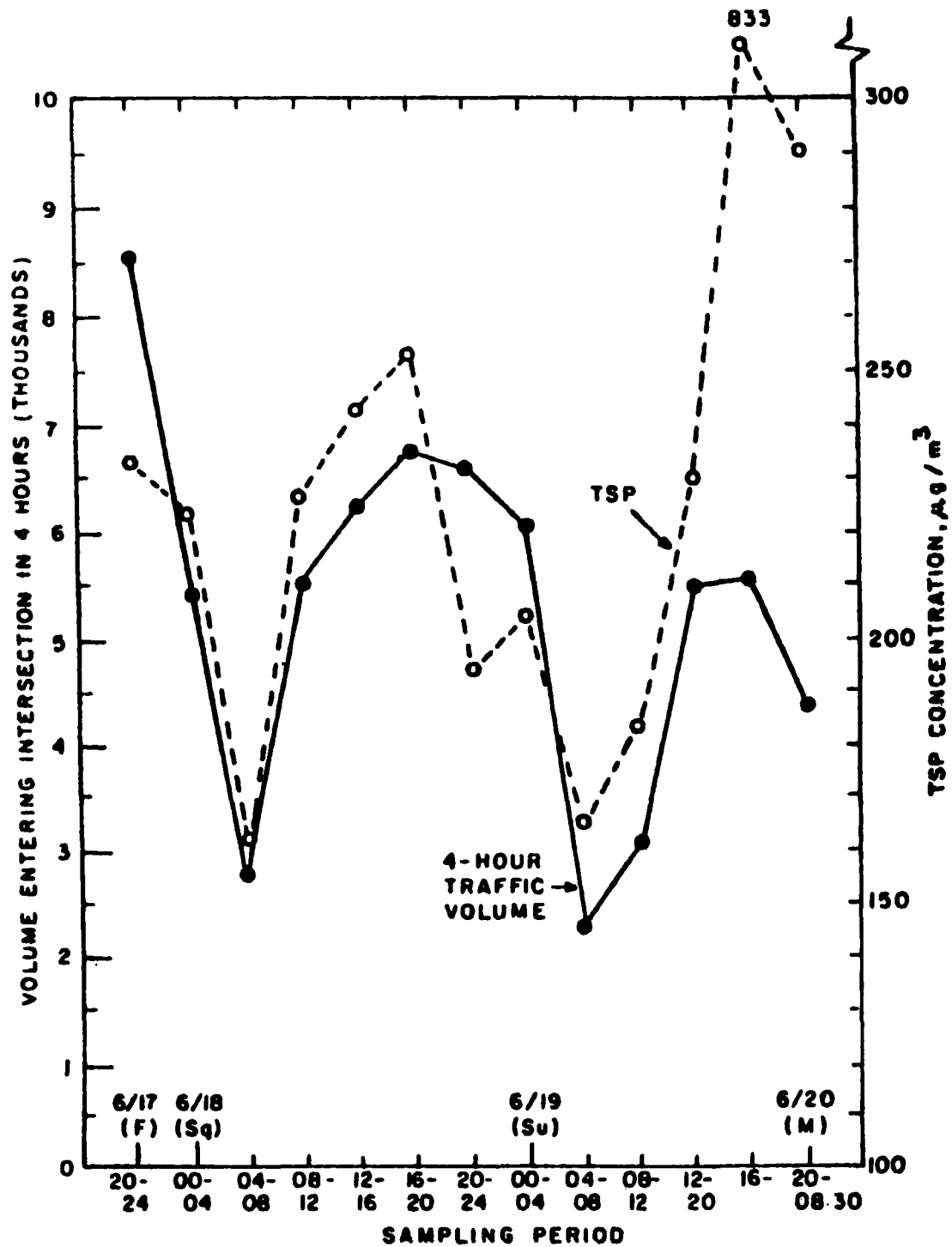


Figure 2-6. Comparison of TSP concentration and traffic volume at Broad and Spruce Streets.

Source: Reference 11

2.5 EMISSION PATTERN/AIR QUALITY RELATIONSHIPS

2.5.1 Description of Technique

In concept, this is a very simple technique. Emission patterns or activity patterns related to emissions from a specific source or source category are compared by standard statistical techniques to TSP data patterns covering comparable time periods. In practice, the technique is complicated by the commonly observed absence of detailed emission/activity data and the limitations imposed by the standard TSP sampling schedule of every sixth day. Emission/activity patterns investigated by this technique have included the following:

- ° Traffic counts for a street immediately adjacent to a sampling site
- ° Heating degree-days (related to space heating emissions)
- ° Product output rates from specific industrial processes
- ° Open field burning

2.5.2 Applicability of Technique

This technique does not help to characterize the aerosol, but it can lead to an identification of source categories impacting upon the sampler. The primary utility of the technique is that it helps to define qualitatively the TSP impact of a specific source or source category.

2.5.3 Relationship to Other Techniques

Generally, there is a direct relationship between this technique and those which analyze the temporal variations in air quality. One determines the existence of a temporal pattern for one parameter (TSP or emission data), then looks for confirmation of a meaningful relationship by analyzing the temporal variation in the other.

Similarly, propositions derived from the analysis of TSP spatial patterns or the effect of meteorological variables can be further investigated by analyzing emission patterns. Thus, if the shape of a pollution rose is inclined toward a nearby roadway and TSP values at that site are inversely related to rainfall, then the potential significance of street dust can be substantiated by a correlation analysis between TSP values and traffic volumes on the nearby street.

Particle size, element, chemical, and morphological data can be used to investigate further the suspicions aroused by the application of this technique. Thus, variations in Pb and Br concentrations could be correlated with traffic volume to extend the example discussed above.

2.5.4 Resource Requirements

Resource requirements vary with the specific situation to which the technique is applied. The estimates given below are based upon the application of the traffic volume/ TSP correlation method:

Manpower	Low
Skill	Low
Computer	Not required
Data	TSP measurements and traffic

2.5.5 Example Application

This example is drawn from a study of the impact of reentrained dust upon TSP concentrations in Kansas City, Missouri.²⁰ Two sites were located on either side of a relatively well-traveled street. High volume samplers were placed at two different heights at each location. Data were collected over a three month period. A linear regression analysis between TSP values at one of the samplers and traffic counts on the street was performed. As Figure 2-7 indicates, a correlation of approximately +0.8 was found. The effect of each additional vehicle

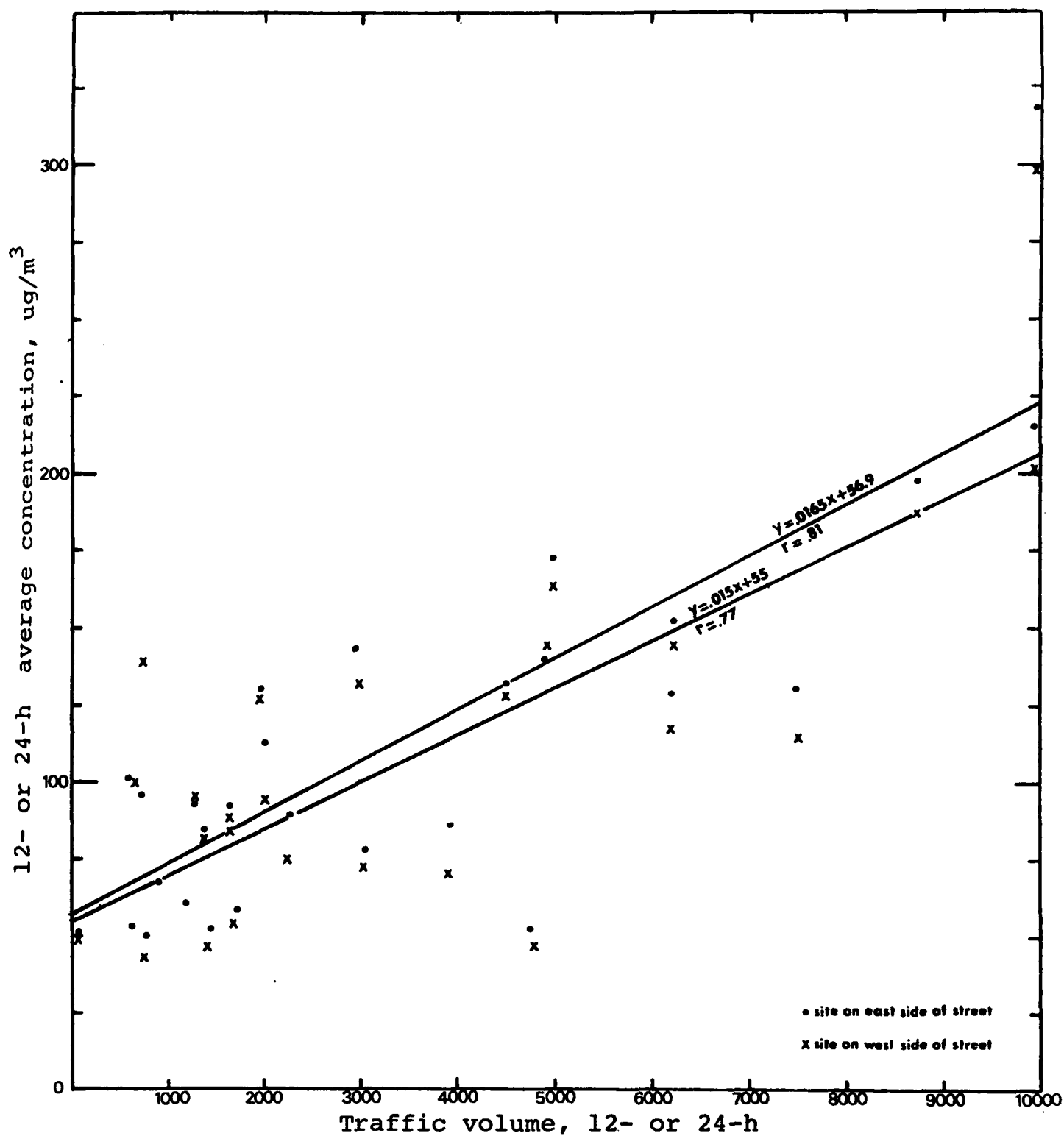


Figure 2-7. Traffic volume versus concentration for McGee Street sites, Kansas City.

Source: Reference 20

upon TSP concentrations was also investigated by comparing concentrations and traffic volumes for different time periods at two different sites. As Table 2-2 suggests, the effect on the sites was not consistent. Qualifications attached to the results of this study were that emissions from other sources may have interfered with the results and that the emission rates per vehicle may actually have varied considerably from site to site, depending upon roadway conditions and other factors.

2.5.6 References

11, 12, 20, 21, 22, 23, 24

Table 2-2. EFFECT OF TRAFFIC VOLUME ON CONCENTRATIONS

Location/ period	Average traffic volume for period	Arithmetic average particulate ₃ concentration, ^a ug/m ³	
		Ground level (5ft) sites	Elevated (20ft) sites
<u>McGee Street, Kansas City</u>			
Weekday	5360	140.4	129.6
Weekend	1560	92.2	81.4
8 a.m. to 8 p.m. (day)	3190	196.4	177.3
8 p.m. to 8 a.m. (night)	863	100.9	80.6
<u>Hamilton Ave., Cincinnati</u>			
Weekday	17792	70.6	63.7
Weekend	17325	66.6	60.8
8 a.m. to 8 p.m. (day)	12682	72.2	63.7
8 p.m. to 8 a.m. (night)	5110	59.8	56.7

^a All samplers are 35 ft from edge of street.

Source: Reference 20

3.0 ANALYZING SPATIAL PATTERNS OF SUSPENDED PARTICULATES

In this chapter, four techniques for analyzing the spatial pattern of suspended particulates are discussed:

- Stratifying concentrations by site environment
- Intersite TSP correlations
- Pollution, dosage, and gradient roses
- Upwind/downwind relationships

3.1 STRATIFYING CONCENTRATIONS BY SITE ENVIRONMENT

3.1.1 Description of Technique

The environments within which monitoring sites are located can be divided into a variety of classes and can be used to account for the spatial variation in air quality. Examples of classification schemes that have been reported in the literature include the following:^{13,16,21,25,26}

Urban	Central city residential/ commercial (fugitive dust sources)	River valley
Rural	Central city residential (no fugitive dust)	High density Town
Traffic	Rural/residential (fugitive dust sources)	Suburban
Non-traffic	Suburban residential (fugitive dust sources)	Background
	Rural residential (no fugitive dust)	Non-urban
	Remote	Residential
		Commercial
		Industrial

The technique itself is rather simple. Once the sites are classified by environment, TSP data associated with those sites are aggregated and summarized statistically in terms of their

average geometric mean. These data can then be analyzed for temporal variations, or for the effect of meteorological variables.

3.1.2 Applicability of Technique

This technique can be useful in generating a more accurate understanding of the regional distribution of suspended particulate air quality whenever topographical conditions negate the level terrain assumptions utilized in most diffusion models or whenever the major impacting emission sources are fugitive dust related. However, the technique cannot be used to characterize the aerosol in any other way.

Major impacting source categories can be qualitatively suggested by this technique insofar as the site classification scheme is developed objectively and not in a manner calculated to prove the culpability of a given source. Such source categories can be implicated by examining each high TSP site type for common attributes. No quantitative assessment of source impact is possible with this technique, however.

No determination of the quantitative impact of a single source has been demonstrated with this technique. No determination of the impact of changes in emissions has been demonstrated.

3.1.3 Relationships to Other Techniques

The temporal variation in suspended particulate concentrations and the effect of meteorological conditions upon those concentrations can be assessed for the various site classes. In combination, these techniques can assess the role of fugitive dust as the dominating source category or can suggest the existence of a dominating group of sources. Likewise, when used in conjunction with other spatial pattern techniques, site classification can suggest the location of dominant sources. When applied to chemical/elemental data, this technique can be used to identify key source categories even more precisely (see Section 6.1).

3.1.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Optional
Data	Site descriptions, TSP measurements

3.1.5 Example Application

In a study recently prepared for the Kansas City region, 31 monitoring sites were divided into five different site exposures as shown in Table 3-1 below:¹³

Table 3-1. EXAMPLE SITE CLASSIFICATION

Class	Description
Background	Sites outside the central city and remote from obvious local man-made sources of particulate matter
Town	A town outside the central city
Suburban	Areas principally residential in character outside commercial-industrial areas
High density	Locations outside river valleys but having a high level of commercial or industrial development
River valley	Low elevation sites in river valleys with surrounding higher elevation

Stratified by their relationships to the primary annual NAAQS, 12 of these sites were found to exceed that standard (see Table 3-2 below).

Table 3-2. EXAMPLE STRATIFICATION OF TSP CONCENTRATIONS
BY SITE TYPE

Class	<75 ug/m ³	>75 ug/m ³
Background	2	0
Town	5	3
Suburban	8	1
High density	1	3
River valley	3	5

Quarterly geometric means were then computed for each of these site classes, yielding a graphical plot showing distinct seasonal changes in TSP concentrations. According to the authors, high concentrations measured during the winter tended to refute the hypothesis that fugitive dust sources (excluding reentrained dust) had a major impact, since fugitive dust impact should be highest in the dry summer and fall. The prevalence of high wind speeds and the occurrence of street sanding during the winter tended to support a reentrained dust impact hypothesis.

Correlation coefficients were calculated for particulate concentrations versus wind speed, mixing height, days since precipitation, and barometric pressure for each of the five site classes. The correlation coefficients shown in Table 3-3 were noted; however the confidence limits around those values were not reported.

Table 3-3. EXAMPLE CORRELATIONS OF METEOROLOGICAL DATA
FOR SITE TYPES

Class	Wind speed	Mixing height	Correlation coefficient	
			Days since precipitation	Barometric pressure
Background	-.033	.221	.555	.051
Town	-.024	.315	.290	-.160
Suburban	-.264	.267	.286	.040
High density	-.169	-.050	.237	.121
River valley	-.157	.085	.426	-.031

The authors concluded that the four meteorological parameters which were investigated failed to account adequately for the observed daily variations in TSP concentrations. The differences in correlations from site type to site type were considerable, however, and suggested that the site classification scheme was a meaningful conceptualizing device.

It should be noted that the authors should have assessed their results in terms of probability or confidence limits. Failure to do so may have resulted in a misinterpretation of the results.

3.1.6 References

13, 16, 21, 25, 26

3.2 INTERSITE TSP CORRELATIONS

3.2.1 Description of Technique

Correlation coefficients can be computed between daily TSP concentrations measured at one site and those measured on the same days at each other site in the region. Two sites which are relatively close together (i.e., less than two miles apart) can be expected to correlate highly if they are being impacted by the same dominant source or source category and are in a location with unobstructed wind flow. The correlation analysis can be performed for a data set of one year or more and improves in accuracy as the number of measurements in the data set increases.

Procedures for computing correlation coefficients are amply described in standard statistical texts and need not be described in detail here. In general, the correlation coefficient is defined as:

$$r = \frac{\sum (x_i - \bar{x}) (y_i - \bar{y})}{[\sum (x_i - \bar{x})^2 \sum (y_i - \bar{y})^2]^{1/2}} \quad (\text{eq.1})$$

where x_i = the i th value of the variable x

and $\bar{x} = \frac{\sum x_i}{N}$ = mean of all x values

In this case, x is the TSP value at one site and y is the TSP value on the same day at another nearby site. The best possible correlation is given by $r = \pm 1$. As r approaches zero, the variables become more randomly related. If $r = +1$, x and y are directly proportional; if $r = -1$, they are inversely proportional. Correlation coefficients indicate how well two variables vary together, not necessarily that there is or is not a causal relationship between the two variables.

3.2.2 Applicability of Technique

This technique can characterize the aerosol in terms of its spatial distribution. This is done primarily in terms of assessing the uniformity of fluctuation in TSP concentrations from site to site.

Where nearby sites intercorrelate highly, the presence of a common dominating set of impacting source categories can be inferred. However, it is not possible to identify what those source categories are or what their respective contributions are. The existence of a unique source or source category can be inferred when one site correlates poorly with a set of sites that otherwise intercorrelate highly.

A large negative correlation implies that one site has large values when the other has small values. This could occur when a large point source is located between the sites.

No determination of the impact of changes in emissions has been demonstrated with this technique.

3.2.3 Relationship to Other Techniques

Given a sufficient number of samples, temporal patterns in intersite correlations can be investigated. If the correlation coefficient changes dramatically, it can be inferred that dominating sources vary over time.

Again assuming a sufficient number of samples are available, measured concentrations can be stratified by meteorological conditions and intersite correlations can be computed therefor. The nature of dominant sources can be inferred from changes in the resulting correlations.

Intersite correlation data can also be computed for different species (e.g., Pb, SO₄, NO₃) and compared to TSP intersite correlations. Where TSP intersite correlations are high and species correlations are high, it can be inferred that the two sites are measuring essentially the same air mass.

3.2.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Moderate
Computer	Preferable, depends upon number of sites
Data	TSP measurements at more than one site (adequate number of measurements to assure statistical significance)

3.2.5 Example Application

An example can be drawn from a recent study of the TSP attainment status in Houston, Texas.²² Two sites are located on the north side of the central downtown area. Less than a mile apart, both of these sites have severe wind flow obstructions to the east but they are somewhat dissimilar in their immediate environments (see Figure 3-1). The daily TSP concentrations correlate well ($r = 0.86$). Pollution roses for each site have

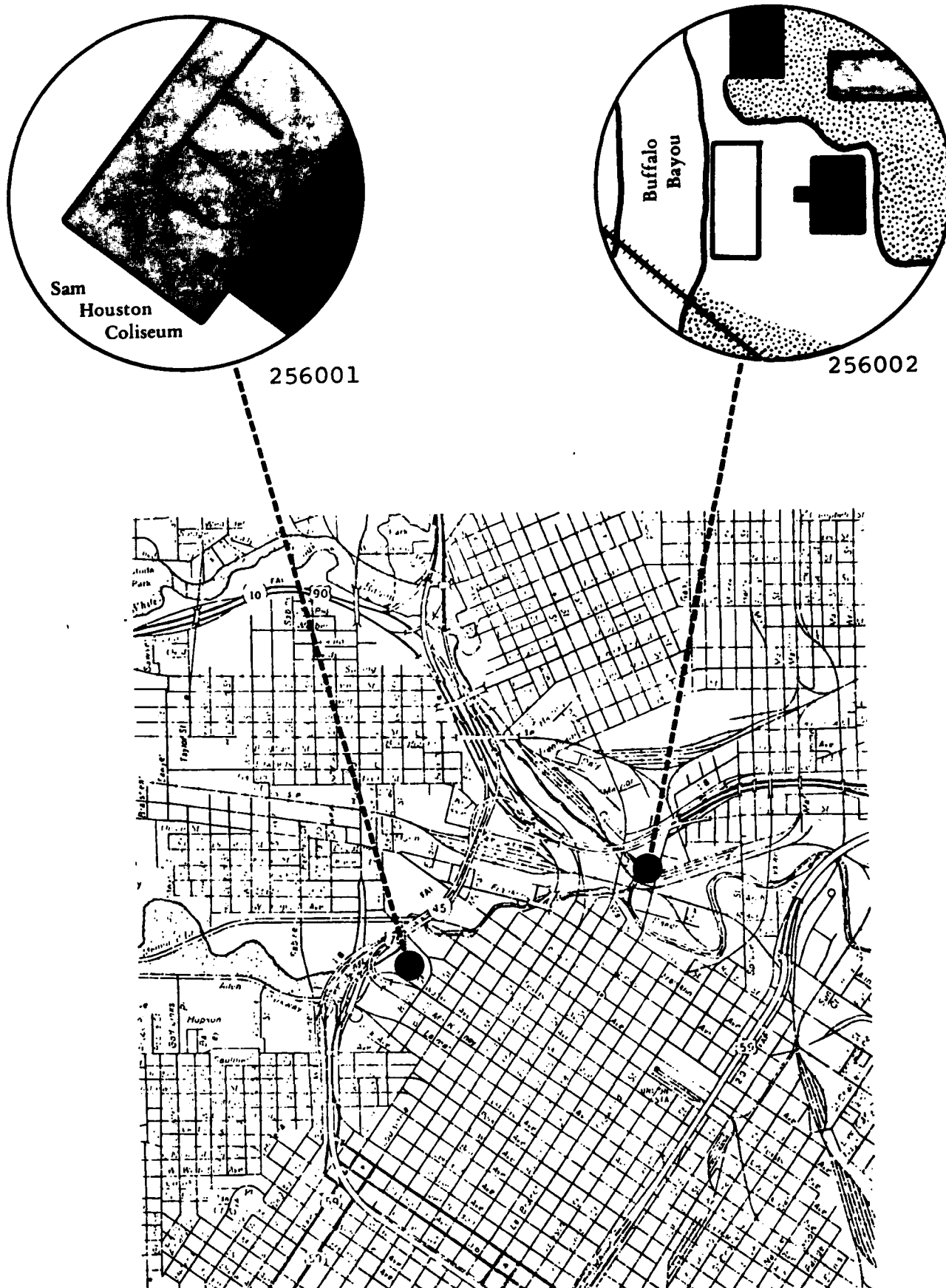


Figure 3-1. Locations of sites to be intercorrelated.
Source: Reference 22

similar shapes. Correlations of TSP and Pb values by wind direction indicate strong relationships when persistent winds come from the north and northwest. Correlations between the two sites in terms of nitrates, sulfates, ashable organics, Mn, and Cu are all greater than +0.75.

From these comparisons, the authors of the study concluded that the two sites measured essentially the same air mass despite the differences in their immediate environments. An analysis of sources in the vicinity of the two sites led the authors to conclude that traffic-related sources to the north and northwest of the sites and industrial sources to the southwest were the primary identifiable contributors to the polluted air mass sampled at those sites.

A matrix display of a typical set of intersite TSP correlations from a recent study in Philadelphia is shown in Table 3-4.¹¹ In this example, daily TSP concentrations at 13 sampling sites are intercorrelated for each of three years. As that table indicates, there was considerable variation in correlations from site pair to site pair and from year to year. Some sites, such as BEL and FRI correlated well from year to year, whereas others, such as S/E and ALL correlated well one year and poorly the next. Again it should be noted that the utility of this matrix would be enhanced, were the results to be expressed in terms of probabilities or assigned confidence limits.

3.2.6 References

11, 22

3.3 POLLUTION, DOSAGE, AND GRADIENT ROSES

3.3.1 Description of Techniques

The three techniques grouped into this category use essentially the same types of information but apply them slightly differently. A pollution rose is produced by directionally analyzing air quality data at a given sampling station. The data

Table 3-4. LINEAR CORRELATION COEFFICIENTS BETWEEN 24-H TSP
LEVELS AT PHILADELPHIA MONITORING STATIONS IN
1974, 1975 AND 1976

Site	DEF	ALL	INT	BEL	ROX	N/E	NBR	FRI*	LAB	SBR	S/E	500	AFS	Average
DEF - '76		0.81	0.69	0.70	0.49	0.76	0.65	0.75	0.78	0.78	0.89	0.79	0.77	0.74
'75		0.61	0.56	0.72	0.64	0.65	0.66	0.68	0.78	0.75	0.59	0.67	0.84	0.68
'74		0.64	0.72	0.59	0.56	0.63	-	0.74	0.79	-	0.80	0.78	-	0.69
ALL - '76			0.57	0.66	0.45	0.60	0.66	0.75	0.70	0.76	0.81	0.85	0.75	0.70
'75			0.52	0.45	0.36	0.66	0.47	0.68	0.66	0.55	0.27	0.47	0.66	0.53
'74			0.50	0.50	0.44	0.51	-	0.74	0.62	-	0.40	0.62	-	0.55
INT - '76				0.37	0.50	0.70	0.55	0.62	0.74	0.67	0.61	0.66	0.59	0.61
'75				0.66	0.51	0.56	0.41	0.60	0.63	0.44	0.47	0.43	0.58	0.53
'74				0.39	0.44	0.60	-	0.56	0.54	-	0.67	0.58	-	0.56
BEL - '76					0.88	0.78	0.85	0.91	0.78	0.81	0.73	0.95	0.56	0.75
'75					0.89	0.80	0.87	0.65	0.86	0.86	0.48	0.76	0.87	0.76
'74					0.56	0.55	-	0.72	0.61	-	0.53	0.62	-	0.56
ROX - '76						0.65	0.60	0.67	0.66	0.62	0.46	0.59	0.67	0.60
'75						0.68	0.66	0.65	0.78	0.64	0.46	0.68	0.54	0.62
'74						0.68	-	0.60	0.61	-	0.19	0.52	-	0.51
N/E - '76							0.63	0.70	0.85	0.71	0.75	0.74	0.71	0.72
'75							0.43	0.72	0.91	0.69	0.54	0.65	0.73	0.67
'74							-	0.54	0.68	-	0.47	0.53	-	0.58
NBR - '76								0.76	0.78	0.89	0.60	0.75	0.72	0.70
'75								0.56	0.66	0.72	0.30	0.66	0.62	0.58
'74								-	-	-	-	-	-	-
FRI - '76									0.75	0.85	0.78	0.83	0.68	0.75
'75									0.80	0.74	0.49	0.63	0.79	0.68
'74									0.70	-	0.67	0.80	-	0.67
LAB - '76										0.81	0.68	0.84	0.87	0.77
'75										0.81	0.54	0.78	0.82	0.75
'74										-	0.54	0.68	-	0.64
SBR - '76											0.74	0.81	0.79	0.77
'75											0.34	0.65	0.83	0.67
'74											-	-	-	-
S/E - '76												0.70	0.84	0.72
'75												0.49	0.50	0.46
'74												0.53	-	0.53
500 - '76													0.77	0.77
'75													0.73	0.63
'74													-	0.63
AFS - '76														0.73
'75														0.71
'74														-

*CAMP station in 1974; roof-top station in 1976.

Source: Reference 11

are segregated according to the wind direction observed while each sample was being collected, and samples with like wind direction are grouped together. Using degree increments of the compass for direction, the result is $\frac{360}{x}$ subsets (where x is the number of degrees in each radial subset), each containing the data points recorded with corresponding wind directions within that $\frac{360}{x}$ degree increment. For each subset, one then calculates an appropriate measure such as geometric mean or arithmetic mean. The resultant numbers indicate the distribution of pollutant readings versus direction. These numbers may be conveniently plotted and interpreted in polar form. A variation of this technique involves computing a pollution rose only for those data that exceed some preselected 24-h concentration (e.g., 75 ug/m^3).

Since the wind blows from some directions more frequently than from others, the impact of source emissions will be weighted not only by the magnitude of the readings associated with that source, but also by the frequency of occurrence of those readings. A dosage rose can take this fact into account. A dosage rose is produced by plotting the total dosage received at the sampling site in terms of concentration-days (ug-days/m^3) rather than the average TSP concentrations.

A gradient rose is similar to a pollution rose, except that the difference in TSP levels between two sites is displayed as a function of wind direction instead of the average TSP level versus wind direction. It is produced by selecting only those days when winds blow persistently toward one sampler from the direction of a nearby (e.g., <8 miles distant) sampler, calculating TSP concentrations at the two sites, and recording the difference. The resulting values are displayed on a polar plot. A negative value is recorded for the first site if its measured mean concentration is lower than that of the nearby site.

3.3.2 Applicability of Technique

Other than by partially describing the spatial variation in TSP concentrations, this technique does not lead to a characterization of the aerosol.

This technique identifies major impacting source categories only insofar as a distinct set of such source categories can be pinpointed by the shapes of pollution roses generated at nearby sites.

A pollution rose can pinpoint the existence of a major source if the shape of the rose clearly points toward an identifiable source. A semiquantitative assessment of the contribution from that source can be made by comparing the average concentrations for other directions with the one pinpointed by the rose. The dosage rose will do the same, but it will also take the impact of dominant wind directions into account.

The gradient rose will add to the understanding provided by the pollutant/dosage roses. If there is a large positive gradient in a particular direction, on the average, TSP levels are higher at the site under study than at the nearby upwind site. This large increase indicates a localized source between the two samplers. A small gradient indicates an area source or the presence of many point sources which affect the two sampling sites approximately equally. A large negative gradient indicates that either an effective sink for airborne particulates is located between the two sampling sites or a large source is located upwind of the upwind sampling site.

A key potential limitation associated with this technique is that the wind direction data upon which it relies may have been generated from a location which is not representative of the location at which the TSP concentrations were measured. The best way to avoid this problem is to generate site-specific wind data whenever possible.

3.3.3 Relationship to Other Techniques

Given a sufficient number of samples to make a statistically valid comparison, pollution roses can be computed for differing time periods. The resulting roses can be related to available knowledge concerning the types, magnitude, and seasonal variations in emissions from nearby sources. Similar computations can

be made for different sets of meteorological conditions. The possible presence of dominating nontraditional sources can be investigated through the use of this latter combination of techniques.

Data from the most current emission inventory can be used to select sampling sites that can be investigated through the use of pollution roses. Likewise, microinventories can be applied to sites which have inexplicable pollution rose shapes. Given adequate data, particle size distributions for various chemicals and elements can also be stratified by wind direction (see Section 7.2).

3.3.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Not required
Data	TSP and site-representative wind direction measure- ments

3.3.5 Example Application

Three sites in the northeast part of the Houston, Texas metropolitan area are shown in Figure 3-2.²² Pollution roses for those sites, also shown in Figure 3-2, indicate that the highest concentrations at each of these sites occur when winds blow from an industrial area to the south and southwest. Examination of the gradient roses (see Figure 3-3) indicates that higher concentrations are measured at the center site than at the other two when winds blow through the residential area. There is a dramatic increase at the center site when winds are from the southwest across both the industrial area and a four-lane highway. The study from which this example is drawn concluded that although the impact of the industrial area was evident, it was not known whether point sources or fugitive emissions from that industrial area were the cause of that impact.

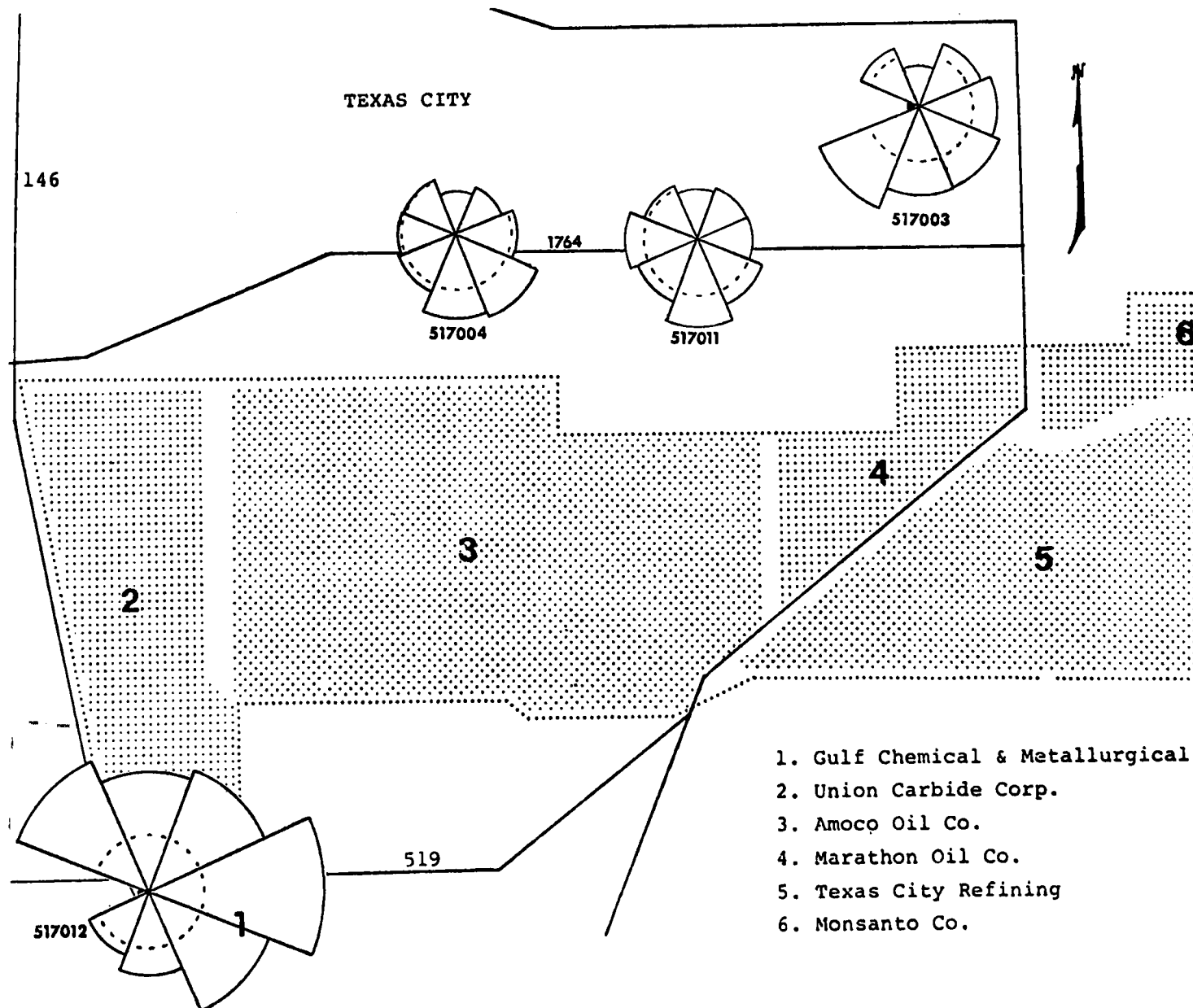


Figure 3-2. Location of sites and pollution roses.

Source: Reference 22

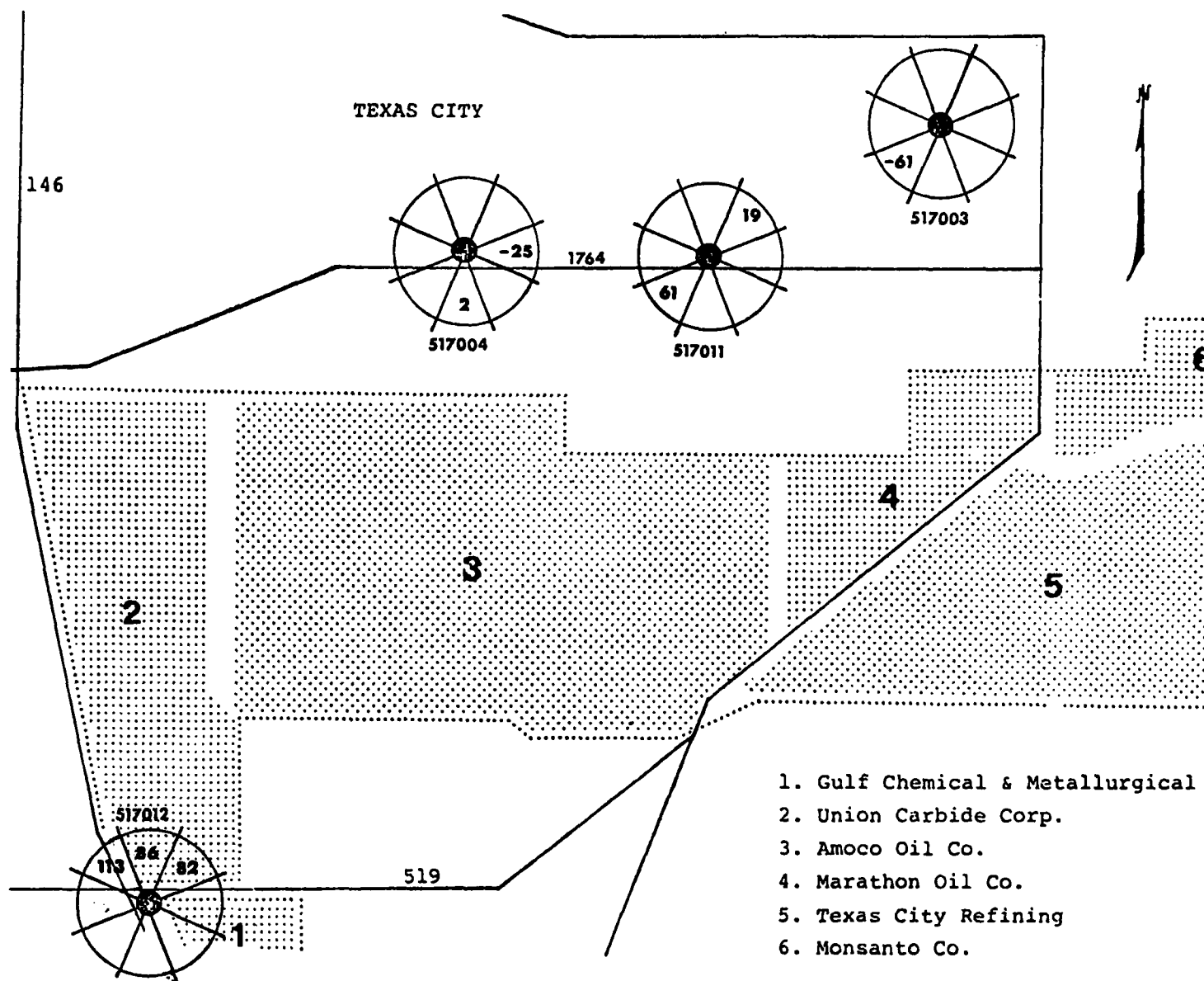


Figure 3-3. Gradient roses.

Source: Reference 22

3.3.6 References

22, 27, 28, 29

3.4 UPWIND/DOWNWIND RELATIONSHIPS

3.4.1 Description of Technique

This technique can be applied on two different scales: regional or local. In both cases the concept is simple: to measure concentrations upwind and downwind from a suspected major source of particulate matter and to compute the difference in concentrations for that period when winds were consistently from the upwind direction. On a regional scale, this technique can be used to determine the relative contributions of natural and urban-generated aerosols. On a local scale, it can be used to estimate the impact of a specific source.

3.4.2 Applicability of Technique

Other than revealing the spatial variation in air quality, this technique does not shed any light on the character of the aerosol. But, the regional scale upwind/downwind analysis can provide some information as to the role of naturally occurring windblown dust combined with the long-range transport of secondary particulates.

The primary utility of the upwind/downwind technique is that it can accurately quantify the impact of identifiable sources. This capability is especially useful in conducting special studies of the impact of a single source. (See example below.)

Once a source's relationship to upwind/downwind measurements has been satisfactorily quantified, then the effect of changes in that source's emissions can be estimated by using linear rollback calculations.

3.4.3 Relationship to Other Techniques

For the upwind/downwind technique to be meaningful, it is necessary to analyze only those data generated when the wind

persistently blew from the upwind direction. This can be accomplished either by sampling only during periods of such winds (as with directionally actuated samplers) or by extracting and statistically analyzing only days of direction-relevant winds. It is possible, however, that unknown sources are contributing to measured differences in upwind and downwind concentrations. Microinventories of the areas immediately surrounding the samplers can be helpful in investigating this possibility. More useful yet is to compare the upwind/downwind relationships revealed by this TSP analysis with those revealed by the analysis of species concentrations at the two sites, taking into account the types of particulate matter emitted by the suspected source. Likewise, microscopic analysis of material collected on individual upwind and downwind samples will add supporting evidence to source origin hypotheses.

3.4.4 Resource Requirements

Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Not necessary
Data	TSP and site-representative wind direction measure- ments

3.4.5 Example Application

An example can be drawn from a recent effort to quantify the rate of dust emissions from aggregate storage piles.¹⁵ The stockpiling operations at a sand and gravel pit were selected for testing. Prevailing winds were from the southwest and south.

The storage area shown in Figure 3-4 covers approximately 17 acres. Fifteen stockpiles were located in this area. Other sources, such as crushing and screening, were either located north of the stockpile area or else were inactive during the sampling period.

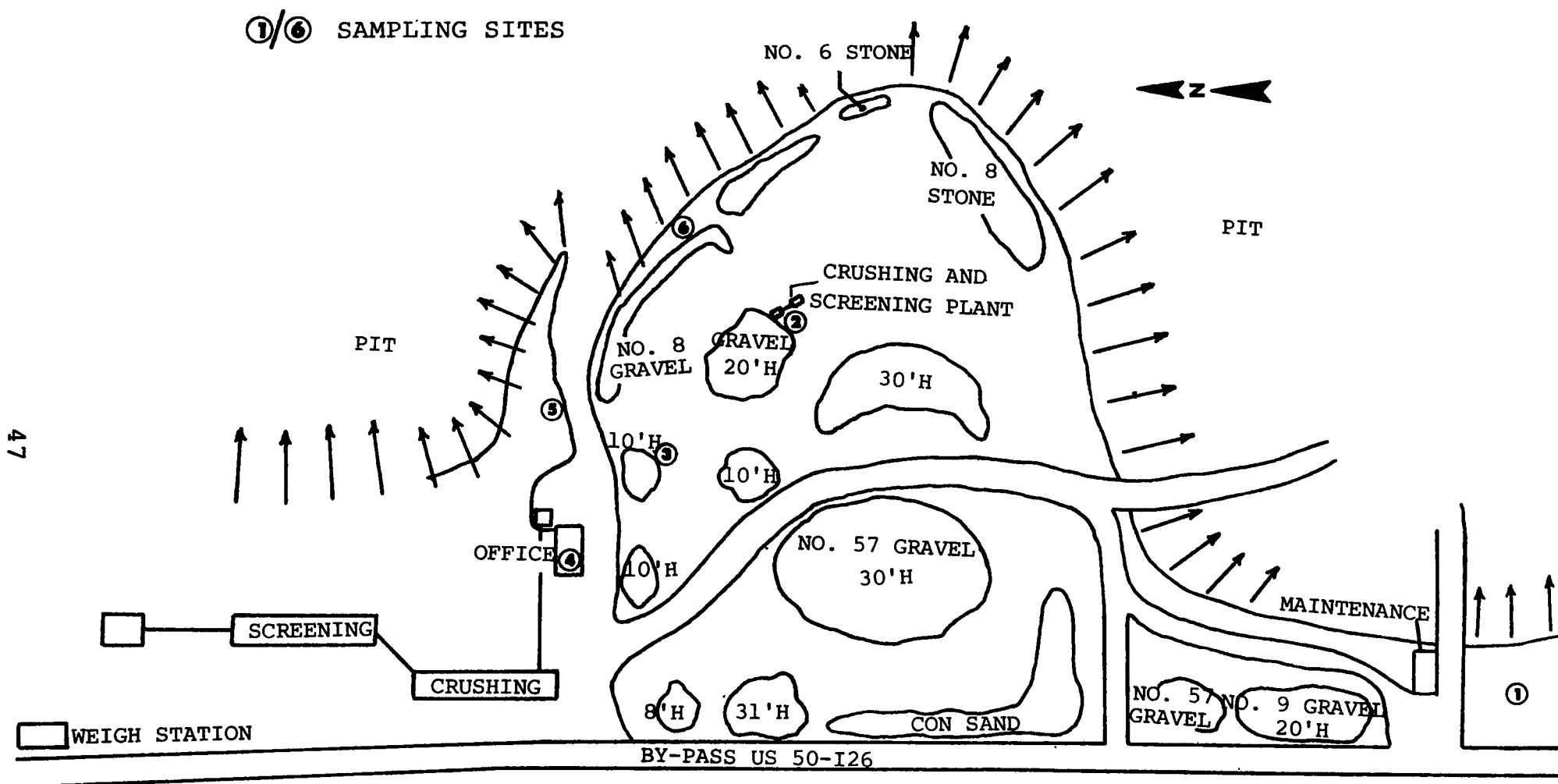


Figure 3-4. Aggregate storage sampling sites.

Source: Reference 15

Field testing was conducted for a one month period and consisted of eleven 24-h runs and eight 12-h runs. Conventional high volume samplers with wind direction activators were used. A 180 degree sector of sampling was employed, so that any wind with a southerly component activated all of the samplers. Five of these samplers were located north (downwind) of the storage area, and one was located to the south (upwind). Wind speed and direction data were also measured and recorded at the site.

As indicated in Table 3-5 below, the test results indicated that the upwind sampler (station 1) recorded lower average concentrations than did the downwind sites:

Table 3-5. EXAMPLE UPWIND/DOWNWIND CONCENTRATIONS

Site	Arith mean, $\mu\text{g}/\text{m}^3$
1 (upwind)	73
2	124
3	76
4	108
5	86
6	107

An evaluation of the effects of four different factors on the emission rates was also performed. Those factors were rainfall, wind speed, type of aggregate material, and amount of activity in the piles.

3.4.6 References

15, 30, 31, 32

4.0 ASSESSING THE EFFECT OF METEOROLOGICAL VARIABLES

Many commonly measured meteorological variables may affect TSP concentrations. This chapter discusses techniques which have been used to determine the relationships between individual meteorological parameters and TSP concentrations. The five techniques discussed are:

- Correlation and regression techniques
- Decision tree analysis
- Analysis of precipitation
- Analysis of wind speed
- Trajectory analysis

4.1 CORRELATION AND REGRESSION TECHNIQUES

4.1.1 Description of Techniques

The most widely used technique to isolate the relationship between an individual meteorological variable and associated TSP concentrations is simple correlation of daily measurements of the two variables (see Section 3.2.1 for discussion of correlation). Several meteorological variables can be evaluated at once and the paired correlations reported in matrix form. Meteorological parameters commonly used as independent variables are listed in Table 4-1. Statistical tables of significance, based on sample size, provide comparative "r" values to determine whether the calculated correlations are significant.

TSP measurements at several different sites in an urban area or region can be placed in the matrix and correlated with the meteorological variables and other TSP measurements. Indicators of particulate air quality other than TSP, such as light scattering or citizen complaints, may also be included as parameters

Table 4-1. METEOROLOGICAL VARIABLES

Meteorological parameters	Alternative forms
Precipitation	Amount, duration (h), days since rain, log of days since rain, 3-day accumulated, amount on preceding day or days
Wind speed	24-h average, max h, min h, resultant, ratio of resultant to average, average through mixing layer
Wind direction	Frequency of each of 8 dir plus calms, total miles of wind from each of 8 dir, most persistent, resultant, variability in percent
Visibility	24-h average, max h, min h
Inversion	Height, base (lower height), intensity, duration, a.m. mixing height
Temperature	24-h average, max h, min h, wet bulb or dry bulb
Dew point temp	24-h average, max h, min h
Cloud cover	Average percent
Sunshine	Hours

in this analysis. Also, significant correlations between two meteorological variables may be used to indicate related meteorological conditions (e.g., winds from the south may correlate well with precipitation).

The time periods for the TSP and meteorological measurements may be offset slightly in a second analysis and the resulting correlations compared with those in the first analysis. Higher correlations reveal a time lead or lag in the relationship between TSP and those meteorological variables.

Many studies have employed the correlation matrix as a screening technique to reduce the number of variables to be considered in a subsequent stepwise multiple regression analysis. Available computer programs such as IBM's subroutine STPRG (Scientific Subroutine Package) report the independent variables in the order of their importance and the total variance in TSP concentration at a site due to all the meteorological variables that increase the multiple correlation coefficient. A stepwise multiple linear regression program also calculates an F-value for analysis of variance, to determine the statistical significance of each additional variable in the multiple regression.

The higher the multiple correlation coefficient, the more the meteorological variables explain or relate to TSP concentrations. Also, the analysis can provide an equation relating TSP, the dependent variable, to all the significant meteorological variables. This equation has predictive capabilities.

4.1.2 Applicability of Techniques

If the spatial distribution of sources and receptors remains fixed, then it is assumed that variations in daily TSP concentrations are a function of meteorological parameters and source strengths. Correlation and regression techniques attempt to explain as much of the variations as possible in terms of meteorological conditions. If some emission parameters are included in the correlation matrix, this technique can determine whether these parameters are related to TSP concentrations more or less so than the selected meteorological variables.

These techniques do not characterize the aerosol. A comparison of the most important variables at different sites can lead to indirect, qualitative judgments as to impacting source categories (e.g., fugitive dust sources are more affected by precipitation than other sources). Wind direction as a meteorological variable can reveal directions associated with high and low TSP concentrations. Higher correlations with a time lag may indicate the type of source or relative distances of impacting sources.

No quantitative assessment of the impact of a specific source or of a change in emission rates can be made with these techniques.

4.1.3 Relationship to Other Techniques

Correlation and regression analyses of meteorological data can explain observed seasonal patterns, but not shorter-term patterns or long-term trends.

In analyzing spatial patterns, a correlation matrix may show natural groupings of sites which relate similarly to changes in wind speed, precipitation, temperature, and other meteorological parameters and therefore may be affected by the same air masses. Also, a correlation matrix for meteorological variables may be done simultaneously with intersite correlation analysis. Use of wind direction as a meteorological variable produces an analysis similar to, but potentially more comprehensive than, the pollution rose technique.

Neither emission inventory nor diffusion modeling techniques have obvious relationships to analysis of meteorological variables. However, source emission patterns can be used in a correlation matrix or in stepwise multiple regression along with air quality and meteorological parameters if the emission patterns can be described on a daily basis (e.g., daily traffic volumes or days with coal firing in a dual fuel power plant).

4.1.4 Resource Requirements

Data requirements for these techniques are quite high, with a resulting need for extensive data handling. Resource requirements are as follows:

Manpower	Moderate
Skill	Moderate
Computer	Required
Data	TSP averages, equivalent meteorological variable measurements

4.1.5 Example Application

The example is taken from a study of the impact of field burning on particulate air quality in the Eugene-Springfield area of Oregon.²⁴ Correlation analyses were followed by stepwise multiple regression analyses. Air quality, emissions, and meteorological data for the 1974, 1975, and 1976 field burning seasons were used, with separate runs for each season's data.

Both types of analyses included 31 variables: 8 measures of air quality (3 daily hi vol concentrations, 2 daily average light scattering measurements, visibility, smoke readings, and daily complaints); 10 emission variables (number of acres of fields and number of tons of slash burned per day by quadrant--N, E, S, and W--plus total field burning and total slash burning); and 13 meteorological variables (daily average temperature, rainfall, logarithm of number of days since rain, relative humidity, wind frequencies from each of eight compass directions, and calm winds).

The 31 x 31 correlation matrices by year showed that the three hi vols correlated well (0.70-0.96) with each other for all three seasons. TSP concentrations correlated with light scattering (0.33-0.75) for all sites and correlated somewhat with smoke observations (0.50-0.59), slash burning activity in 1976 (0.16-0.65), logarithm of days since rain (0.45-0.68), and relative humidity (-0.36 to -0.61). There was no correlation in any season between field burning activity and TSP concentrations.

The regression analyses with air quality parameters as dependent variables can be summarized as follows: in all three seasons, air quality variables (TSP, light scattering, visibility, and smoke observations) were most strongly related to meteorological variables. Slash burning had a greater effect than field burning. The only exception was complaints, which were definitely affected by increased field burning. The ranked independent variables for the regression analyses with 1974 data are shown in Table 4-2.

4.1.6 References

13, 16, 18, 24, 29, 33, 34, 35

4.2 DECISION TREE ANALYSIS

4.2.1 Description of Technique

This technique utilizes the AID decision tree program developed at the University of Michigan Institute for Social Research.³⁶ Starting with a large number of daily observations of TSP (dependent variable) and several meteorological variables (independent variables), the AID program sequentially splits the sets of daily observations into two subgroups of an independent variable, at each split choosing the partition of the data that maximizes the difference in TSP concentrations as measured by the residual sum of squares (RSS).^{*} The partitioning process is repeated on successively smaller subgroups of daily observation sets until one of the following circumstances occurs:

$$* \text{ RSS} = \sum_{j=1}^n (A_j - \bar{A})^2, \text{ where}$$

A = TSP concentration for one day

n = number of days of data in subgroup

$$\Delta \text{RSS} = \text{RSS}_{\text{initial}} - \text{RSS}_{G1} - \text{RSS}_{G2}$$

Table 4-2. STEPWISE MULTIPLE REGRESSION ANALYSIS
WITH 1974 DATA

Dependent variable	Independent variables	Cum mult correlation	Sign
TSP Eugene Airport	Rel humidity	0.484	-
	Calm winds	0.576	+
	W winds	0.634	-
	SE winds	0.660	+
	Log of days	0.678	+
	NW winds	0.692	-
	N winds	0.703	+
TSP Eugene Commerce	Rel humidity	0.606	-
	W winds	0.695	-
	Calm winds	0.752	+
	Log of days	0.768	+
	S fields	0.775	+
	W slash	0.784	+
TSP Springfield Library	Rel humidity	0.541	-
	Calm winds	0.660	+
	W winds	0.714	-
	Log of days	0.735	+
	S fields	0.751	+
	NW winds	0.768	-
	W slash	0.783	+
B _{scat} Eugene	W winds	0.383	-
	W slash	0.476	+
	N fields	0.550	+
	Rel humidity	0.593	-
	NW winds	0.628	-
	NE winds	0.664	+
	Calm winds	0.689	+
	N winds	0.702	-
	SE winds	0.709	+
B _{scat} Springfield	W winds	0.400	-
	N fields	0.560	+
	Calm winds	0.602	+
	NE winds	0.641	+
	Av temperature	0.669	+
	NW winds	0.690	-
	Log of days	0.709	+
	Rainfall	0.718	-
	SE winds	0.725	+
Visibility	NE winds	0.340	-
	Rainfall	0.436	-
	N fields	0.519	-
	W winds	0.555	+
	SE winds	0.569	-
	Av temperature	0.581	+
	Total slash	0.590	-
Smoke observations	Av temperature	0.294	+
	SW winds	0.395	-
	N slash	0.438	+
	NE winds	0.461	+
Complaints	NE winds	0.610	+
	N fields	0.662	+
	S winds	0.689	+

Source: Reference 24

- ° No subgroups can be split to achieve a Δ RSS above a preset lower bound, e.g., 1 percent of original RSS of TSP concentrations
- ° Further division would produce a subgroup with less than a preset number of days of data, e.g., 3
- ° The number of terminal subgroups reaches a preset limit, e.g., 10

The independent variables (meteorological parameters) in this program must be represented by discrete numbers. To achieve discreteness, the raw meteorological data are usually divided into ranges. For example, if the variable Z represents wind speed, the investigator may assign the following values for Z:

<u>Z</u>	<u>Wind speed, mph</u>
1	0 - 3
2	3 - 7
3	7 - 15
4	15 - 25
5	> 25

The version of AID used in TSP analyses is simplified so that meteorological variables must be assigned discrete numbers representing monotonic intervals (as in the wind speed example above). Also, the simplified program performs only monotonic splits of the data (i.e., G1 = 1, 2, 3 and G2 = 4, 5 but not G1 = 1, 4 and G2 = 2, 3, 5).

The result of decision tree analysis is a listing of the independent variables in order of importance according to the percent of variance in TSP concentrations that each explains. The analysis also identifies the ranges of meteorological conditions that account for large percent variances in TSP. It has been found that the AID program generally explains more of the variance in TSP data than does multiple linear regression, because it does not involve restrictive assumptions such as linearity or additivity.

4.2.2 Applicability of Technique

Decision tree analysis of meteorological data has the same applications as correlation and regression techniques. It also has the same relationships with other techniques and similar resource requirements.

As with correlation and regression, emission parameters can be included along with meteorological parameters as independent variables in the analysis. This allows determination of relative effects of emissions and meteorology on TSP variance.

The meteorological classes used in AID provide more information on TSP-meteorology relationships than regression coefficients. Also, these meteorological classes can be used to normalize TSP trend data for differing meteorological conditions.

4.2.3 Example Application

The example is an application of the AID program to TSP and meteorological data at 25 hi vol sites in EPA Region VI.³⁷ Data for the analysis were from the years 1973-1975. Eighteen meteorological variables were input:

Month of the year	p.m. average wind speed
Average visibility	thru mixing layer
No. observations blowing	Max temperature
dust	Min temperature
Average wind speed	Amt of precipitation
Resultant wind dir	Max wind speed
Average relative humidity	Three-day accumulated
Wind variability	precipitation
a.m. mixing height	No. of days since last
a.m. average wind speed	precipitation
thru mixing layer	No. of precipitation
	observations

For each meteorological parameter, six to nine data ranges were specified. Termination criteria for the program were the same as discussed previously: $\Delta RSS < 1$ percent RSS_0 , $n < 3$, or 10 terminal subgroups.

An example of the program output is presented in Figure 4-1. The percent variance explained at each of the 25 sites is shown in Table 4-3; values range from 28 to 72 percent and average 51 percent. These are equivalent to total correlation coefficients of 0.53 to 0.85 which average 0.71.

The variables that were found to be most important in explaining TSP variance were month of year (appearing at 16 sites), the two mixing height variables (at least one at 20 sites), number of days since rain (14 sites), wind variability (14 sites), average relative humidity (13 sites), maximum daily temperature (13 sites), resultant wind direction (12 sites), and three-day accumulated precipitation (12 sites).

To provide an assessment of the decision tree technique's performance, multiple regression analyses were also run on the 25 data sets. The percents of TSP variance explained by linear and log-linear multiple regression are presented in Table 4-3 for purposes of comparison. The results indicate that the decision tree technique is considerably better than multiple regression in explaining TSP levels in terms of meteorological variables. The performance of this technique is particularly notable because the decision trees use only a few of the meteorological parameters while the regression equations use all 18 parameters. One possible explanation for the increased capability is that the AID program discerns significant nonlinear dependencies, whereas regressions are limited to linear or log-linear relationships.

4.2.4 References

36, 37

4.3 ANALYSIS OF PRECIPITATION

4.3.1 Description of Technique

In most cases, precipitation is considered to have the greatest impact of the meteorological variables on TSP concentrations. Precipitation has three effects, all of which tend to

FORREST CITY, AR

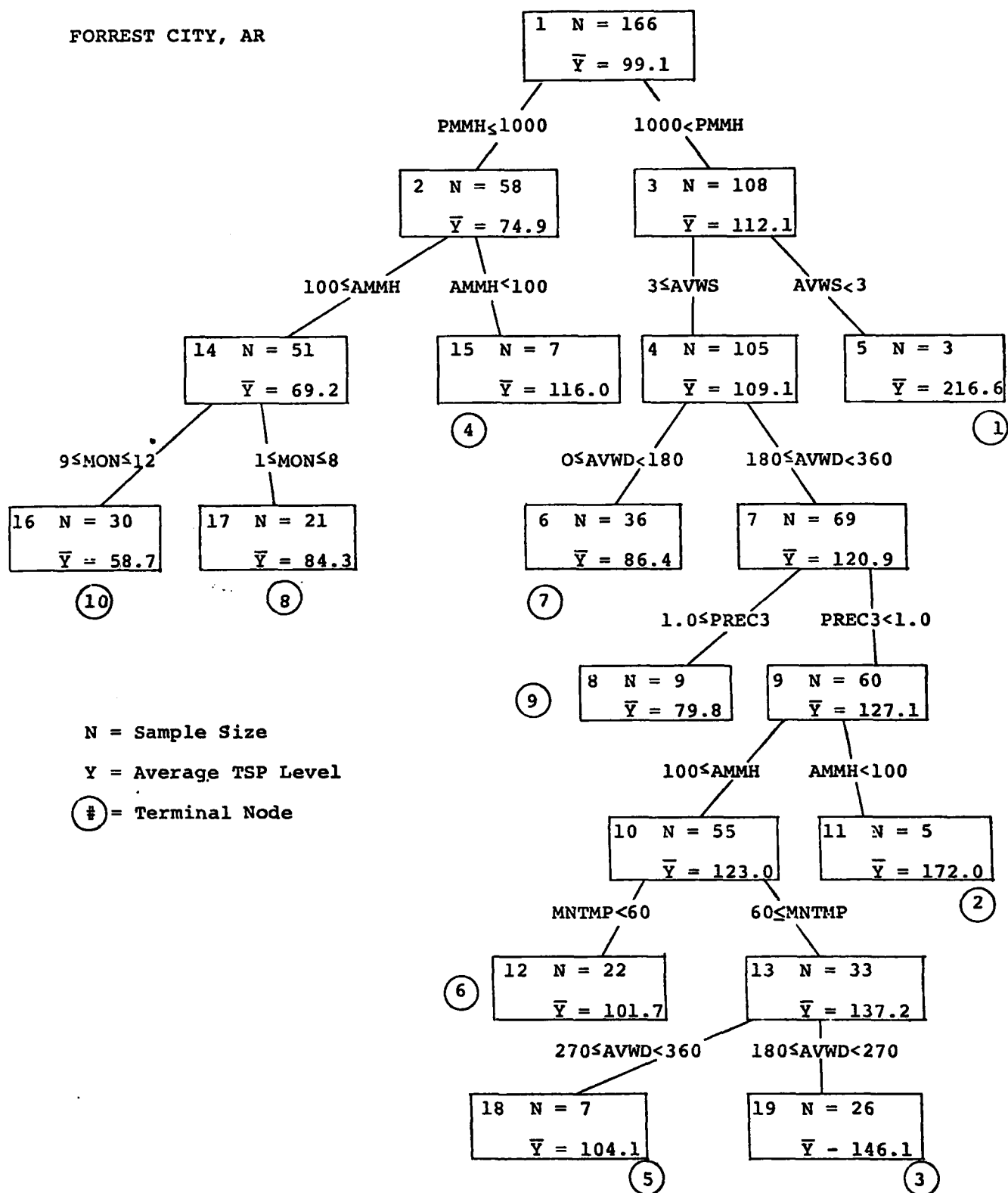


Figure 4-1. AID decision tree for Forrest City, AR.
Source: Reference 37

Table 4-3. PERCENT VARIANCE EXPLAINED BY AID COMPARED TO
PERCENT VARIANCE EXPLAINED BY MULTIPLE REGRESSIONS
AGAINST ALL 18 METEOROLOGICAL VARIABLES

Site	Percent variance explained			
	AID program		Regression	
	15 groups	10 groups	Linear	Log-linear
Forrest City, AR	50%	42%	31%	44%
Helena, AR	60%	52%	31%	42%
Jonesboro, AR	60%	51%	42%	47%
Springdale, AR	63%	52%	38%	43%
Stuttgart (1), AR	61%	53%	29%	41%
Stuttgart (2), AR	67%	62%	49%	52%
Albuquerque (1), NM	70%	62%	56%	45%
Albuquerque (2), NM	73%	59%	42%	31%
Dona Ana, NM	63%	54%	58%	63%
Las Cruces, NM	62%	56%	33%	50%
Raton, NM	52%	44%	19%	23%
Bethany, OK	51%	39%	31%	41%
Muskogee, OK	36%	28%	24%	29%
Oklahoma City, OK	55%	50%	37%	43%
Roger Mills County, OK	47%	45%	32%	40%
Sequoyah, OK	69%	61%	50%	54%
Tulsa (1), OK	69%	59%	28%	29%
Tulsa (2), OK	44%	40%	24%	13%
Corpus Christi (1), TX	50%	43%	28%	18%
Corpus Christi (2), TX	67%	49%	13%	27%
Corpus Christi (3), TX	63%	54%	40%	28%
Harlingen, TX	71%	61%	60%	57%
Lubbock, TX	76%	72%	66%	53%
McAllen, TX	47%	38%	24%	24%
San Benito, TX	59%	55%	20%	40%
Mean	59%	51%	36%	39%

Source: Reference 37

reduce TSP levels: (1) it condenses on particles within clouds (rainout) and washes out particles as it falls; (2) it wets surfaces and thereby suppresses generation of fugitive dust; and (3) it is often associated with frontal passages which bring in relatively clean air.

A large number of analytical techniques and graphical data presentations have been employed to quantify the effect of rainfall in reducing concentrations. Some of the more useful techniques are described briefly below.

- Correlation/Regression. As discussed in Section 4.1, one or more precipitation-related variables are usually involved in correlation and regression analyses.
- Daily TSP concentration plotted against amount of precipitation. Time periods for the precipitation values can be the sampling day, the day before sampling, the previous 48 hours, sampling days with no rain on prior day, and weekday or weekend sampling days only.
- Comparative frequency distributions of TSP concentrations for rain/no rain data subgroups. Possible subgroups are no rain within 48 h, no rain within 7 days, rain $>.01$ in. within 48 h, rain $>.10$ in. within 48 h, and rain $>.25$ in. within 48 h.
- Average TSP concentration plotted against days since rain. If TSP data from several sites are used, they should be normalized. This analysis provides better results if hi vol samples are obtained daily rather than every sixth day. Different amounts of minimum rainfall can be specified (e.g., days since .25 in. rainfall) to produce a series of curves. The effect of different rainfall intensities is shown by the interval between the curves.
- Time plots of average TSP on the same graph as inverse precipitation (e.g., average monthly rain/rain for month), using monthly, quarterly, or annual averaging periods. To the extent that precipitation affects TSP concentrations, the two curves should track one another. A correlation coefficient can measure the correspondence of the two curves.
- Quarterly average TSP concentrations plotted against number of days of rain in quarter.

- ° A precipitation rose. This can be compared with a pollution rose for the same site and may explain some of the apparent directional impacts.

4.3.2 Applicability of Technique

Rainfall is usually associated with a reduction in fugitive dust emission rates. With this association, the relative effects of rainfall at different sites or in different urban areas can be used to identify areas with high fugitive dust impacts. Because of the washout action of rainfall, quantitative relationships between rainfall-related TSP reductions and fugitive dust impact cannot be established.

Analysis of precipitation cannot be used to characterize the aerosol or to estimate the impact of a specific source or of a change in emissions.

4.3.3 Relationship to Other Techniques

Seasonal or monthly TSP patterns can be explained with rainfall analyses. Data for some analyses (such as weekday/weekend, upwind/downwind, or source emission patterns) can be stratified for days with rain and days with no rain. By comparing the two data subsets, some additional information on source contributions can be obtained.

Precipitation roses can be compared with wind roses, pollution roses, element roses, etc. to further clarify directional impacts. With sufficient data, the effect of rainfall on particle size distributions (cumulative or species-specific) can also be assessed.

4.3.4 Resource Requirements

These techniques are relatively simple and do not require extensive data. Resource requirements are summarized below:

Manpower	Low
Skill	Low
Computer	Optional
Data	TSP and various measurements of precipitation

4.3.5 Example Application

The example is an analysis of TSP concentrations versus time since rain. TSP readings for 1974 at two sites in Birmingham were sorted into six classifications of days since rain, then averaged.²¹ To facilitate comparison with results from other areas, the TSP concentrations were normalized by dividing by the five-week running average concentration (samples were taken every day). The results are shown in Figure 4-2. Calculations were performed twice, using alternate amounts of precipitation of 0.01 in. and 0.25 in. to classify a day as one with precipitation. The curves in Figure 4-2 indicate that concentrations return to near normal in one day after rain in downtown Birmingham but are depressed for a second day at an industrial exposure in North Birmingham.

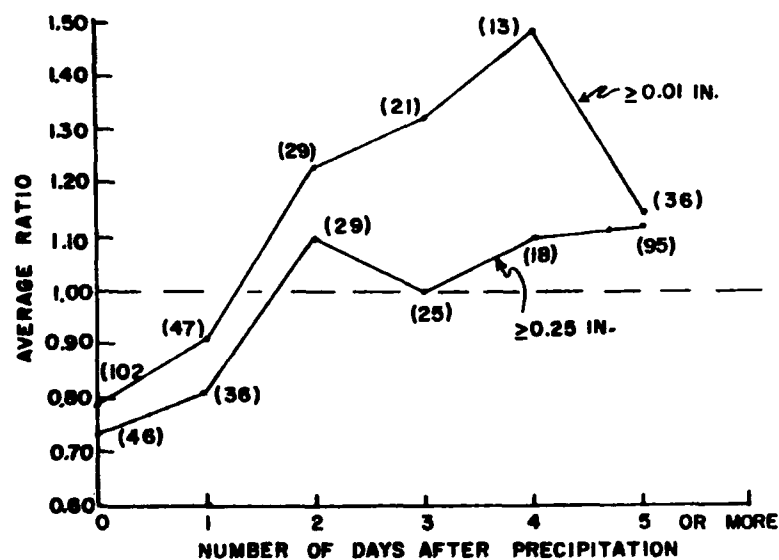
4.3.6 References

11, 12, 13, 15, 16, 21, 24

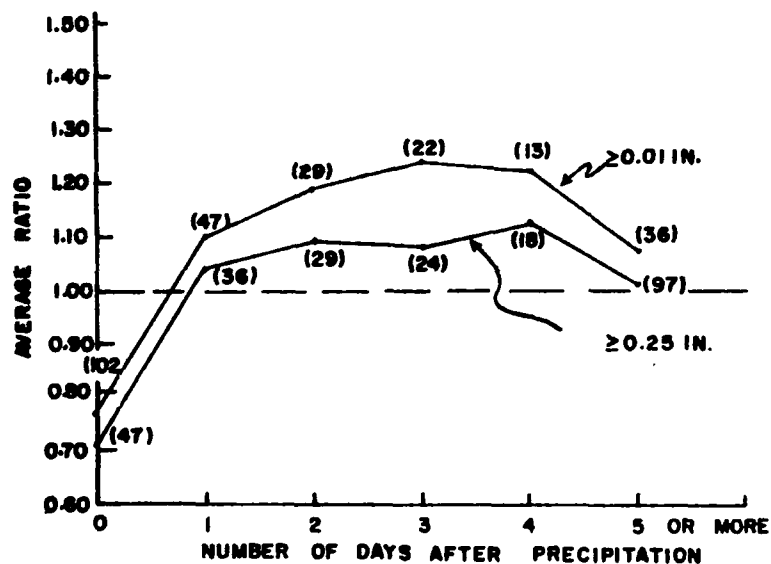
4.4 ANALYSIS OF WIND SPEED

4.4.1 Description of Technique

Wind speed potentially affects TSP concentrations in two contrary manners. As wind speed increases, the effective volume of air available for dilution increases and, for constant source strengths, downwind concentrations are inversely proportional to wind speed. However, emission rates also increase with wind speed because this is the agent by which soil and dust particles are naturally entrained. At instantaneous wind speeds up to 12 or 13 mph, almost no dust is entrained, but at higher speeds the emission rates can become substantial.



a) NORTH BIRMINGHAM



b) DOWNTOWN BIRMINGHAM

Number of observations shown in parentheses

Figure 4-2. Effect of rainfall in reducing TSP concentrations at two sites in Birmingham.

Source: Reference 21

The main interest in analysis of wind speed is to determine the relative effects of dilution and wind erosion generation on TSP concentrations. This is done most simply by plotting TSP concentration versus 24-h average wind speed for all samples at a site or for selected data subsets. The characteristic curve shows high TSP values at extreme high and low wind speeds and lower TSP values with moderate wind speeds.^{15,16,21,37} Distortions from this shape can be interpreted in terms of the relative impact of wind-generated sources.

The analysis can be further refined by plotting TSP concentration versus 24-h average wind speed separately for each wind direction. This analysis provides an indication of wind-generated emissions by direction. One requirement for directional analysis is that only data taken on days with persistent wind directions be used. In this case, wind persistence is defined as a ratio of resultant wind speed to average wind speed equal to or greater than 0.9. Variable wind direction (ratio <0.9) can be an additional subset for plotting TSP versus wind speed.

Wind speed is often used as a meteorological variable in correlation and regression analyses. However, because of the characteristic U-shaped TSP versus wind speed curve, correlations are usually low and may be insignificant. In some studies, TSP concentrations have been correlated with wind speed by speed range.

At least one other technique has been employed to analyze wind speed impact.³⁸ For a set of two sampling sites, one in an urban area and one a background sampler upwind of the city, the ratio of TSP concentrations is calculated for each sampling day. The sampling days are then ranked according to increasing wind speed. If the ratio increases consistently with wind speed, this has been interpreted to mean that extra-urban particulate is a major contributor to concentrations. Conversely, if the ratio decreases consistently, major contributors are located between the two samplers.

4.4.2 Applicability of Technique

As with other meteorological analyses, wind speed techniques assist in identifying source categories but cannot characterize the aerosol, estimate the impact of a specific source, or estimate the impact of changes in emissions from a source.

4.4.3 Relationship to Other Techniques

If wind speed varies substantially on a seasonal or diurnal basis, it may explain some of the temporal TSP patterns. The wind speed variable has been combined with wind direction in some correlation/regression analyses by using total miles of wind blown from each direction as independent variables. This is related to the pollution rose concept.

Because wind speed affects the deposition rate of large particles, an analysis which combines wind speed and particle sizing may provide more information on the impact of wind speed.

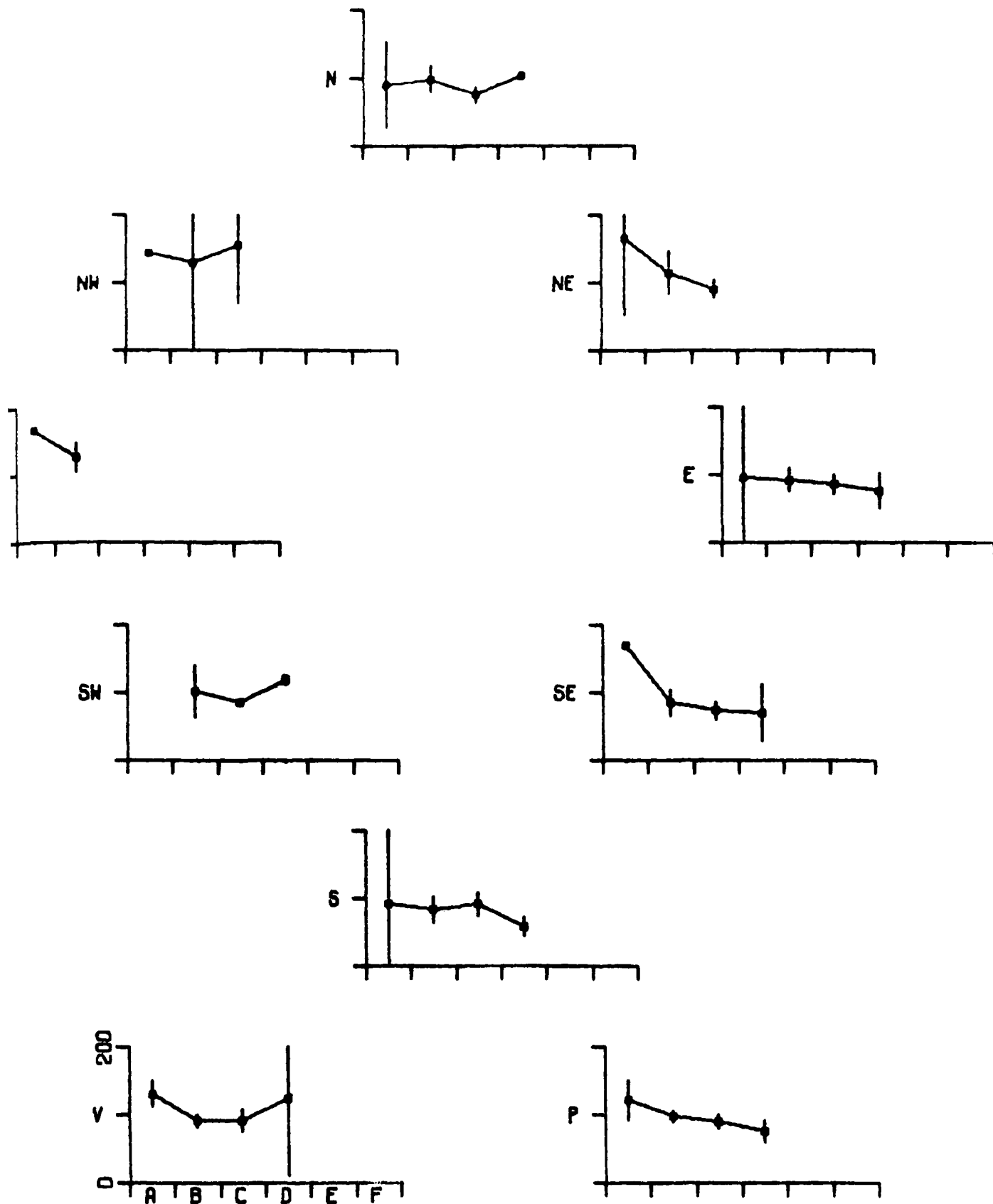
4.4.4 Resource Requirements

Wind speed analysis techniques are relatively simple and do not require extensive data. Resource requirements are as follows:

Manpower	Low
Skill	Low
Computer	Optional
Data	TSP and various wind speed measurements

4.4.5 Example Application

TSP data from Houston and Dallas-Fort Worth were processed into several preliminary displays for further analysis.²² One of these displays was plots of average TSP concentrations versus average wind speed as a function of wind direction. In addition to plots for the eight compass directions at each hi vol site, additional plots were generated for all days with variable winds and for all persistent winds. The plots for one site are shown in Figure 4-3.



AVG TSP VS AVG WIND SPEED

91V SAMPLES 173P SAMPLES
 HIVOL 4060002B NWS 12960
 MONTHS JAN - DEC YEARS 70 71 72 73

Figure 4-3. Plots of average TSP versus wind speed as a function of wind direction.

Source: Reference 22

4.4.6 References

13, 15, 16, 21, 29, 35, 37, 38, 39

4.5 TRAJECTORY ANALYSIS

4.5.1 Description of Technique

This technique attempts to identify the approximate origin of air parcels sampled and to determine portions of the TSP sample that have been transported. A backward trajectory from the sampling site is calculated, usually with upper air (850 or 900 millibar level) rather than surface wind speed and direction measurements.

For 24-h TSP samples, there are actually a number of different trajectories that reach the sampler throughout this time period. However, none of the techniques to date have assumed more than a single trajectory; the resulting air parcel that is traced is assumed to arrive at the site at either the midpoint or end of the 24 hours.

Upper air wind measurements are not taken continuously, so some interpolation between the 12-h interval reported readings must be made. Also, because of the distances involved in 24-h air movements, wind data from several upper air meteorological stations should be used and appropriate values interpolated for locations between these stations. The National Weather Service prepares and distributes via teletype and facsimile circuits 6-h trajectory vectors at the 1000, 850, and 700 mb levels for various stations in the U.S. EPA has a trajectory analysis program available which linearly interpolates between the three closest meteorological stations, computes backward trajectories in 2-h increments, and gives values for the u and v components of the wind at each intermediate location.

After identifying the approximate area of origin of the air parcel, the next step is to find TSP and possibly sulfates and nitrates data at a nearby location. These ambient concentrations should be for the day prior to the downwind sample. If a number

of data pairs of these upwindbackground versus downwind-impacted areas samples are generated, correlations and other comparisons of the data pairs can be performed.

This technique is based on several estimates and assumptions. Therefore, its results should only be used qualitatively.

4.5.2 Applicability of Technique

Trajectory analysis can estimate the portion of a measured TSP concentration due to long-range transport, which helps describe the aerosol. Also, if transported particulate is considered as a source category, it can help to determine contributing source categories. However, it cannot quantitatively estimate the impact of a specific source or the impact of changes in emissions from a source.

4.5.3 Relationship to Other Techniques

Transported particles are those in the small particle size range, generally less than 2 to 3 microns diameter. Therefore, particle sizing and possibly chemical analysis of the <3 um fraction may increase the information gained by trajectory analysis. Also, much of the transported material may be secondary particulate (sulfates, nitrates, ammonium, and organic compounds), so chemical analyses of upwind and downwind samples would aid in establishing the amount of transport.

Trajectory analysis is similar in concept to correlation/regression analyses with wind directions as independent variables. If performed in a comprehensive manner, the latter analyses are much more quantitative.³³

4.5.4 Resource Requirements

Availability of wind measurements that appropriately reflect air mass movements within the mixing layer in the vicinity of the sampling site is the key to this technique. Evaluation of data by a qualified meteorologist is a prerequisite. Other resource requirements can be summarized as follows:

Manpower	Moderate
Skill	Moderate
Computer	Probably required
Data	TSP and upper air wind speed and direction measurements

4.5.5 Example Application

This "descriptive" trajectory analysis was done after a correlation analysis of TSP concentrations with wind direction in Albany, New York showed that higher particulate concentrations were associated with southerly winds and that lower concentrations were associated with winds from the northwest.³⁹ Twenty-four hour trajectories for 10 days during the study period, representative of various wind directions, were generated using National Weather Service 6-h vectors. In all 10 cases, predicted trajectories were verified satisfactorily by comparison with local wind records for the days in question.

The resulting trajectories and corresponding TSP concentrations are shown in Figure 4-4. They showed that air masses from the south had passed through the Washington, D.C.-Philadelphia-Allentown-Bethlehem areas or New York-Long Island during the preceding 24 hours, and that air masses from the Northwest passed through low population areas of Upper New York State and Canada.

4.5.6 References

33, 38, 39, 40

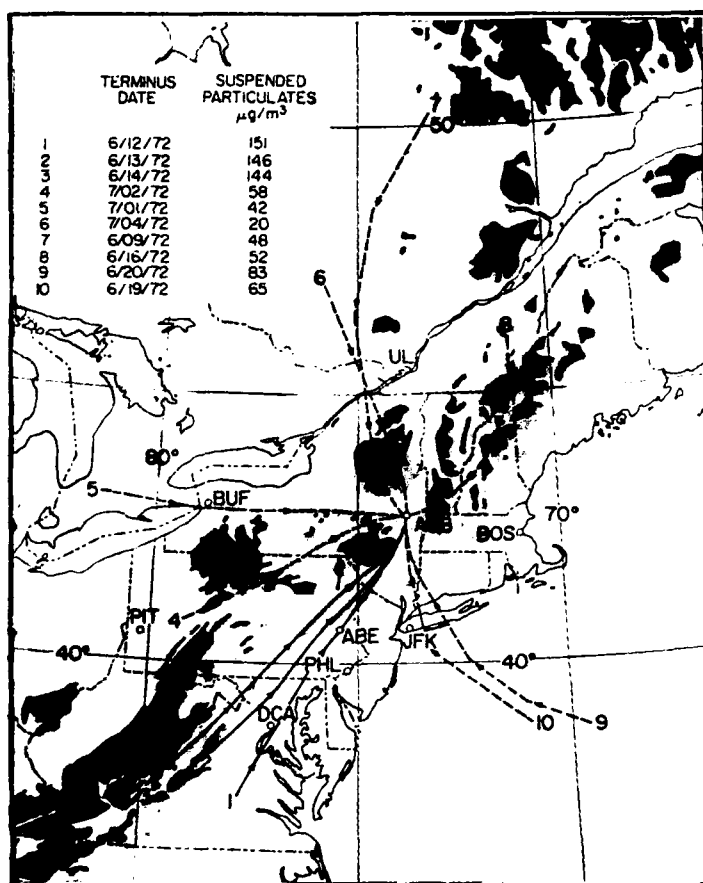


Figure 4-4. Air flow trajectories for 10 nonprecipitation days terminating in Albany.

Source: Reference 39

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5.0 ANALYZING EMISSIONS DATA

Previous chapters have presented several techniques for analyzing suspended particulate data. Such techniques can provide evidence implicating source categories possessing certain emission characteristics, but additional techniques must be used to quantify those emission characteristics. In this chapter three such techniques are discussed:

- Emission inventorying
- Microinventorying
- Diffusion modeling

5.1 EMISSION INVENTORYING

5.1.1 Description of Technique

Procedures for compiling an emission inventory are amply described in a number of EPA publications and need not be repeated in detail here.^{28,41,42,43} In brief, the technique is to inventory the major sources of particulate emissions in terms of their magnitude, degree of control, and location.

Emission sources have been classified a number of different ways. One of the most common has been to use the following classification scheme:

- Industrial process
- Fuel combustion
- Transportation
- Electricity generation
- Incineration
- Miscellaneous

Table 5-1 lists subdivisions for each of these source categories and the type of source (point, area, or line) included in each. Point sources are any stationary source emitting more than some designated minimum (usually 25 ton/yr) of particulate matter. An area source is a collection of sources whose individual emission rates are small but whose collective impact may be large. A line source is a source that can be geometrically described best as a line (e.g., a highway). The line source description is used primarily in carbon monoxide microscale analyses and is rarely used in particulate emission inventories.

In addition, there has been a growing trend toward making finer distinctions among various types of "unconventional sources" of particulates. Fugitive emissions are particulate matter emitted from industrial facilities without the benefit of flow or direction control. Fugitive dust, on the other hand, is a type of particulate emission driven airborne by man's activity or by the effect of wind erosion upon exposed surfaces. A summary of the various significant categories of fugitive dust sources is also presented in Table 5-1.

The major deficiencies of the technique include the following:

- ° Emission factors represent typical emission rates and may not apply to the specific sources of concern
- ° Methods for apportioning area and fugitive dust sources to geographical subdivisions of a region are subject to large potential errors
- ° Inventorying, by itself, tells the analyst nothing about air quality; other techniques, such as diffusion modeling, must also be used

5.1.2 Applicability of Technique

Emission inventorying, by itself, does not help to characterize the aerosol. Rather, the fundamental utility of the technique is to identify the major sources and source categories which may contribute to measured or predicted TSP concentrations.

Table 5-1. EMISSION SOURCE CATEGORIES

Source category	Subdivisions	Source description
Industrial process	Chemical manufacture Food/Agriculture Primary metals Secondary metals Mineral products Petroleum industry Wood products Evaporation Metal fabrication Leather products Textiles In-process fuel Fugitive emissions Other	Point, area
Fuel combustion	Internal combustion External combustion	Point, area
Transportation	Highway vehicles Lt duty gas autos Lt duty gas trucks Motorcycles Hvy duty gas trucks Hvy duty diesel trucks Off-highway vehicles Rail locomotives Vessels Aircraft	Area, line
Electricity generation		Point
Incineration		Point, area
Miscellaneous	Solvent evaporation Fires Fugitive dust Anthropogenic (unpaved roads, agri tilling, const activities, street dust, off-road mtr veh activities, inactive tailing piles) Wind erosion (unpaved rds, agri fields, disturbed soil surfaces)	Point, area

Source: Adapted from References 41 and 44.

Recognizing the unique and vital role that this technique plays in any effort to control TSP concentrations, there are still a number of deficiencies associated with this technique that the analyst needs to be aware of. The most important are the following:

- ° The inventory is usually limited to man-related sources and may downplay the role of natural sources. Questions which are still the subject of research concern the roles of naturally occurring windblown dust and sea spray, and secondary particulates transported over long distances.
- ° The types of sources which are inventoried have changed over time as new information has become available (e.g., industrial fugitive emissions and fugitive dust from such source categories as paved roads have only recently been included in inventories).
- ° Actual emissions from industrial facilities may differ from estimated emissions by a rather large amount. Fluctuating loads may cause actual collection control efficiencies to differ from measured efficiencies. Likewise, malfunctions and deterioration in equipment may occur and cause significant deviations from calculated emissions.

This technique is a prerequisite for determining the impact of specific sources upon air quality, for that impact cannot be determined without first identifying the locations and approximate magnitudes of those sources' emissions. Similarly, the technique is a prerequisite for assessing the effect of changes in emissions upon TSP air quality.

5.1.3 Relationship to Other Techniques

Most emission inventory data are prepared in terms of annual emissions. As a result, quantitative comparisons between air quality and emission data are normally possible (depending upon the availability of data) only in terms of annual trends. However, in some cases data on temporal variations in emissions from specific sources or source categories may be available. Techniques for relating short-term variations in air quality and emissions are discussed in Section 1.5.

Emission inventorying relates to the analysis of spatial patterns of air quality, in that upwind/downwind analyses begin with the assumption that a certain source may be an excessive contributor to measured air quality and in that the shapes of pollutant roses initially lead the analyst to check the current emission inventory for major particulate sources which are located in the direction suggested by those shapes.

The major relationships between this technique and the analysis of the effect of meteorological variables relates to rainfall. Certain emission sources, such as fugitive dust sources, vary in magnitude inversely with rainfall. Where rainfall is shown to have a major effect upon TSP values at a specific site, then it could be inferred that fugitive dust sources are a contributing source.

Diffusion modeling is dependent upon emission inventory data since the emission rates from modeled sources are necessary input values.

Emission inventories are also used in the chemical element balance technique to approximate the contribution of sources for which suitable tracers cannot be found. Most industrial processes fall within this category. Likewise, where detailed elemental concentrations of a source's emissions are available, such data can be used with element roses and interspecies correlations at a given sampling site to verify that source's impact upon TSP values at that site. Occasionally, as in the case of windblown dust, data concerning the particle size distribution of a source category is available. When such is the case, sampling site particle size distribution data can be used to implicate major contributing source categories.

5.1.4 Resource Requirements

Resource requirements for this technique are summarized below:

Manpower	High
Skill	Low-moderate
Computer	Optional
Data	Emission factors, source activity rates and locations, climatological data

5.1.5 Example Application

Examples of the application of the emission inventorying techniques can be found in any major city. As a result, a detailed example will not be presented. Rather, one brief example from Philadelphia will be discussed.^{12,30}

The Metropolitan Philadelphia Interstate AQCR is a major industrial area encompassing over 1,300 individual plants in Pennsylvania, New Jersey, and Delaware. National emission data system (NEDS) data for 1972 provided a breakdown for most of the source categories in the AQCR (see Table 5-2). Locations of the 50 largest sources in the Philadelphia area are graphically displayed in Figure 5-1.

The NEDS data did not include any estimate of the emissions from possible fugitive dust sources. The authors used data from other recent studies of dirt roads, dirt airstrips, construction, and agricultural tilling. Subsequent calculations indicated that the fugitive dust sources contributed over 10 times the total tonnage as the traditional inventoried sources. The majority of fugitive emissions were calculated to be contributed by dirt roads and construction activity (see Table 5-2).

5.1.6 References

12, 28, 41, 42, 43, 44, 45

5.2 MICROINVENTORYING

5.2.1 Description of Technique

The term "microinventory" refers to the procedure of estimating annual particulate emissions in a relatively small area surrounding a high volume sampler site. When first applied, this

Table 5-2. NEDS TSP EMISSIONS FOR PHILADELPHIA AQCR

Source category	Emissions, ton/yr		Total
	Point	Area	
Fuel combustion	(133,440)	(31,463)	(164,903)
External	133,410	31,463	164,873
Residential	-	6,334	6,334
Electrical	12,954	-	12,954
Industrial	114,341	19,729	134,070
Commercial-Institutional	6,115	5,400	11,515
Internal	30	-	30
Industrial process	(61,322)	-	(61,322)
Chemical	1,810	-	1,810
Food/Agriculture	1,665	-	1,665
Metals	9,195	-	9,195
Mineral	22,521	-	22,521
Petroleum	26,116	-	26,116
Other	15	-	15
Solid waste disposal	(8,989)	(6,989)	(15,978)
Government	8,651	-	8,651
Residential	-	1,415	1,415
Commercial-Institutional	5	4,906	4,911
Industrial	333	668	1,001
Transportation	-	(23,074)	(23,074)
Gasoline	-	17,857	17,857
Diesel	-	2,912	2,912
Aircraft	-	1,022	1,022
Vessels	-	1,283	1,283
Fugitive emissions	-	(2,808,640)	(2,808,640)
Dirt roads	-	1,954,629	1,954,629
Dirt airstrips	-	2	2
Construction	-	850,594	850,594
Agricultural tilling	-	3,415	3,415
Totals	203,751	2,870,166	3,073,917

Source: Reference 30

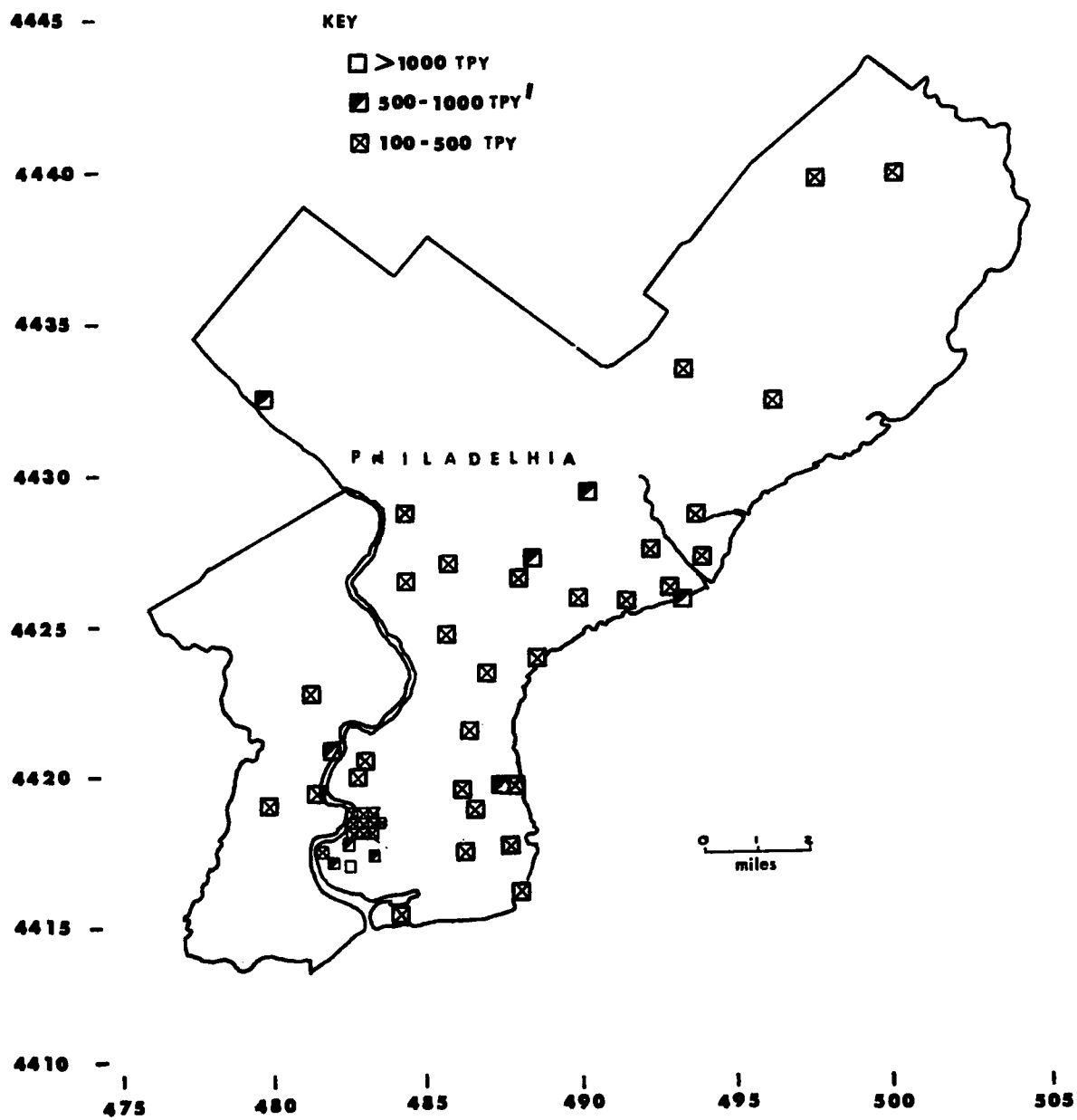


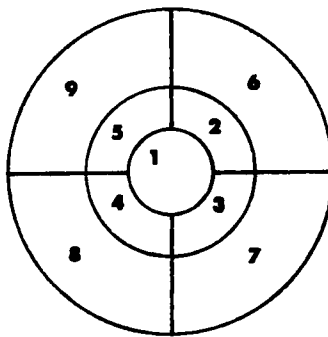
Figure 5-1. Location of major sources.

Source: Reference 12

technique was rather simple, but it has since evolved into a detailed compilation of sources and their distance and direction from a sampler.^{46,47}

Although a relatively small area is inventoried, the area must be large enough to include the location of most of the sources significantly affecting air quality at the site being investigated. The most recent approach uses a five mile radius (from the sampling site) for point sources and one mile for area sources.

Distances and compass directions from the sampling site to impacting sources should also be calculated. For point sources, this is done out to the five mile radius limit. For area sources, however, such an approach is impractical. Instead, the area within a one mile radius of the sampling site is divided into four 90 degree quadrants and three radial distances (as shown below).



Within Sector 1 (1/4 mile radius from the site), the major sources very close to the monitor are described as to their distance and direction from the monitor. Emissions from all sources in that sector are then tabulated. Similar tabulations are made for sources within the 1/4 to 1/2 mile radii and the 1/2 to 1 mile radii with these radii being divided into quadrants. Thus, a total of nine sectors are considered separately for the inventory of area source emissions.

Fugitive dust sources are generally the dominant area source within one mile. These sources include dust from paved roads, unpaved roads, parking lots, cleared areas, construction, rail yards, agricultural tilling, and so on. Industrial processes

smaller than the point source cutoff of 25 ton/yr actual emissions are also included. The area source inventory also includes combustion sources (such as space heating, small boilers, and incinerators), which were not included in the point source inventory, and transportation exhaust sources.

A procedure for determining the locations of the sources and their activity levels should be developed for each urban area to minimize time required in the field. A presurvey is suggested if the sites to be inventoried are not located in the same region as the staff performing the inventory. This presurvey requires a one- to two-day general tour of the sites to be inventoried. Table 5-3 indicates the information obtained during the presurvey.

Following this presurvey, a 1/4 mile radius map of the road network is drawn to scale using USGS maps. Then the distances and directions from the sites to the point sources are calculated. Aerial photos can be used to prelocate or preinventory cleared areas or other fugitive dust sources. The return, or survey visit, consists of a detailed visual inspection of the vicinity of the site. The condition of the streets (dirt level and presence of paving, curbs, etc.) is described, so that appropriate emission factors can be selected later. The location and activity level of unpaved or gravel parking, for example, is estimated by the best available means (talks with the owner or attendant, or estimated lot capacity and turnover rate). The locations of very close point sources (as calculated from UTM coordinates) are confirmed. The mile area is checked for omissions from the point source inventory. Any obvious, unusual characteristics that may not have been considered accurately in the area source allocation scheme (such as a concentration of coal or wood fired space heaters and fireplaces) are noted, and appropriate adjustments are made to the inventory. The survey usually requires two persons for about one-third to one-half day in the field per site, depending upon the complexity of the area and the number of fugitive sources.

Table 5-3. INFORMATION OBTAINED IN PRESURVEY

-
- ° U.S. Geological Survey maps of the area from a local engineering supply store
 - ° Land use maps from the local Council of Governments (COG)
 - ° Traffic data from local COG or city/state transportation agency
 - ° TSP annual air quality data and the site UTM coordinates from the local air pollution control agency
 - ° Gridded inventory of area combustion sources from the local air pollution control agency
 - ° Point source inventory, including annual TSP emissions over 25 ton/yr and UTM coordinates, from the local air pollution control agency
 - ° Precise location of hi vol including height and distance to all major sources (such as roads, parking lots, etc., within 60 m) obtained during site tour
 - ° Photographs of site and surrounding area obtained during site tour
 - ° Aerial photographs (optional) usually available through COG
-

Source: Reference 45

Standard emission factors are then applied to activity factors such as vehicle miles traveled (VMT), acres of tilled land, etc., to compute the emission estimates for each sector.

Predominant land use in each sector can be estimated by visual inspection or through land use maps. For further accuracy, the 1/4 mile radius (Sector 1) can be divided into quadrants with assigned land use classifications. Such information may be useful for future planning and projection activities. Table 5-4 indicates the land use categories and classification criteria which have been found to be useful.

5.2.2 Applicability of Technique

This technique does not contribute to an improved characterization of the aerosol. However, it can qualitatively estimate the relative impacts of fugitive dust sources and point sources upon measured air quality. This can be accomplished by performing intersite comparisons of emissions for sites with equivalent TSP concentrations. One recent application used microinventory data at several sites to produce a multivariable, empirically-based predictive equation.⁴⁵

When combined with standard diffusion models to provide refined detail in the vicinity of receptors with measured TSP data, the technique can be used to assess the impact of a change in emissions from given sources. But, by itself, the technique will neither determine the impact of a specific source or changes in emissions from a specific source.

5.2.3 Relationship to Other Techniques

No attempt has yet been made to compare temporal variations in TSP concentrations with temporal variations of emissions within microinventoried areas, and the state-of-the-art suggests that such comparisons will not be possible except in terms of long-term trends.

The technique is potentially quite supportive of and complementary to techniques which analyze spatial patterns of TSP.

Table 5-4. LAND USE CATEGORIES AND
CLASSIFICATION CRITERIA

Descriptor	Characteristics
Undeveloped	No significant activity; includes parks and pasture land
Agriculture	Active farming
Light residential	Four or less dwelling units/ acre
Dense residential	Greater than four dwelling units/acre
Suburban commercial	Retail businesses in strip development or shopping centers
Central commercial	Central business district
Light industry	Metal fabrication, warehousing, trucking, etc.
General industry	Controlled industrial processes
Heavy industry	Steel mills, foundries, coking, etc.
Airport	Municipal airport

Source: Reference 45

For example, this technique can be used to classify sites by type for stratification by concentration. Likewise, it can be used to search for sources implied by the shapes of pollutant roses. Intersite correlations can be cross-checked with this technique. Where a correlation is high but the microinventories are quite dissimilar, then a reasonable inference would be that sources outside the microinventoried area are the primary cause of the problem. Where correlations are high and microinventoried emissions are equivalent, then sources within that area are probably the primary causes.

Other than being used in conjunction with seasonal variations in activity patterns (in terms of their effect upon fugitive dust emission factors), this technique bears no strong relationship with techniques which assess the effect of meteorological variables.

The resolution of diffusion modeling grid systems could theoretically be improved by using microinventories which are converted to small detailed grid systems as shown in Figure 5-2.⁴⁵ A successful application of this technique has not yet been reported in the literature, however.

No attempts have yet been made to relate microinventory data to particle size, chemical, elemental, or morphological data; however, it is possible (although challenging) that the technique could be refined to use element-specific emission factors to produce element-specific emission totals for the microinventoried areas. These totals could then be correlated with measured concentrations for that element at a series of sampling sites.

5.2.4 Resource Requirements

Resource requirements for the microinventory technique are summarized below:

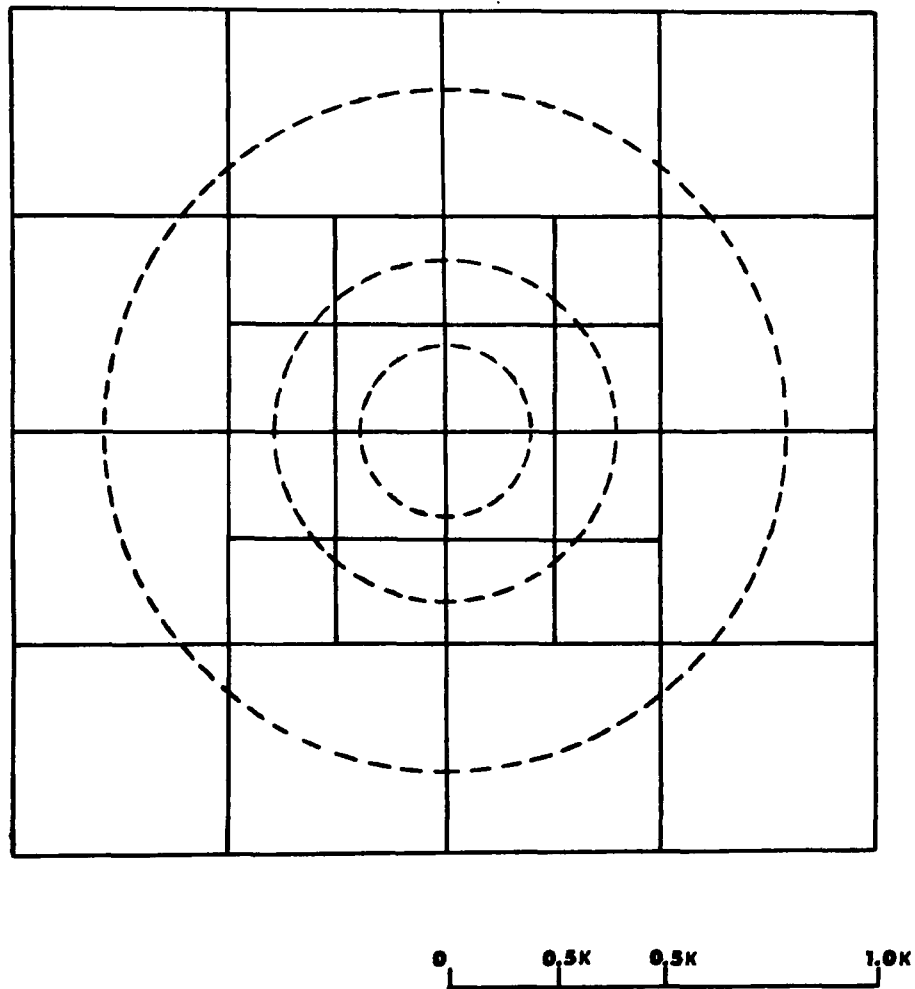


Figure 5-2. Converting microinventory area to diffusion model grid.

Source: Reference 45

Manpower	Moderate-high
Skill	Low
Computer	Not required
Data	Same as for emission inventory plus more detail in vicinity of samplers

5.2.5 Example Application

The example is drawn from a recent report produced for EPA.⁴⁶ The example microinventory is summarized in Tables 5-5 through 5-7 and Figures 5-3 and 5-4 in terms of the following data:

- Description of the site
 - SAROAD code
 - Location
 - Monitor height
 - Land use description by sector
 - Localized sources within 200 feet of the sampler
 - Air quality data
 - A USGS map of the one mile radius area around the sampler
 - A map of the 1/4 mile radius area around the sampler
- Point source summary in terms of emissions, distance, and compass direction from the sampler
- Area source summary in terms of activity rate and emissions by sector for each source category

5.2.6 References

13, 45, 46, 47, 48, 49, 50

5.3 DIFFUSION MODELING

5.3.1 Description of Technique

An atmospheric simulation model can be defined as a mathematical description of the transport, dispersion, and transformation processes that occur in the atmosphere. In its simplest form, such a model relates pollutant concentrations (x) to pollutant emission rates (Q) and a background concentration (b), as in the following equation:

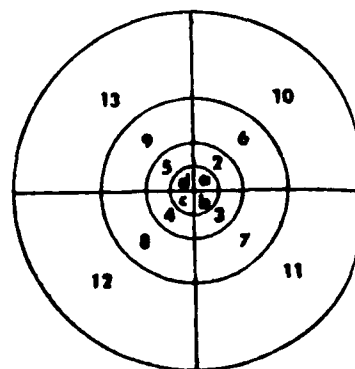
Table 5-5. DESCRIPTION OF MICROINVENTORY SITE

DESCRIPTION OF SITE

SAROAD code - 26 2380 005 H01

Location - 6402 East 37th,
Kansas City, Missouri

Monitor height - 35 ft, on roof of
fire station



Land use, by sector -

1a light industry	6 light industry
1b light residential	7 light industry
1c undeveloped	8 dense residential
1d light residential	9 dense residential
2 light industry	10 general industry
3 general industry	11 dense residential
4 undeveloped	12 dense residential
5 light residential	13 dense residential

Localized sources, within 200 ft of monitor -

<u>Source</u>	<u>Distance</u>	<u>Description</u>
Stadium Drive	55 ft	7870 ADT, dirty, uncurbed
Unpaved parking lot	105 ft	0.1 acres, 10 cars
Unpaved alley	100 ft	5 cars
Fremont	60 ft	75 ADT, dirty, uncurbed

Air quality data -

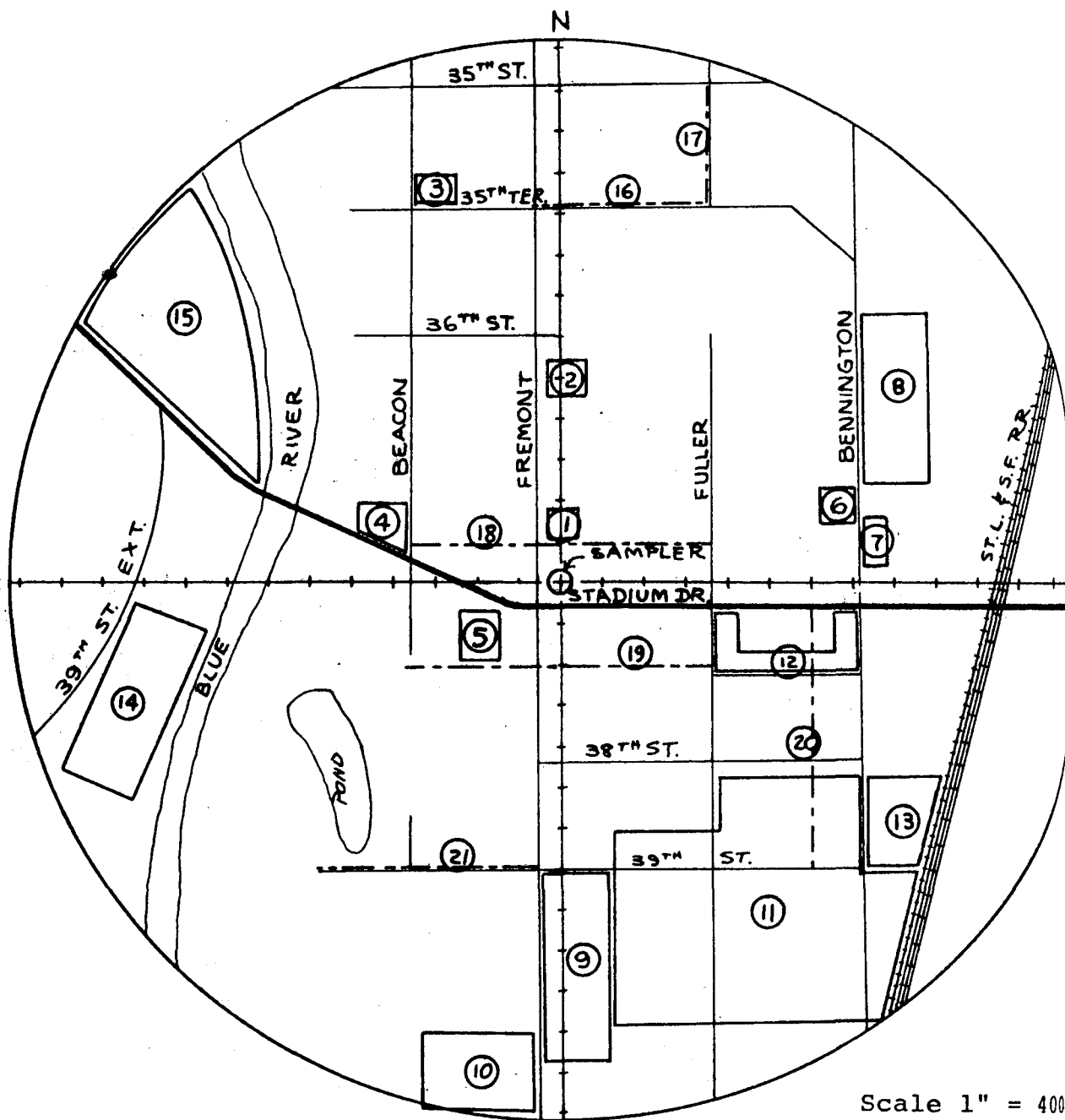
<u>Year</u>	<u>Annual geometric mean, ug/m³</u>	<u>No. of samples</u>
1977	85	-
1976	89	53
1975	86	54
1974	89	47
1973	101	51

Source: Reference 46



Figure 5-3. One mile radius around sampling site.

Source: Reference 46



- | | |
|---------------------------------------|--------------------------------------|
| 1,7. Unpaved pkg lot, 0.1 ac, 10 cars | 12. Unpaved pkg lot, 0.8 ac, 20 cars |
| 2. Cleared area, 0.1 ac | 13. Unpaved pkg lot, 0.7 ac, 50 cars |
| 3,6. Unpaved pkg lot, 0.1 ac, 5 cars | 14. Unpaved storage area, 3.4 ac |
| 4. Unpaved storage area, 0.2 ac | 15. Unpaved storage area, 2.3 ac |
| 5. Unpaved pkg lot, 0.3 ac, 25 cars | 16. Unpaved road, 400 ft, 5 cars |
| 8. Unpaved pkg lot, 1.7 ac, 35 trucks | 17. Unpaved road, 300 ft, 10 cars |
| 9. Unpaved pkg lot, 1.5 ac, 150 cars | 18,19. Unpaved alley, 750 ft, 5 cars |
| 10. Unpaved storage area, 2.0 ac | 20. Unpaved road, 675 ft, 5 cars |
| 11. Unpaved storage area, 8.1 ac | 21. Unpaved road, 600 ft, 5 cars |

Figure 5-4. One-quarter mile radius around sampling site.

Source: Reference 46

Table 5-6. MICROINVENTORY POINT SOURCE SUMMARY

POINT SOURCE SUMMARY

Site: 6402 East 37th

Plant number	Emission level, t/yr	Distance from site, mi	Compass direction, °
26	48	.39	180
25	893	1.7	35
24	98	2.4	25
23	43	2.7	20
22	1,233	3.5	10
16	249	4.3	5
15	41	4.4	0
13	341	4.7	350
14	327	4.8	0
9	46	5.1	330

Source: Reference 46

Table 5-7. MICROINVENTORY AREA SOURCE SUMMARY

AREA SOURCE SUMMARY

Site: 6402 East 37th

Source category	Activity rate	Emissions by sector, ton/yr									
		1	2	3	4	5	6	7	8	9	Total
COMBUSTION:											
Residential fuel	9491 pop	0.2	0.1	0.1	0.1	0.1	0.5	0.5	0.5	0.5	2.6
Comm/Ind fuel	8081 emp	0.5	0.4	0.4	0.4	0.4	1.5	1.5	1.5	1.5	8.1
Incinerators											
Rail/Air											
Auto exhaust	97.4x10 ⁶ VMT	1.9	0.6	0.6	0.2	0.6	27.0	16.7	3.2	13.5	64.3
INDUS PROCESSES:											
Ind processes	4 sources	1		10					1.6	1.0	13.6
FUGITIVE DUST:											
Railroad yards	78 ac	0.4	2.0	1.2			3.0	0.5	0.7		7.8
Clean streets	94.5x10 ⁶ VMT	10.6	3.6	3.6	1.4	3.6	152.3	95.6	14.1	70.8	355.6
Comm streets	3.0x10 ⁶ VMT	1.6					9.6		19.1	19.1	49.4
Unpaved roads	2.7x10 ⁴ VMT	1.2									1.2
Cleared areas	0.1 ac	neg									
Construction											
Agriculture											
Storage areas	16 ac	3.2									3.2
Unpaved pkg lots	25.4 ac	4.8					12.0		7.4		24.2

Recap	Emissions by sector, t/yr			
	1	2-5	6-9	Total
COMBUSTION	2.6	4.0	68.4	75.0
IND PROCESSES	1.0	10.0	2.6	13.6
FUGITIVE DUST	21.8	15.4	404.2	441.4
Total	25.4	29.4	475.2	530.0

$$x = kQ+b \quad (\text{eq.2})$$

The constant k is a function of atmospheric conditions and the spatial relationship between source and receptor. Atmospheric simulation models are ultimately concerned with the variability of k , and of emission rates and their impact on pollutant concentrations.

Modeling procedures have been generally categorized into four generic classes: Gaussian, numerical, statistical, and physical.⁵¹ Gaussian models are generally considered to be state-of-the-art techniques for estimating the impact of non-reactive pollutants such as total suspended particulates.

The extent to which a specific model is suitable for use depends upon a number of factors, among which are the following:

- The detail and accuracy of the data base
- The meteorological and topographic complexities of the area
- The technical competence of the persons performing such modeling
- The resources available
- The situation being modeled (i.e., the type and number of sources, and the time frame of concern)

For assessing the impact of point sources over all typical averaging times, a number of models are available. These models are summarized in three recent EPA publications, and need not be repeated in detail here.^{51,52,53} Where refined analyses are required and no significant meteorological or topographical complexities exist, EPA has recommended the use of the Single Source (CRSTER) Model. Where such complexities do exist, EPA has recommended that each complex situation be treated on a case-by-case basis with the assistance of expert advice.

The Climatological Dispersion Model (CDM/CDMQC), the Air Quality Display Model (AQDM), and the Texas Climatological Model (TCM) have been recommended for evaluating the long-term impact of urban multisource complexes. In the case of multisource

short-term average situations, the Gaussian Plume Multiple Source Air Quality Algorithm (RAM) and the Texas Episodic Model (TEM) have been recommended. The statistical conversion mechanisms of CDM and AQDM can also be used in urban multisource areas; however, their use has not been recommended for situations dominated by large point sources.

AQDM, CDM, and the other above-described models may also be inappropriate for areas which may be dominated by fugitive dust sources. In such locations, models such as the Hanna-Gifford Model, a modified CDM/Rollback model, the Atmospheric Transport and Diffusion Model, or other techniques may be more applicable.²⁸ A fact which must be considered is that models with a wide applicability (such as AQDM and CDM) are not generally available for dealing with long-range transport, deposition, windblown particulate matter, and certain unique meteorological circumstances.

Although specific model impact requirements vary with the model being applied, the general input requirements can be classified as source, meteorological, receptor, and background air quality data. Typical point source emission data includes the emission rate, stack height, stack diameter, stack exit velocity and temperature, and location. Typical area source data include the emission rate prorated over a predetermined rectangular grid network, a representative average emission height, and locational coordinates. Meteorological data typically include such variables as wind direction, wind speed, atmospheric stability, and mixing height. Efforts must be made to assure that these meteorological data are as representative of the transport and dispersion conditions in the modeled area as possible. Receptor sites are usually chosen to provide an adequate degree of spatial resolution to TSP values and/or to provide an estimate of the peak concentration that would be caused by the modeled source(s).

It is commonly assumed that the annual mean background TSP concentration is 30-40 $\mu\text{g}/\text{m}^3$ over much of the Eastern United States. However, such an assumption is not necessarily valid for

short-term situations or for other parts of the U.S. Methods for determining appropriate background concentrations are discussed in a recent EPA publication.⁵¹

Once estimated values are generated, the applied model should be validated and/or calibrated. As one aspect of the validation process, statistical methods including skill scores, contingency tables, correlation analysis, time series and spatial analysis, and others are often attempted. Calibration of a model, the process of identifying systematic errors and applying a correction factor, usually involves the application of regression analysis or a similar statistical technique. The statistical reliability of such procedures is limited by uncertainties associated with input variables and the normally small number of "calibration" sites. Calibration may be the only alternative for improving estimated concentrations, but it should only be applied to long-term models at the present time.

5.3.2 Applicability of Technique

Commonly used models characterize the aerosol only in terms of absolute TSP concentrations over varying time periods at a number of receptor locations. However, there has been a recent trend toward developing models which incorporate a fallout function for larger particles.²⁸

In theory, one of the attributes of the diffusion model is its ability to calculate the contribution of various source categories to predicted air quality. This capability is limited in practice, however, by the facts that not all major emission sources categories are always identified (e.g., street dust prior to circa 1975), that the emission factors and activity rates associated with those sources are of varying degrees of accuracy, and that the procedures for allocating area source emissions to gridded areas can be subject to significant error.

One of the most important uses of diffusion modeling is to assess the air quality impact of emissions or changes in emissions from a specific source or identified set of sources. The

increasing use of diffusion models, and the limitations associated with the other techniques discussed in this digest, suggests that there are no better techniques currently available for performing this task.

5.3.3 Relationship to Other Techniques

Diffusion modeling, largely because of the resources required for implementation, is rarely related to techniques which analyze the temporal patterns of particulate air quality. There is, however, a relationship with those techniques which analyze spatial patterns. It has already been noted that most diffusion models assume level terrain conditions. Other techniques, such as intersite correlations and pollution roses, help to compensate for the errors this assumption induces by providing supplementary information. No quantitative relationships have yet been developed, though.

Meteorological conditions are, of course, used as input variables to diffusion models, but they bear no other significant relationship to that technique. Similarly, emission inventory data are required as a model input but bear no other distinct relationship. Microinventories have the potential of providing refined emission data in the immediate vicinity of samplers.

In one recent study, the Hanna-Gifford diffusion model was used as an ingredient in the development of a new "factor model" of air pollution.⁵⁴ No other relationships between diffusion modeling and physical, elemental, chemical, or morphological data have been demonstrated. The techniques provide dissimilar ways of approaching the same problem and can be used to supplement each other.

5.3.4 Resource Requirements

Resource requirements for diffusion modeling vary considerably depending upon the specific model used. Screening procedures discussed in a recent EPA publication are quite simple

and are not resource intensive.⁵¹ More sophisticated models, such as the RAM, are much more resource intensive. The following summarizes the requirements related to urban multisource models:

Manpower	Moderate ^a
Skill	High
Computer	Necessary
Data	Emission inventory, meteorological data, TSP measurements

^a Assumes that emission inventory data have already been obtained.

5.3.5 Example Application

Examples of the application of specific diffusion models can be found for any major city. What follows is a very brief description of how one model was applied to a hypothetical situation. The example, an application of AQDM, is drawn from the EPA publication entitled Air Quality Analysis Workshops: Volume I--Manual.⁴⁴

The Air Quality Display Model (AQDM) was used to perform the air quality analysis for County X. Figure 5-5 illustrates the particulate air quality computed for 1975 and 1985 as plotted by a computerized routine. Table 5-8 is an example of the computed air quality from the AQDM output tables. In making these computations, it was assumed that no new control programs were in force. Only existing regulations and Federal new source performance standards were assumed to be in effect. Source compliance data, where available, were used to determine actual emissions.

It is evident from Figure 5-5 that there were several areas in the county exceeding the primary NAAQS for particulates in 1975 and that there were much more widely spread violations of the secondary standard of 60 ug/m³. By 1985, growth and development has caused significant increases in the geographical extent of primary and secondary standard violations.

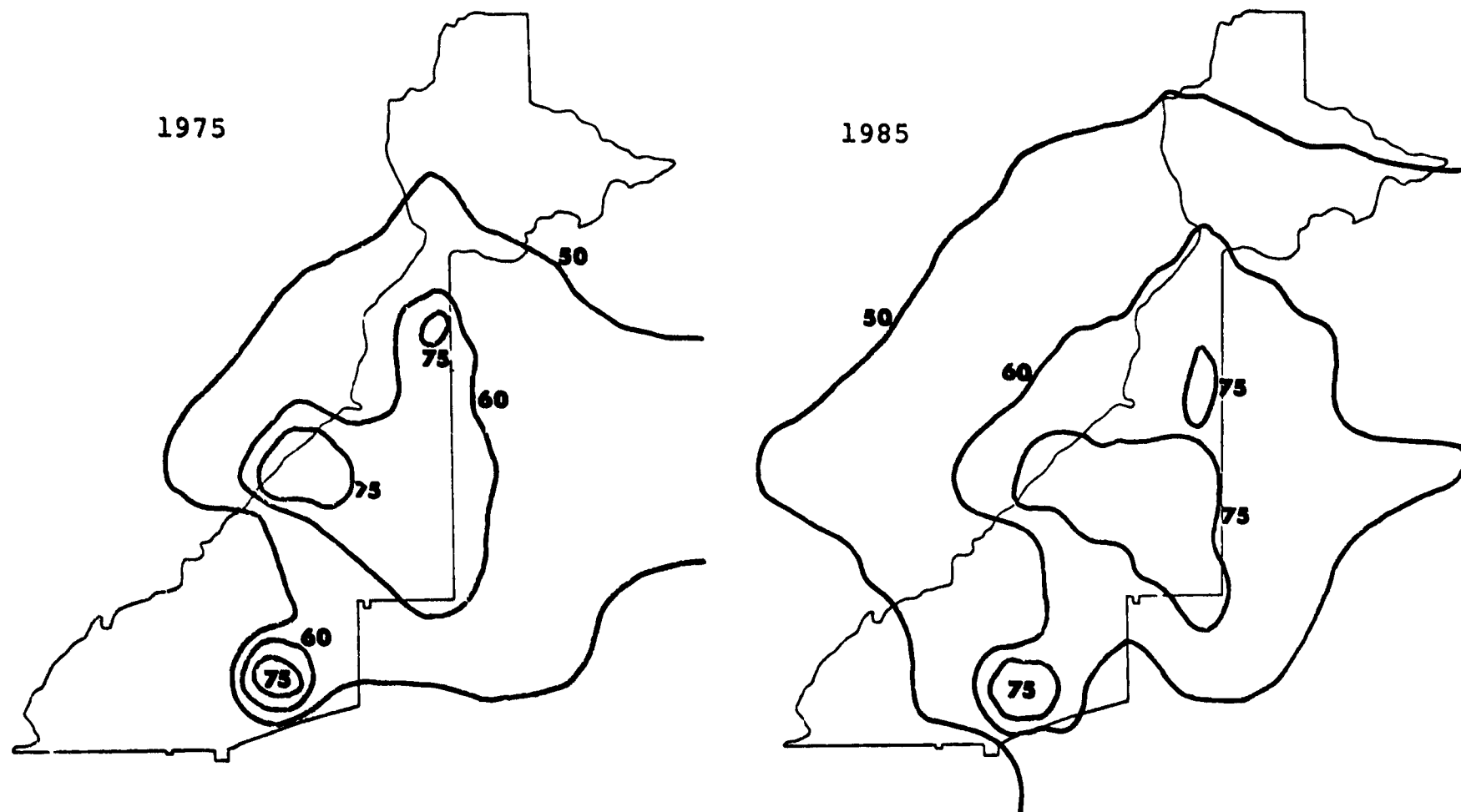


Figure 5-5. County X particulate air quality in 1975 and 1985.

Source: Reference 44

Table 5-8. COMPOSITE OF COMPUTED AIR QUALITY FOR
COUNTY X FROM AQDM

RECEPTOR CONCENTRATION DATA					
RECEPTOR NUMBER	RECEPTOR LOCATION		EXPECTED ARITHMETIC MEAN (MICROGRAMS/CU. METER)		
	(KILOMETERS)		1975 PARTICULATES	1980 PARTICULATES	1985 PARTICULATES
	HORIZ	VERT			
1	700.0	3710.0	42.	44.	45.
2	700.0	3715.0	43.	45.	46.
3	700.0	3720.0	43.	47.	47.
4	700.0	3725.0	44.	47.	47.
5	700.0	3730.0	44.	49.	48.
6	700.0	3735.0	45.	50.	50.
7	700.0	3740.0	46.	50.	50.
8	700.0	3745.0	46.	50.	50.
9	700.0	3750.0	45.	48.	49.
10	700.0	3755.0	44.	48.	48.
11	700.0	3760.0	44.	47.	47.
12	700.0	3765.0	43.	46.	46.
13	700.0	3770.0	43.	46.	46.
14	700.0	3775.0	42.	45.	45.
15	700.0	3780.0	42.	44.	44.
16	700.0	3785.0	42.	44.	44.
17	705.0	3710.0	43.	45.	46.
18	705.0	3715.0	43.	46.	47.
19	705.0	3720.0	44.	47.	48.
20	705.0	3725.0	44.	48.	48.
21	705.0	3730.0	45.	49.	49.
22	705.0	3735.0	46.	51.	51.
23	705.0	3740.0	47.	52.	52.
24	705.0	3745.0	47.	51.	51.
25	705.0	3750.0	46.	50.	50.
26	705.0	3755.0	45.	49.	49.
27	705.0	3760.0	44.	48.	48.
28	705.0	3765.0	44.	47.	47.
29	705.0	3770.0	43.	46.	46.
30	705.0	3775.0	43.	46.	46.
31	705.0	3780.0	42.	45.	45.
32	705.0	3785.0	42.	44.	44.
33	710.0	3710.0	44.	47.	48.
34	710.0	3715.0	44.	48.	49.
35	710.0	3720.0	45.	49.	49.
36	710.0	3725.0	45.	50.	50.
37	710.0	3730.0	46.	50.	51.
38	710.0	3735.0	48.	53.	53.
39	710.0	3740.0	49.	55.	55.
40	710.0	3745.0	48.	53.	53.

Source: Reference 44

An analysis of the TSP contribution by the various sources at five of the receptors used in the example is given in Table 5-9.

5.3.6 References

11, 13, 28, 44, 51, 52, 53, 54

Table 5-9. SOURCE CONTRIBUTION ANALYSIS FOR COUNTY X

																	Point Source Compliance Status		
			Receptor 119			Receptor 132			Receptor 151			Receptor 171			Receptor 233			Baseline Emissions T/yr	Allowable Emissions ^a T/yr
			1975	1980	1985	1975	1980	1985	1975	1980	1985	1975	1980	1985	1975	1980	1985		
Calculated Air Quality (ug/m ³)			70	86	86	57	66	68	73	82	83	55	62	63	77	84	86		
% Reduction for:																			
Primary Standard			-	13	12	-	-	-	-	8	10	-	-	-	3	11	13		
Secondary Standard			15	30	30	-	10	12	17	26	28	-	3	5	22	29	31		
Source Contributions (%)																			
<u>Points</u>																			
Stone Quarry	#1		1.7	2.0	2.0	8.4	10.0	9.8	2.6	4.4	4.3	1.3	1.6	1.6	3.0	4.7	4.6	7620	6265
	2					1.2	1.4	1.4	3.2	5.2	5.0	6.1	7.5	7.4	1.8	2.3	2.2	7297	4130
	3					1.6	1.8	1.8										5495	3115
	4		1.5	1.7	1.8		1.0		2.0	2.6	2.6	1.4	1.7	1.7	1.9	2.4	2.4	5896	3847
	5		27.2	35.1	32.4	13.7	18.0	16.1	9.5	13.6	12.2	4.9	6.6	5.9	12.5	17.3	15.4	16350	9240
	6								1.4	1.3	1.2	8.0	8.0	8.9	1.0			5280	2974
	7				1.1	3.2	2.8	3.7	1.0	1.0	1.3			1.0	1.1	1.0	1.4	6315	3570
Steel Mill	#1		2.1	1.3	1.5				7.5	5.1	5.8				3.1	2.0	2.3	3446	3222
	2		1.2						7.8	5.3	6.0				1.5	1.0	1.1	3446	3222
Lead Smelter			1.4						2.2	1.6	1.3				2.7	1.9	1.6	2530	38
Steam Plant	#1		3.2	3.0	3.7				2.2	2.3	2.7				8.0	7.8	9.3	2322	1103
	2		1.0						1.8						1.8			1205	382
Brick Plant	#1		3.6	3.8	3.8					1.1	1.0				1.2	1.4	1.4	3660	74
	2					1.5	1.6	1.6							1.0	1.1	1.1	3660	74
Concrete Plant	#4									1.4	1.4							50	b
	6									1.6	1.6					1.5	1.5	8	b
<u>Areas</u>																			
	#10		2.2	2.4	2.7	5.4	5.3	6.6	1.9	2.0	2.4	1.4	1.5	1.7	2.1	2.2	2.6		
	77														1.4	1.2	1.2		
	83														1.2	1.0	1.0		
	108						1.0	1.0	1.4	1.9	1.8	7.3	9.1	9.0	1.0	1.3	1.3		
	112								1.1	1.5	1.5	5.8	7.3	7.2		1.1	1.1		
<u>Background</u>			42.6	34.9	35.0	52.5	45.2	44.3	41.3	36.8	35.9	55.0	48.4	47.6	41.2	35.6	34.7		
			87.7	84.2	84.0	87.5	88.1	86.3	86.9	86.7	88.0	92.4	91.7	92.0	87.5	86.8	86.2		

^aBased on current SIP regulations.^bIn compliance.

Source: Reference 44

6.0 INTERPRETING CHEMICAL, ELEMENTAL, AND MORPHOLOGICAL DATA

Recent advances in techniques which measure the elemental or chemical concentration (by total mass or particle size range) of the aerosol and the morphology and chemical composition of individual particles have led to concurrent advances in the analyst's ability to characterize the ambient aerosol, identify major contributing sources and source categories, and further define and solve air pollution control problems.

This chapter discusses several techniques that have seen progressively more common usage:

- Temporal, spatial, and meteorologically-affected (TSM) patterns
- Enrichment factor (E)
- Chemical element balance (CEB)
- Interspecies correlations
- Pattern recognition
- Factor analysis
- Interpretation of morphological data

6.1 TEMPORAL, SPATIAL, AND METEOROLOGICALLY-AFFECTED PATTERNS (TSM TECHNIQUES)

6.1.1 Description of Techniques

Depending upon data availability, the TSM techniques described in Chapters 2, 3, and 4 can be applied to elements and chemical compounds. Normally, however, the analyst will not have access to enough data to assess temporal patterns in a meaningful manner. Thus, for the most part, the analysis of temporal variations in element and compound concentrations will focus upon short-term time intervals. Pollution, dosage, gradient roses, and upwind/downwind techniques can easily be adapted to use for

analyzing spatial variations in element and compound concentrations. Intercorrelations within and among sites can also be adapted to such uses (see Section 6.4 for a discussion of interspecies correlations).

Similarly, the techniques described in Chapter 4 for analyzing the effect of meteorological variables can be easily adapted for use with chemical/element data.

6.1.2 Applicability of Techniques

In combination, these TSM techniques characterize the aerosol to the extent that they document the degree to which that aerosol varies in composition over time and space.

Source categories impacting upon a sampling site can be identified to an extent with this technique. Specifically, data on the spatial and temporal variation of elements and their relationships to meteorological variables can be used to investigate hypotheses formed by a preceding analysis of TSP data. Such an analysis may show, for example, that Si, Al, Mg, K, and other elements normally associated with soil dust are covariant over time with TSP. Similarly, a weekday versus weekend analysis may reveal higher concentrations of Pb and Br (automotive-related) on weekdays than weekends.

The impact of a specific source can be assessed through use of the pollution, dosage, and gradient roses, and upwind/downwind techniques discussed in Chapter 3. The only difference is that one investigates the variation in specific elements/compounds rather than in TSP.

The ability of these techniques to assess the effect of changes in emissions has not yet been successfully demonstrated.

6.1.3 Relationship to Other Techniques

As indicated in the previous subsection, there is an integral relationship between these techniques and those discussed in Chapters 2 through 4. Trajectory analysis is especially amenable to analysis for variations in SO_4 and NO_3 concentrations.

These techniques can be combined with various types of diffusion modeling and emission inventorying techniques. For example, upwind/downwind measurements of Pb concentrations have been used in conjunction with basic Gaussian diffusion equations to back-calculate Pb emissions from motor vehicles.⁵⁵ Likewise, elemental concentrations upwind and downwind of urban areas have been used to estimate overall emission rates for those elements.⁵

Spatial variations in concentrations can be used with the chemical element balance technique to assess spatial variations in source contributions.⁵⁷ Particle size distribution data can be used with this set of techniques to substantiate or invalidate the relative impact of fugitive dust sources. In general, this technique is closely related to or supportive of all chemical/elemental/morphological techniques.

6.1.4 Resource Requirements

Resources required to implement these techniques are much greater than those for similar techniques which relate to TSP concentrations due to the much larger data set. As a result, resources required to implement these techniques are largely a function of the availability of standard computer programs and the number of sites and chemicals/elements to be investigated. The following estimates are based upon a 10 site, 20 chemical/element analysis involving the application of only one of the specific techniques (adapted to chemical/element use) described in detail in Chapters 2 through 4:

Manpower	Low ^a
Skill	Moderate ^a
Computer	Necessary
Data	Chemical/element measurements, other measurements dependent upon specific application ^b

^a Does not consider the equivalent laboratory time or skills required to utilize the instrumentation indicated.

^b Specific choice of instrumentation depends upon specific elements investigated.

6.1.5 Example Application

Due to the large number of possible techniques that have been subsumed under this section, four different examples will be presented. One will demonstrate the analysis of diurnal variations in various elements, another two will address the element rose and upwind/downwind techniques, and a fourth will exemplify the use of linear regression as a way of quantifying the relationship between chemical/element concentrations and meteorological variables.

6.1.5.1 Diurnal Variations - The example is drawn from a recent study performed for the New York metropolitan area.¹⁷ Four-hour nucleopore samples were collected six times daily at one site for a period of 13 days (minus invalid data) during December. These samples were then analyzed with X-ray fluorescence (XRF) for Si, S, Ti, V, Mn, Fe, Zn, As, Br, and Pb. In addition, three nucleopore samples of street dust and four of local soil were also collected and analyzed with XRF. The authors suspected that there were major analytical errors for Ti, As, and Mn; thus, those elements are not addressed herein.

Figure 6-1 presents plots of TSP and element data for the sampling period normalized to the first 4-h period (2200-0200) concentration. Since meteorological conditions were identical for TSP and element measurements, any deviation from the TSP concentration pattern could be attributed to either sampling error or differing emission patterns for the sources of the various elements. As Figure 6-1 indicates, Si, V, Fe, and Zn are covariant with TSP. Pb and Br, on the other hand, are covariant with each other but differ significantly from TSP. S shows a unique pattern.

6.1.5.2 Element Rose - The element rose technique was used in a recent study for Miami, Florida.⁵⁸ Samples were collected at three different sites. The wind direction at each of these sites was structured into 36 separate 10 degree groups, and average concentrations for each of 15 elements were calculated.

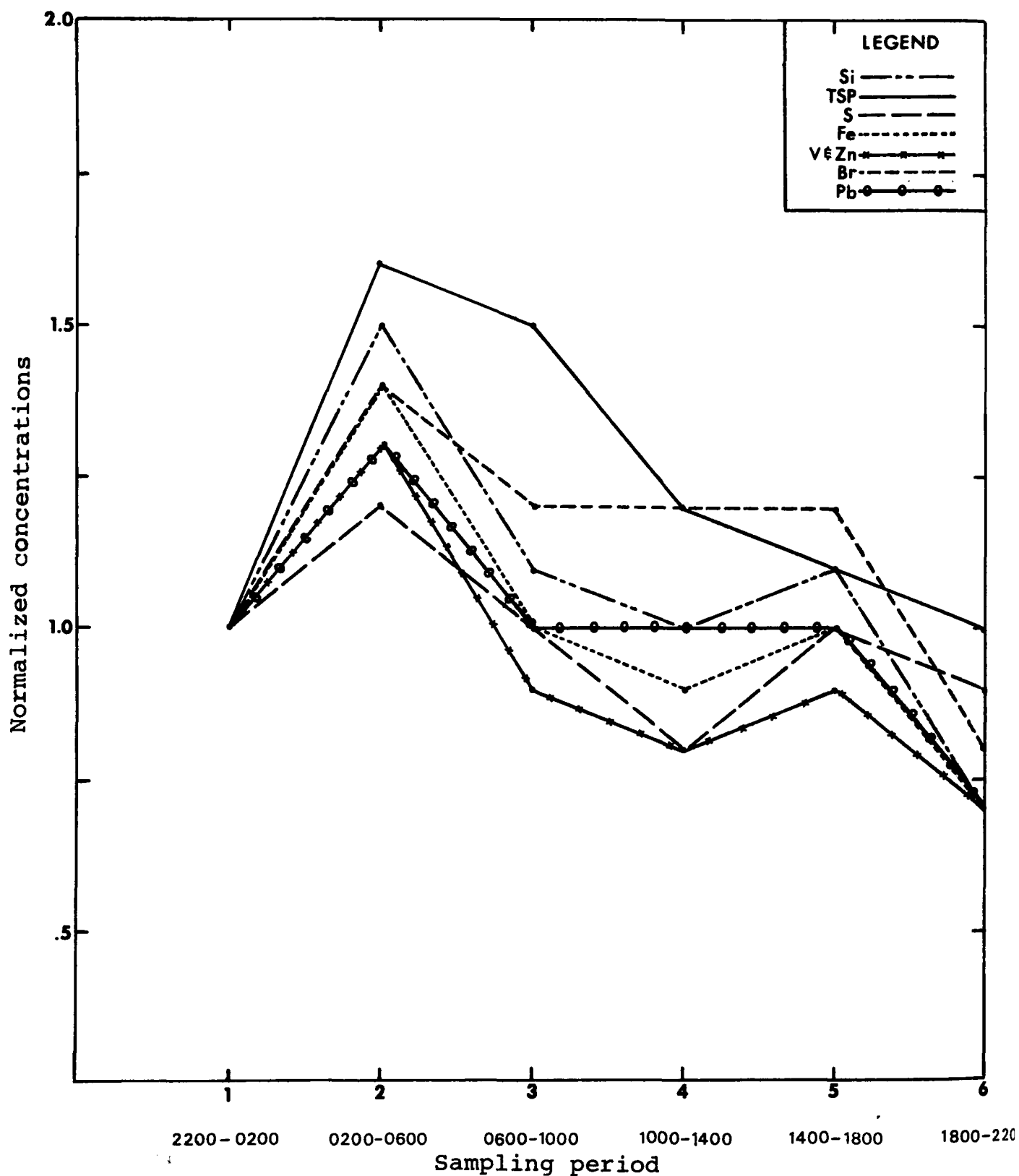


Figure 6-1. Normalized diurnal variation in TSP and selected elements.

Source: Reference 17

According to the authors of the study, the most striking example of the usefulness of the method was provided by the distribution of Pb and Br at a background site. The peak in each case was toward the southwest, pointing toward a large Dade County-operated vehicle service station.

6.1.5.3 Upwind/Downwind Analysis - The example for this technique is drawn from Phoenix, Arizona.⁵⁹ Element data were collected for five 6-h sampling periods at sites upwind and downwind of the Phoenix urbanized area. Four-stage Andersen size fractionating sampler and dichotomous sampler filters were subjected to elemental analysis by X-ray fluorescence.

Downwind samples of two of the six elements investigated (Br, Pb) were generally about twice as great as upwind ones. Elevated values of those elements were attributed to inversion conditions. Cu, Zn, and As concentrations were slightly higher downwind than upwind. It was suspected that mixing of air masses introduced Cu emissions from nearby smelters into the Phoenix area. Fe concentrations were similar at both types of sites.

6.1.5.4 Linear Regression for Meteorological Variables - Drawn from a study of Tucson, Arizona, this example demonstrates the use of linear regression analysis to assess the impact of meteorological variables upon chemical/element concentrations.³⁴ Initial variables included the following:

- Relative humidity (day before sampling)
- Dew point (day before sampling)
- Wind speed (day before sampling)
- Most persistent and resultant wind direction
- Hours of sunshine and cloud cover
- Precipitation (occurrence and amount)
- Maximum, minimum, and average visibility

Samples were divided into two categories according to the two dominant resultant directions. When their chemical/element concentrations were compared, no significant differences were

found. The authors concluded that in order to investigate the effect of wind direction in more detail, shorter sampling periods would be necessary.

Selected meteorological parameters were correlated with chemical/element data at two sites, one urban and one rural. Results of the analysis for the urban site, shown in Table 6-1, indicated to the authors that TSP and Al (and the other soil-related elements) showed a direct dependence upon wind speed. TSP and Al were inversely related with humidity and dew point. No apparent relationship between precipitation and chemicals/elements was revealed. TSP and Al were inversely related with visibility. No significant relationships were demonstrated for non-soil species like SO_4 and Pb. Results of the rural site analysis, shown in Table 6-2, were similar to those of the urban site with two exceptions: at the rural site, the inverse relationship between Al and humidity and dew point was much stronger, and the dependence of TSP and Al with wind speed was less certain. The authors attributed these results to the greater effect of soil moisture upon soil dust than fugitive dust, and to the possible unrepresentative nature of the wind data used for the rural sampling site.

6.1.6 References

17, 22, 30, 34, 55, 56, 57, 58, 59

6.2 ENRICHMENT FACTOR (E)

6.2.1 Description of Technique

The enrichment factor method uses data concerning the elemental concentrations of suspended particulates collected from the atmosphere and of the continental crust or sea to produce estimates as to the degree to which a given element is "enriched" hence produced by non-natural sources.*

* An excellent discussion of the enrichment factor technique can be found in Reference 60. Much of the material herein is based upon that reference.

Table 6-1. DEPENDENCE OF CHEMICAL COMPOSITION ON METEOROLOGICAL PARAMETERS (URBAN LOCATION)

	MASS	Al	SO ₄ ⁼	NO ₃ ⁻
WS	.28(.02)*	.18(.14)	-.11(.40)	-.12(.34)
RH	-.48(.00)	-.55(.00)	.03(.84)	.01(.94)
DP	-.46(.00)	-.39(.00)	-.17(.17)	-.13(.30)
T	.00(.99)	.18(.14)	-.29(.02)	-.12(.34)
P	-.41(.00)	-.54(.00)	.31(.01)	.12(.34)
VIS	-.34(.01)	-.26(.03)	-.21(.09)	-.28(.02)
WS(DB)	.42(.00)	.36(.01)	-.14(.255)	.02(.87)
RH(DB)	-.48(.00)	-.57(.00)	.17(.18)	.06(.66)
DP(DB)	-.52(.00)	-.39(.001)	-.10(.42)	-.05(.69)

	Pb	Zn	Cu	Cd
WS	-.27(.02)	-.15(.24)	-.15(.22)	-.15(.24)
RH	.07(.58)	.07(.58)	.10(.42)	-.03(.82)
DP	-.31(.00)	-.05(.72)	.08(.52)	-.02(.88)
T	-.42(.00)	-.20(.12)	-.06(.66)	.07(.60)
P	.39(.00)	.30(.015)	.26(.05)	.13(.29)
VIS	-.01(.94)	-.10(.45)	-.01(.94)	.00(.99)
WS(DB)	-.21(.10)	-.21(.09)	-.21(.10)	-.16(.20)
RH(DB)	.15(.23)	.06(.66)	.13(.29)	-.04(.73)
DP(DB)	-.25(.04)	-.03(.80)	.09(.50)	.01(.91)

*Values in parentheses indicate the probability of true correlation coefficient being zero (i.e., no correlation exists).

WS: Wind Speed

P: Barometric Pressure

RH: Relative Humidity

VIS: Visibility

DP: Dew Point

WS(DB): Wind Speed (day before sampling)

T: Temperature

RH(DB): Relative Humidity (day before sampling)

DP(DB): Dew Point (day before sampling)

Source: Reference 34

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Table 6-2. DEPENDENCE OF CHEMICAL COMPOSITION ON METEOROLOGICAL PARAMETERS (RURAL LOCATION)

	MASS	Al	SO ₄ ⁼	NO ₃ ⁻
WS	.18(.18) [*]	.20(.13)	-.09(.51)	.10(.46)
RH	-.52(.00)	-.70(.00)	.20(.14)	-.06(.64)
DP	-.50(.00)	-.56(.00)	-.08(.57)	-.14(.32)
VIS	-.14(.31)	-.14(.30)	-.16(.24)	-.19(.17)
WS(DB)	.20(.13)	.28(.04)	-.12(.36)	.17(.21)
RH(DB)	-.40(.00)	-.65(.00)	.32(.02)	-.17(.22)
DP(DB)	-.46(.00)	-.54(.00)	-.02(.86)	-.14(.30)

	Pb	Zn	Cu	Cd
WS	-.13(.35)	-.06(.67)	-.09(.51)	-.12(.38)
RH	.12(.37)	-.12(.39)	.28(.04)	.22(.10)
DP	-.22(.11)	-.08(.55)	.09(.51)	.08(.55)
VIS	-.06(.65)	.00(.99)	.14(.30)	.05(.69)
WS(DB)	.03(.81)	-.03(.81)	-.07(.60)	.00(.97)
RH(DB)	.25(.06)	-.11(.42)	.32(.02)	.26(.05)
DP(DB)	-.19(.17)	-.10(.45)	.09(.53)	.07(.60)

* Values in parentheses indicate the probability of true correlation coefficient being zero (i.e., no correlation exists).

WS:	Wind Speed	WS(DB):	Wind Speed (day before sampling)
RH:	Relative Humidity	RH(DB):	Relative Humidity (day before sampling)
DP:	Dew Point		
VIS:	Visibility	DP(DB):	Dew Point (day before sampling)

Source: Reference 34

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The enrichment factor itself is calculated for the various elements in the aerosol relative to the crust or sea or both, usually normalized to an element considered to be the clearest indicator of the naturally occurring source material. The general formula for such an enrichment factor is:

$$E(X)_{\text{aerosol-source}} = \frac{(X/\text{Ref})_{\text{aerosol}}}{(X/\text{Ref})_{\text{source}} (\text{crust or sea})}$$

where $E(X)_{\text{aerosol-source}}$ is the enrichment factor of the element X in the aerosol, relative to some source material and a reference element; X/Ref is the ratio of the concentrations of element X and the reference element in the aerosol and the source material.

Elements most frequently used as crustal indicators include Si, Al, Fe, and Sc, whereas for sea salt Na is almost always used. An enrichment factor of close to 1.0 indicates that the crust/sea is the primary source of the measured aerosol. A factor significantly greater than 1.0 can normally be interpreted as the result of man-generated emissions.

A number of concepts inherent to the enrichment factor technique are still under active investigation, among which are the following. First, the element fractionation of the crust/sea material as it interfaces with the atmosphere is largely unexplored. What little data are available relate primarily to sea salt fractionation. Second, although average elemental concentrations of crustal rock are normally used for calculation purposes, the true contribution of this natural source is probably much more closely related to the elemental concentrations of soil in the region of aerosol sampling. Few data are available concerning the regional variation in elemental concentrations of soil. Third, the most common crustal reference element, Al, is used frequently in the vicinity of samplers and risks being subjected to biased calculations. Si, a better reference material, has only recently become amenable to reasonably available measurement techniques.

Some limitations to the technique have been emphasized in the literature: first, man-induced fugitive dust (from traffic on unpaved roads, for example) will cause significant amounts of dust, but will not necessarily produce high enrichment factors due to the natural (crustal) origin of the material disturbed by the activity; second, moderate or high enrichment factors in relatively remote areas may be the result of natural soils which differ from crustal rock, or the action of such other material sources as volcanoes, forest fires, or vegetation.

6.2.2 Applicability of Technique

The enrichment factor technique characterizes the aerosol in terms of its elemental concentration relative to the reference material. This is perhaps the second best attribute of the technique. The best attribute is the fact that this technique will provide evidence as to the degree to which anthropogenic sources contribute to ambient concentrations measured at rural or remote sites. In other words, it will provide evidence concerning the nature and impact of long-range transport of man-related source emissions.

The enrichment factor method has not been shown to be useful in assessing the impact of a single source or in assessing the effect of changes in emissions from that source. Nor has it been shown to be useful in identifying major source categories impacting upon a sampler in an urban area.

6.2.3 Relationship to Other Techniques

No relationships between the enrichment factor technique and those which analyze temporal or spatial patterns of air quality have been reported in the literature, with one exception. The variation between rural and urban sites in the vicinity of a large city can be analyzed in terms of enrichment factors.³⁴ Developments in this area are possible.

The only relationships between this technique and those which assess the effect of meteorological variables derives from

the fact that enrichment factors calculated at rural sites are a way of deducing the impact of long-range transport.

No relationships with previously discussed emission inventorying and diffusion modeling techniques have been reported. However, enrichment factors can be used in conjunction with element intercorrelations to provide evidence of soil dust impact. In addition, enrichment factors can be stratified by particle size ranges to investigate further the impact of windblown and fugitive dust. Again, further developments in this area are possible.

6.2.4 Resource Requirements

Resource requirements for the enrichment factor technique are summarized below:

Manpower	Low ^a
Skill	Moderate ^a
Computer	Not necessary
Data	Chemical/element measurements for aerosol and crust or sea ^b

^a Does not consider the laboratory skills required to utilize the instrumentation indicated.

^b Specific choice of instrumentation depends upon specific elements investigated.

The dependency of this technique upon sophisticated analytical instrumentation to produce element data is a fact which makes this technique simultaneously possible yet costly to apply on a routine basis.

6.2.5 Example Application

An example of this technique is drawn from a recent study of Tucson, Arizona.³⁴ Al was used as the reference element and soil was used as the reference material. A comparison was made between average enrichment factors (E's) for urban and background particulates. Elements which the authors of the study considered

to be attributable to the airborne soil-derived material (Al, Mg, Sr, Fe, Na, Si, Mn, Ti, Rb, Ca, K, and Li) showed E's very close to unity. Other species (Zn, Cu, Cd, Pb, In, Tb, Bi, NO₃, SO₄ and NH₄) were observed to have E's of 50 to 2000 regardless of their urban/rural location. Non-soil sources for these species were deduced. Four other elements (Ce, Co, Ni, and Cs) showed E's of close to 1.0, but their poor correlation with the soil-derived elements led the authors to conclude that either the local crustal composition of these elements was highly variable or that non-soil sources were responsible for introducing a significant fraction of those species into the atmosphere.

The fact that E's were higher for most non-soil species at the rural location than they were at the urban location (see Figure 6-2) suggested to the authors that nonurban sources for those species contributed in large measure to the total atmospheric burden measured in the urban area (as well as the rural area).

Enrichment factors were also calculated for large (>2 um) and small (<2 um) particles. In comparison with data for the entire size range, non-soil species E's were found to be lower for large particles and higher for small particles (see Figures 6-3 and 6-4), thereby adding further credence to the proposition that windblown and fugitive dust were dominant source categories

6.2.6 References

26, 34, 59, 60, 61, 62, 63, 64 [103, 104]

6.3 CHEMICAL ELEMENT BALANCE (CEB)

6.3.1 Description of Technique

The purpose of the CEB technique is to permit the analyst to use available data concerning typical elemental emission rates from selected source categories and elemental concentrations of the aerosol at a given sampling site to back-calculate the fractions of the overall aerosol contributed by all major source

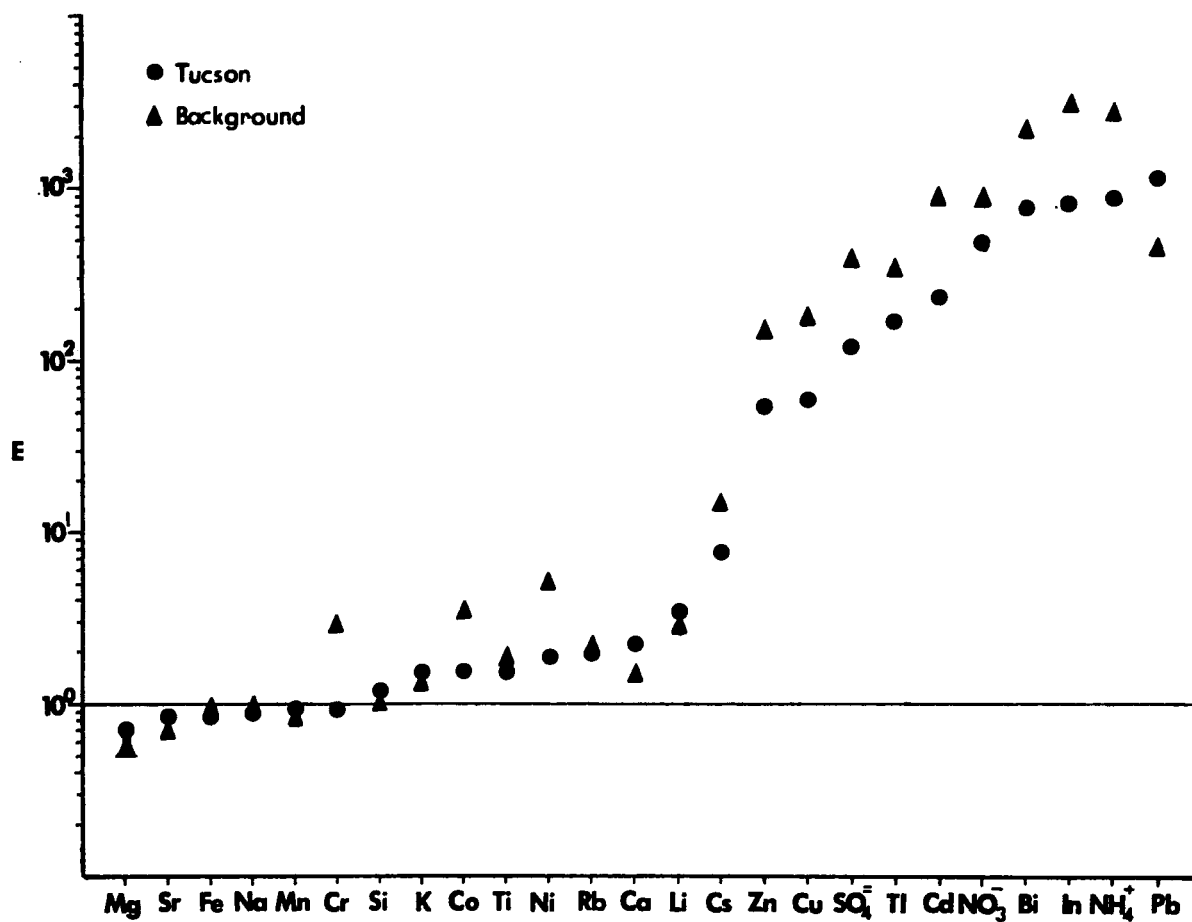


Figure 6-2. Enrichment factors for species in desert background and urban particulate matter.

Source: Reference 34

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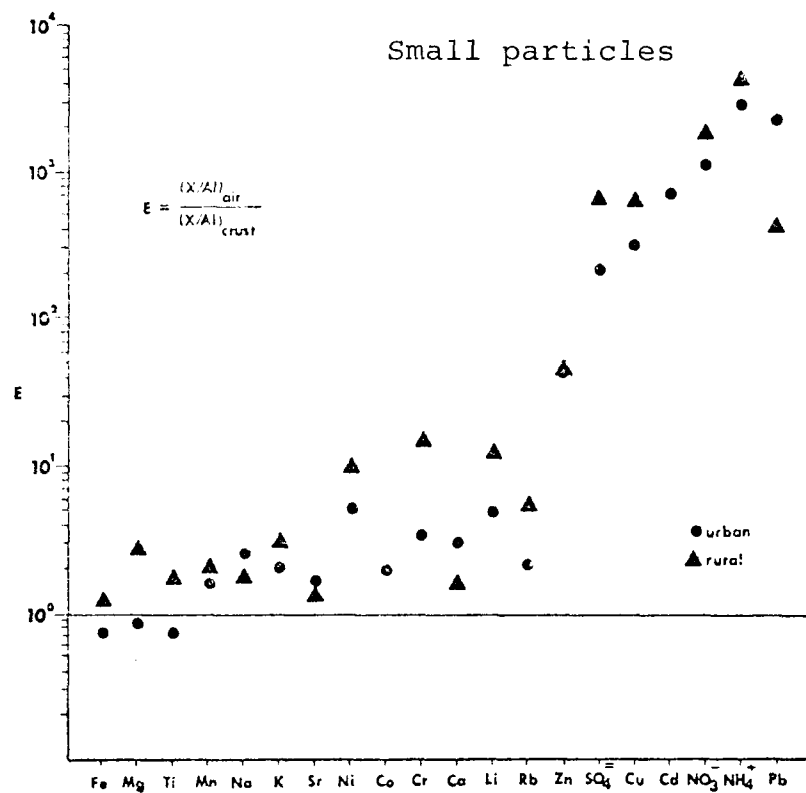


Figure 6-3. Enrichment factors for species in desert background and urban small particle (<2 μm) particulate matter.

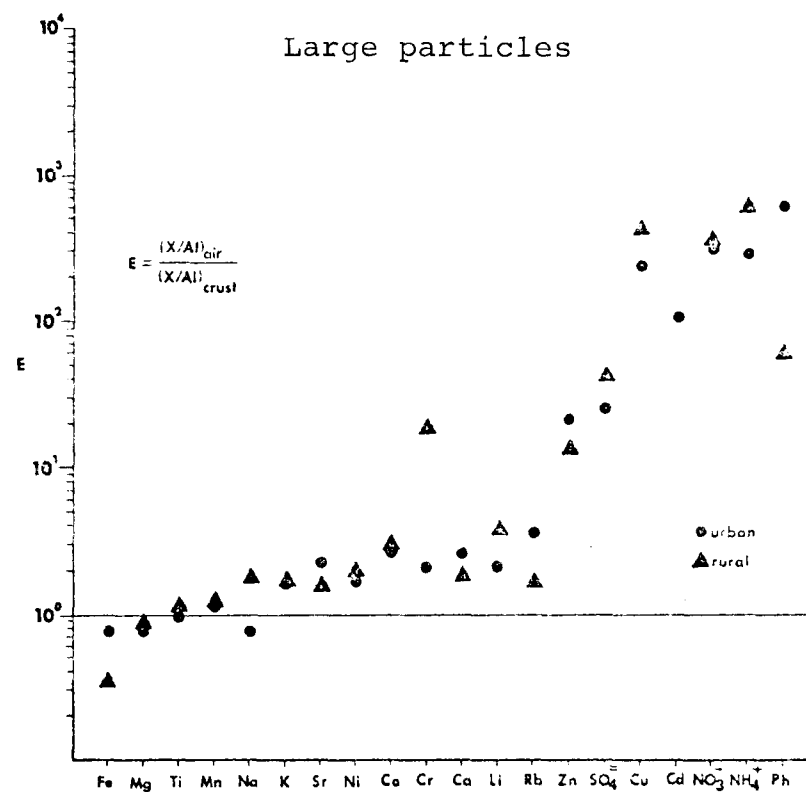


Figure 6-4. Enrichment factors for species in desert background and urban large particle (>2 μm) particulate matter.

Source: Reference 34

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categories. Certain preselected chemical elements are used as tracers for primary particulate emissions from the selected source categories. Secondary contributions, if deemed to be significant enough to warrant inclusion, can be estimated from measured concentrations of SO_4 , NO_3 , and organic vapors.

The fraction (P_i) of element i in an aerosol sample is given by the following simple mass balance equation:

$$P_i = \sum_{j=1}^W a_{ij} x_{ij} S_j \quad (\text{eq.3})$$

where S_j = fraction of the aerosol contributed by source category j

a_{ij} = fraction of source category j 's emissions contributed by element i

x_{ij} = coefficient of fractionation of the element i between the source category j and the atmosphere

W = number of source categories emitting element i

If n elements are considered, then n such equations must be satisfied.

From the definition of S_j , the continuity relation must hold:

$$\sum_j S_j = 1 \quad (\text{eq.4})$$

Values for P_i are obtained from the elemental analysis of atmospheric samples. Values for a_{ij} are extracted from published references or, where practicable, from recent source sampling. Values for x_{ij} , representing the fraction of the emitted species i which appears at a sampling site, are usually assumed to be equal to 1.0. There is, however, some evidence of ion fractionation in sea spray, elemental fractionation in blowing dust, and fractionation resulting from diffusion and sedimentation if particles of different sizes have different chemical compositions.^{65,66} The fraction of the aerosol due to source category j (S_j) is initially unknown.

Where an element comes entirely from one source category, that element can be used as a tracer. In such cases, the mass balance equation is very simple. Values for S_j might be found by solving the following equation:

$$S_j = \frac{P_i}{a_{ij} x_{ij}} \quad (\text{eq.5})$$

For example, Pb is predominantly emitted by the combustion of leaded gasoline. The generally accepted value for the fraction of lead in auto exhaust is 0.4. If the measured Pb fraction of the aerosol is .025, then the mass balance for Pb is simply:

$$S_{\text{auto}} = \frac{.025}{0.4 (1.0)}$$

This yields an: $S_{\text{auto}} = .0625$ or 6.25 percent. The resulting S_j 's for the tracer elements are then used to generate predicted ambient concentrations for non-tracer elements. The overall accuracy of the chemical element balance can then be assessed by comparing measured versus predicted ambient concentrations.

If there were as many tracer elements as source categories (as in the above example), then the analyst could calculate a unique solution for the source contributions. This is rarely the case, however. If there are more tracer elements than source categories, then one could find a linear least squares solution to the mass balance equations of the tracers. Where there are fewer tracers than sources, then linear programming solutions must be found.⁵⁴ The least squares approach, which has been used most frequently in the literature, will be described herein.^{56,61}

The principle behind the least squares method is finding the combination of S_j 's that minimizes the sum of the squares of the deviations between measured and predicted elemental concentrations.⁶⁷ The quantity to be minimized, F , is given by:

$$F = \sum_i \frac{(P_i - \sum_j a_{ij} x_{ij} S_j)^2}{\sigma_i^2} \quad (\text{eq.6})$$

where σ_i = experimental uncertainty in P_i

Setting the partial derivatives of F with respect to the S_j 's to zero results in a system of equations of the form:

$$\sum_j S_j \sum_i \frac{a_{ik} x_{ik} a_{ij} x_{ij}}{\sigma_i^2} = \sum_i \frac{P_i a_{ik} x_{ik}}{\sigma_i^2} \quad (\text{eq.7})$$

The subscript k denotes the kth equation. This yields one equation for each source rather than one for each element. The system of equations is solved by standard matrix methods.^{68,69}

Source categories typically investigated with this technique, tracers used for those sources, and the cities in which those sources/tracers have been used are given below:

<u>Source category</u>	<u>Tracer</u>
Soil dust ^{a,b,c,d,e,f}	Al, (Si) ^e
Sea Salt ^{a,b,f}	Na
Automobile emissions ^{a,b,c,d,e,f}	Pb, (Br) ^c
Fuel oil combustion ^{a,b,c,d,e,f}	V
Portland cement ^{b,c,d,e,f}	Ca
Coal burning and coke production ^{c,e}	Al ^c (Fe, Ti) ^e
Iron and steel manufacturing ^{c,d}	(Mn, Fe) ^c

- ^a Pasadena, CA (1971)
- ^b Pasadena, CA (1973)
- ^c Chicago, IL (1974)
- ^d St. Louis, MO (1974)
- ^e St. Louis, MO (1975)
- ^f Miami, FL (1975)

It will be noted that the list of source categories shown above is not all inclusive. Other source categories, such as incinerators and diesel powered motor vehicles, are not included. The contribution of such source categories is estimated by using

a scaling factor derived from routinely available emission inventories. This scaling factor is the ratio of the emissions of the source category not included in the chemical element balance to the emissions of a source category which is included in the balance.

Most of the referenced applications of the CEB technique terminate at this point. In other words, they discount the effect of secondary particulates. In some cases, notably those of Friedlander, the role of secondary particulates is explicitly accounted for.⁶⁵ The reader is referred to Reference 65 for a detailed explanation of this development in the CEB technique.

The technique clearly has some drawbacks that should be noted by the potential user:

- ° A detailed knowledge of the elemental composition of each source--not routinely available--is necessary. As a result, a large number of assumptions (e.g., a_{ij} is the same for all sources in a given category) and approximations are required.
- ° The technique has no predictive modeling capability.
- ° Source categories, soil dust for example, are indistinct. Within this category are such important subcategories as naturally occurring windblown dust, anthropogenic fugitive dust sources, and wind erosion fugitive dust sources.

6.3.2 Applicability of Technique

The chemical element balance technique uses elemental data that partially characterize the aerosol in terms of its composition, but the primary utility of the technique lies in its unique ability to identify the relative contribution of various source categories to measured TSP concentrations. No other technique has demonstrated an equal ability to quantify the impact of soil dust or sea salt. This capability is limited to the extent that suitable tracers have not yet been identified for many industrial processes and that the technique has not yet shown an ability to distinguish among sources within a given source category.

Even though the CEB technique can identify major impacting source categories, it has not yet been shown to be capable of defining the impact of a specific source. For the same reason, it is not yet possible to use this technique to determine the effect of changes in emissions or the location of specific sources.

6.3.3 Relationship to Other Techniques

The chemical element balance technique has not yet been used in conjunction with techniques that assess temporal variations in air quality. One reason is that the technique depends upon costly analytical techniques such as neutron activation to produce the ambient elemental concentrations (P_i 's). It is possible that temporal variations could be assessed by using aggregate sets of elemental data, but this possibility is dependent upon the emergence of less expensive, real time, analytical instrumentation.

It is possible to use the technique to assess the spatial variation in TSP values by performing CEB for different sites for the same time period. By doing so, one can identify the relative contributions of source categories at various sites.⁵⁷

No efforts to relate this technique to those which assess the effect of meteorological variables have been reported. The reasons are identical to those expressed for temporal variations. Developments in this area are possible.

An emission inventory is a necessary ingredient of the CEB technique at the present time because, in the absence of suitable tracers for all source categories, some means of accounting for "untraced" source categories must be used. That means has been the emission inventory.

Diffusion modeling and CEB technique results can be used to cross-check each other. In the case of diffusion modeling the analyst begins with "known" emission rates and predicts TSP concentrations. In the case of the CEB technique, he begins with known ambient concentrations and back-calculates diffused source contributions. Further, in one recent study the Hanna-Gifford

diffusion model was combined with the CEB and factor analysis techniques to produce a new factor model of air pollution.⁵⁴ The pattern recognition, interspecies correlation, and microscopy techniques could be used to support the findings of the CEB technique.

6.3.4 Resource Requirements

In the following estimate of requirements, it is assumed that the technique will be applied in a fashion similar to that reported in the literature (i.e., applied to small sets of short-term data):

Manpower	Low ^a
Skill	High ^a
Computer	Necessary
Data	Chemical/element measurements for aerosol and major source categories, coefficients of fractionation for sources, emission inventory, tracer, elements for major sources ^b

^a Does not consider the laboratory skills required to use the instruments indicated.

^b Specific choice of instrumentation depends upon specific elements investigated.

6.3.5 Example Application

An example of how the CEB technique is applied can be drawn from a recent study in St. Louis.⁶⁷ Major anthropogenic sources of primary particulates in that city were drawn from a 1975 emission inventory (see Table 6-3).

Table 6-3. SOURCES OF PRIMARY PARTICULATES IN ST. LOUIS

Source	Emissions, ton/yr
Coal combustion	19,817
Fuel oil combustion	862
Waste incineration	1,018
Automobile exhaust	1,276
Diesel exhaust	287
Industrial processes	1,328
Tire dust ^a	128
Soil dust	-
Cement dust	-

^a Assumed equal to one-tenth of automobile exhaust.

Source: Reference 67

Data on the chemical composition of soil dust, automobile exhaust, fuel oil fly ash, and cement dust were extracted from previous reports which applied the CEB method (see Table 6-4). Similar data for emissions from coal-fired power plants were derived by averaging the results of four previous studies. The resulting averages are also presented in Table 6-4. Two 24-h periods were studied: July 23 and 25, 1975. Hi vol samples for those two periods were subjected to organic analyses for benzene, methanol, and water extracts. Two-hour nucleopore filter samples collected over the same time intervals were analyzed for their elemental concentrations using X-ray fluorescence. Analysis for nitrates and ammonium were not made. [It should be noted that other applications have included these variables.] Measured concentrations for these variables are given in Table 6-5.

Al and Si were used as tracers for soil dust; Ca, Pb, and V for cement dust, automobile exhaust, and fuel oil, respectively; and Fe and Ti were used for coal combustion. Resulting balances for elemental concentrations are also given in Table 6-5.

Calculated source contributions are given in Figure 6-5. Industrial, diesel, and tire dust contributions were calculated

Table 6-4. SOURCE CONCENTRATIONS OF PARTICULATE MATTER
(percentages)

	Soil dust ^a	Auto exhaust ^b	Fuel oil fly ash ^c	Cement dust ^b	Coal combustion ^d
Al	5	U	5.0	2.4	11.7
Ca	0.8	U	0.4	46.0	3.8
Fe	3	0.4	2.5	1.09	10.6
K	2	U	0.10	0.53	3.3
Si	20 ^b	U	1 ^b	10.7	24
Pb	0.005	40.0	0.18	-	0.09
Ti	0.3	U	0.03	0.14	1.0
V	0.007	U	2.5	-	0.08
Cd	-	U	-	-	0.004
Co	0.002	U	0.15	-	0.008
Cr	0.005	U	0.12	-	0.065
Cu	0.003	U	0.16	-	0.031
Mg	0.7	U	0.3	0.48	0.9
Mn	0.03	U	0.03	-	0.034
Na	0.6	U	1.5	0.4	
Ni	0.005	U	6.0	-	0.037
Zn	0.01	0.14	0.05	-	0.52
Br	-	7.9	-	-	-

^a Gatz (1975)

^b Miller, et al. (1972)

^c Winchester and Nifong (1971)

^d Average of results in a table provided in Reference 64

U = Unknown

Source: Reference 67

Table 6-5. RESULTS OF CHEMICAL ELEMENT BALANCE FOR ELEMENTAL CONCENTRATIONS

(ug/m³)

	July 23, 1975			July 25, 1975		
	Predicted	Measured		Predicted	Measured	
Al ^a	1.5	2.5	± 1.2	1.0	1.2	± 0.5
Ca ^a	3.0	3.0	0.2	3.5	3.4	0.3
Fe ^a	1.26	1.23	0.05	0.70	0.68	0.04
Si ^a	3.5	3.3	1.0	3.5	2.9	1.2
Pb ^a	0.92	0.93	0.05	0.07	0.70	0.05
Ti ^a	0.121	0.132	0.009	0.070	0.082	0.007
V ^a	0.017	0.017	0.002	0.0082	0.0083	0.0023
Cl	0.156	0.108	0.009	0.12	0.19	0.02
K	0.42	0.33	0.02	0.33	0.41	0.03
Cr	0.0098	0.021	0.002	0.0033	0.023	0.003
Mn	0.0053	0.048	0.003	0.0043	0.043	0.003
Ni	0.0068	0.0068	0.0007	0.0032	0.0019	0.0011
Cu	0.0036	0.36	0.03	0.0014	0.023	0.002
Zn	0.066	0.54	0.04	0.021	0.24	0.01
Br	0.18	0.24	± 0.02	0.14	0.103	± 0.006
Benzene	n.d.	4.2		n.d.	4.8	
Organics						
Methanol	n.d.	11.8		n.d.	13.6	
Organics						
Water	n.d.	14.2		n.d.	17.8	
Organics						

^a Used in chemical element balance.

n.d. = no data

Source: Reference 67

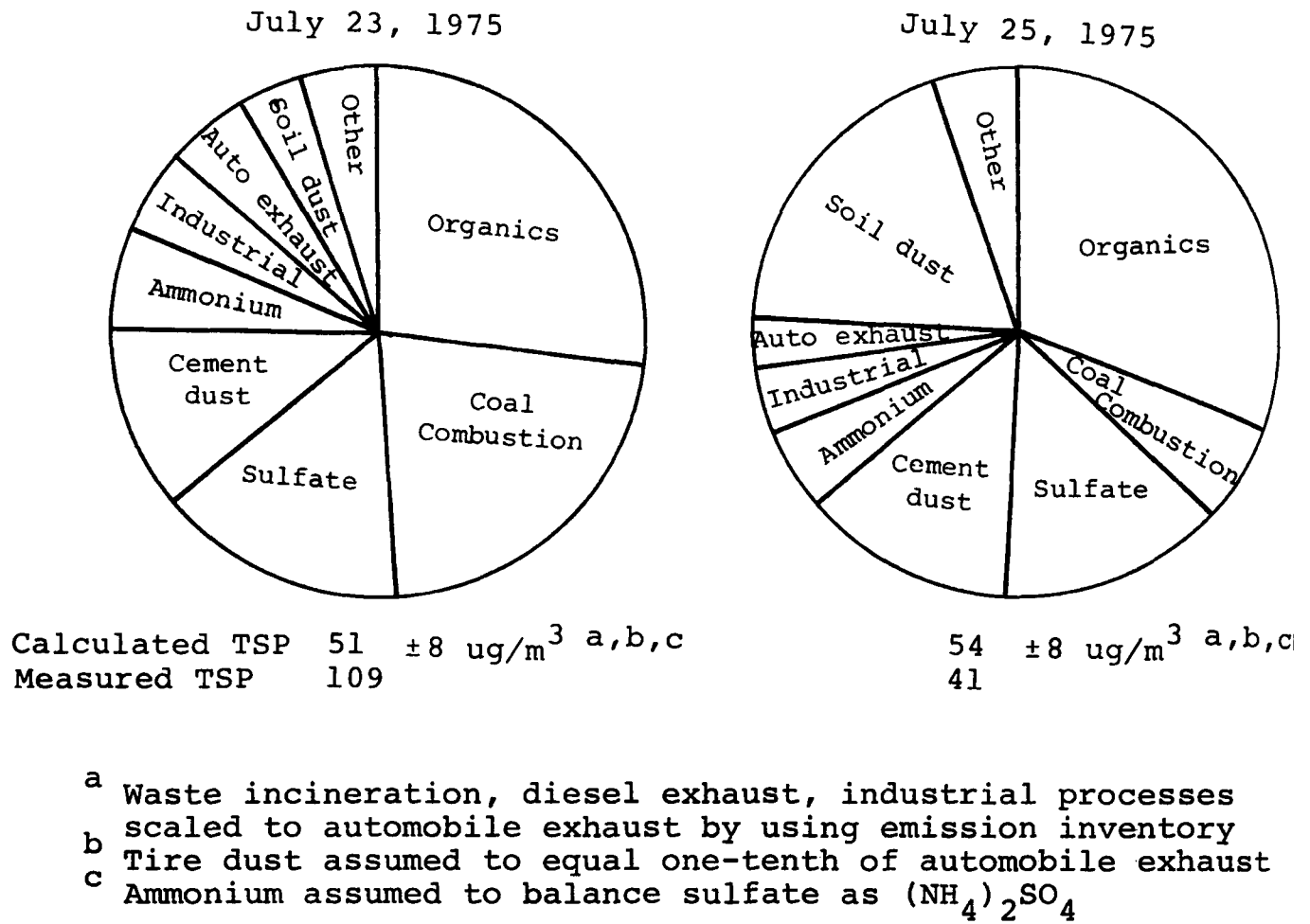


Figure 6-5. Source contributions to St. Louis aerosol.

Source: Reference 67

by scaling the automobile exhaust contribution according to the Table 6-3 emission inventory. Secondary sulfate contributions were calculated by assuming that all sulfur was secondary sulfate. All water-extractable organics were assumed to be secondary.

6.3.6 References

34, 54, 56, 57, 58, 65, 66, 67, 68, 69, 70, 71, 72 [105]

6.4 INTERSPECIES CORRELATIONS

6.4.1 Description of Technique

An important prerequisite for applying this technique is a knowledge of the concentrations of each trace constituent present in a set of particulate samples. Once the trace constituents or species present in a set of particulate samples are quantified, it is possible to evaluate the relationships between species using a correlation analysis. Correlation coefficients are calculated for each possible species pair measured in the set of data. Examples of this technique in the literature include a tabulation of the coefficients in a matrix.^{22,34,54,73,74,75}

There are several types of correlations that can be used. One of these involves the correlation between species concentrations as measured at two different sites. For example, a correlation matrix can be generated for Al or TSP as a function of location. Another type of correlation involves the correlation between species measured at only one location. For instance, the correlation between TSP and Pb or between Pb and Br can be examined at one sampling site.

An important limitation of interspecies correlations is that the correlation coefficients indicate how well two variables vary together, not necessarily that there is or is not a causal relationship between the two variables.

6.4.2 Applicability of Technique

A matrix of correlation coefficients can be developed for species quantified at one site or several sites. By doing so, the aerosol is quantified in terms of its composition and spatial distribution. A good correlation between TSP and another species implies that a large part of the variations in measured TSP can be explained by the variation in the emissions from the source of that species. For example, a high correlation between TSP and Al at one site would indicate that the variation in TSP could be explained by the variability of Al.

When this technique yields good correlations between a species measured at different locations, the results indicate a common source or type of source for the species. For instance, a high correlation between Pb at two different locations would indicate that the Pb measured at the sites has a common source.

The resulting correlations between species associated with a specific type of source serve to identify general impacting source categories. However, no quantitative assessment of source impact is possible. For example, a good correlation between Pb and Br at a particular site would suggest an impact from vehicle exhaust, but it would not indicate how much of an impact (in terms of TSP concentrations) that source category had.

No determination of the impact of a single source or the impact of changes in emissions has been demonstrated with this technique.

One aspect of these correlations should be noted. A low correlation between species suggests that the species may not be related to each other. However, a low correlation does not preclude the possibility that there is a relationship between species. It is possible that the true relationship is hidden by sampling errors, the range of variation among the variables, or other interferences.

6.4.3 Relationship to Other Techniques

With a sufficient number of samples, temporal patterns in interspecies correlations can be investigated for a single site or several sites. If the correlation coefficient changes significantly, it can be inferred that the source category associated with the species varies over time or that the meteorological conditions affecting the measured concentrations changes with time.

Detailed elemental analyses of source emissions can be used with interspecies correlations to tentatively identify major impacting sources.

TSP intersite correlation data can also be compared to the interspecies correlation data. Where good correlations are found between sites for TSP and different species, it can be inferred that the sites are measuring essentially the same air mass. For example, if a good correlation between TSP and Al are found to exist for two or more sites, the implication is that the same air mass is being measured by both sites. Interspecies correlations can also be compared when using the pollution rose and upwind/downwind techniques.

Assuming a sufficient data set is available, interspecies correlations can be determined based on a stratification of meteorological parameters. The nature of the dominant sources can be inferred from observed changes in the correlations. For instance, if TSP data are separated into two categories of precipitation--days with rainfall greater than 0.1 inch and days without rain--a comparison of the correlations between TSP and Al could be made. The results of the analysis might indicate whether the Al is of soil origin or is being emitted from a stationary source. In addition, correlations among NO_3 , SO_4 , O_3 and visibility measurements can be used with trajectory analysis to assess the impact of long-range transport.

Interspecies correlations are direct inputs to the pattern recognition and factor analysis techniques, and can be used with the techniques described in Section 6.1.

6.4.4 Resource Requirements

As the number of samples increases, the resource requirements change. A small number of samples can be evaluated rather easily. However, given a sufficiently large set of data, the resource requirements are:

Manpower	Low ^a
Skill	Moderate
Computer	Necessary
Data	Chemical/element measurements for more than one site (number of values must be adequate for statistical significance) ^b

^a Does not consider the laboratory skills required to use the instruments indicated.

^b Specific choice of instrumentation depends upon specific elements investigated.

6.4.5 Example Application

A recent study of urban air quality in Tucson, Arizona serves as an example of this technique.³⁴ Table 6-6 presents the linear correlation coefficients calculated for the data collected at an urban location. The matrix includes the coefficients calculated for all possible species pairs. Based on the amount of data used, coefficients of 0.3 and greater would indicate a statistically significant relationship. The principal correlations, observed by the authors, from the matrix are:

- The elements Al, Fe, Si, Ti, Li, Tb, K, Ca, Mg, Na, Mn, Sr, and Cr are all correlated with each other
- TSP concentrations are significantly correlated only with the above elements
- The species pair $\text{SO}_4\text{-NH}_4$ is very strongly correlated to each other
- Zn and Cu appear to have some correlation with SO_4 and NH_4

Table 6-6. LINEAR CORRELATION COEFFICIENTS FOR
URBAN SAMPLES, TUCSON, ARIZONA

	Al	SO ₄	NH ₄	Pb	Mass	Fe	NO ₃	Si	Ti	Cs	Li	Rb	Zn	K	Ca	Mg	Na	Cu	Mn	Sr	Ni	Co	Cr	Cd	Bi	Tl	In
Al	---																										
SO ₄	.08	---																									
NH ₄	.10	.81	---																								
Pb	.33	.33	.32	---																							
Mass	.72	.19	.25	.43	---																						
Fe	.75	.15	.14	.34	.68	---																					
NO ₃	.31	.24	.30	.52	.35	.23	---																				
Si	.89	-.01	.08	.24	.70	.72	.27	---																			
Ti	.84	.04	.15	.19	.70	.83	.18	.85	---																		
Cs	.44	-.11	-.03	.30	.47	.25	.39	.42	.44	---																	
Li	.90	.08	.10	.32	.63	.70	.36	.80	.75	.44	---																
Rb	.74	.01	.11	.35	.64	.52	.39	.78	.68	.54	.76	---															
Zn	.01	.54	.50	.25	.08	.42	.09	.0	-.08	-.10	.12	-.02	---														
K	.72	.19	.17	.43	.67	.98	.27	.67	.58	.25	.68	.50	.45	---													
Ca	.78	.25	.23	.23	.66	.91	.25	.70	.66	.22	.73	.45	.33	.89	---												
Mg	.79	.22	.15	.25	.61	.88	.25	.70	.60	.24	.81	.63	.35	.87	.93	---											
Na	.69	.15	.08	.45	.58	.85	.34	.64	.52	.27	.68	.47	.37	.88	.79	.83	---										
Cu	-.05	.55	.41	.23	-.10	-.05	.14	-.12	-.20	.0	.07	-.01	.36	-.03	.0	.05	.0	---									
Mn	.92	.00	.08	.26	.63	.68	.37	.80	.76	.49	.95	.70	.06	.68	.75	.79	.88	.04	---								
Sr	.90	.13	.15	.35	.67	.68	.35	.82	.77	.42	.94	.80	.09	.65	.71	.77	.67	.05	.90	---							
Ni	.30	.19	.25	.23	.25	.34	.43	.27	.15	.21	.42	.33	.30	.36	.34	.47	.41	.23	.40	.40	---						
Co	.25	.11	.05	.17	.23	.20	.31	.19	.20	.38	.35	.27	.15	.19	.15	.20	.25	.15	.37	.32	.24	---					
Cr	.50	.0	.01	.05	.25	.45	.19	.54	.40	.05	.61	.41	.25	.45	.45	.54	.49	.06	.55	.57	.57	.18	---				
Cd	.15	.07	.12	.15	.04	.12	.31	.15	.11	.30	.25	.23	.36	.14	.13	.16	.15	.13	.27	.20	.30	.29	.02	---			
Bi	.08	.07	.02	-.18	-.10	-.07	-.12	.05	.05	.09	.18	-.01	-.03	-.11	.0	.10	-.07	.52	.20	.09	.15	.27	.15	.05	---		
Tl	.0	.08	.12	-.02	-.05	.03	.07	-.03	.04	.10	.18	-.06	.03	.03	.11	.13	.09	.20	.14	.14	.14	.27	.18	-.12	.29	---	
In	.20	-.13	-.20	-.13	-.03	.0	-.16	.23	.05	.0	.25	.15	.0	-.04	-.02	.05	-.05	.13	.23	.20	-.06	.09	.54	-.02	.35	-.03	---

Source: Reference 34

- ° The nitrate ion is only weakly correlated with lead
- ° The elements Cd, Bi, In, and Tl show no apparent relationship to any measured chemical component

In the Tucson study, the elements Al, Fe, Si, Tn, Na, Mg, Ca, Rb, Sr, K, Mn, and Si were all related to each other at all of the locations sampled. This relationship suggested to the authors that a possible common source for these species was the airborne soil material generated by erosion processes.

6.4.6 References

22, 34, 54, 73, 74, 75

6.5 PATTERN RECOGNITION

6.5.1 Description of Technique

Pattern recognition is a statistical technique used for evaluation of complex data sets. Pattern recognition can involve either supervised or unsupervised learning. In supervised learning, a data set is characterized based on previously established patterns. Unsupervised learning, referred to as cluster analysis, requires no preconceived patterns. Discrete elements of a data set are grouped depending on the degree of similarity between elements. In this section, only unsupervised learning will be discussed. As the discussion is only a cursory examination of cluster analysis, the reader interested in a more complete description of the technique is directed to a standard text on the subject.⁷⁶

Cluster analysis lends itself to the study of air pollution in cases where patterns are only vaguely defined. To perform clustering, some measurement of the similarity between elements of a data set is needed. As applied to TSP samples, this measure is usually Euclidean distance. The measure of distance between species is based on the correlation coefficient (r), where $1-r$ represents the distance between two species.

The first step in a clustering algorithm is to join the two most similar clusters. Then, the distance between the new cluster and each remaining cluster is computed and the process is repeated. Reiterations of this process result in all of the elements forming a single cluster. Results of this hierarchical clustering are usually displayed in a dendrogram, (i.e., a diagram of the clusters present at each stage of clustering).^{34,54,77,78} An alternative way to present the clustering results is to generate a nonlinear mapping of the clusters.^{34,61,77} An example of a dendrogram is given in Figure 6-6.

6.5.2 Application of Technique

This technique can characterize the aerosol by providing a measure of the way in which TSP concentrations and the chemical/elemental constituents thereof vary over space.

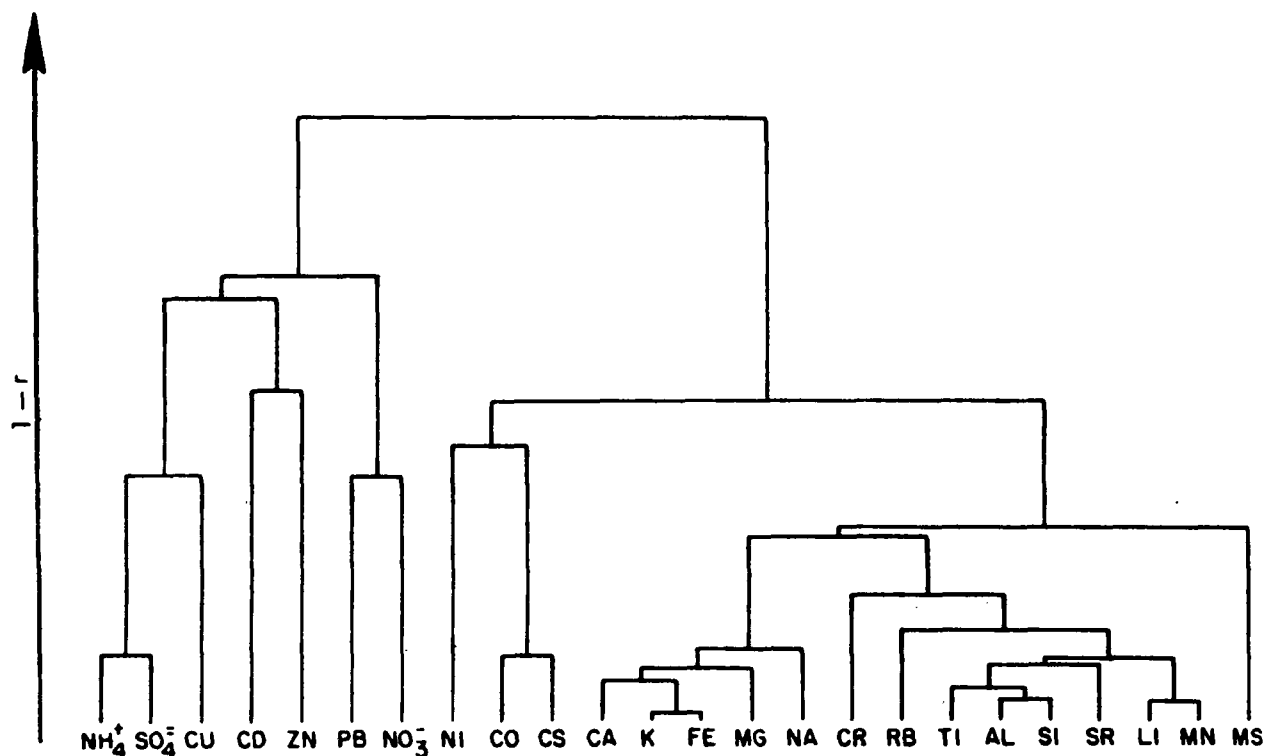
The cluster analysis technique can be used to identify general source categories present in the aerosol. In order to apply the technique in this fashion, a knowledge of the concentrations of the species present in the aerosol is needed. By analyzing the clusters formed from different species, general emission categories can be identified. For example, if Al and Si cluster together, one probable source for these elements is windblown soil particles.

This technique does not lead to the quantification of the impact of a specific source of emissions. The technique has no predictive capabilities and it cannot be used to trace changes in emissions over time.

6.5.3 Relationship to Other Techniques

The most useful relationship of this technique to other techniques is in the interpretation of the results of correlation analyses. The pattern recognition technique, cluster analysis, condenses the information obtained from previously discussed correlation techniques and then presents the information graphically. As such, this technique allows general relationships to

HIERARCHICAL CLUSTERING



The vertical distance is proportional to the degree of dissimilarity between clusters (l-r).

Figure 6-6. Dendrogram of feature clustering for desert urban particulate matter.

Source: Reference 34

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be viewed more efficiently. However, no new information is obtained using the technique that could not have been observed from a careful study of the correlation matrices. The same sets of species with mutually high correlations and similar behavior noted in the matrices will normally be observed in the cluster analysis.

The cluster analysis technique can also be used to support findings of chemical element balance, factor analysis, and the techniques described in Section 6.1. In addition, a knowledge of the existing emission inventory is necessary in order to interpret the results of the pattern recognition technique properly.

6.5.4 Resource Requirements

Given a sufficiently large data set, the resource requirements are:

Manpower	Low ^a
Skill	Moderate
Computer	Necessary
Data	Chemical/element measurements for more than one site (number of values must be adequate for statistical significance) ^b

^a Does not consider the laboratory skills required to use the instruments indicated.

^b Specific choice of instrumentation depends upon specific elements investigated.

6.5.5 Example Application

An example of the application of cluster analysis can be drawn from a recent analysis of air quality in Tucson.³⁴ Based on the correlation matrix presented previously in Table 6-6, hierarchical clustering was performed on the species using 1-r as a measure of the dissimilarity among species. The resulting dendrogram is depicted in Figure 6-6.

As shown in the figure, there is a clustering of the species Ca, K, Fe, Mg, Na, Cr, Rb, Ti, Al, Si, Sr, Li, and Mn. These

elements are probably of soil origin. The remaining features, NH_4 , SO_4 , Cu, Cd, Zn, Pd, NO_3 , Ni, Co and Cs, are well separated from the soil cluster. This separation suggests different sources and/or different chemical and physical behavior in the atmosphere.

Ni, Co, and Cs exhibit a behavior intermediate between soil and nonsoil indicating that a portion of these elemental concentrations may have a soil origin.

The high degree of correlation between NH_4 and SO_4 is probably explained by the chemical combination of these species resulting from the acid base processes after SO_2 is oxidized to H_2SO_4 .

6.5.6 References

34, 54, 61, 76, 77, 78

6.6 FACTOR ANALYSIS

6.6.1 Description of Technique

Factor analysis is related to cluster analysis in its intuitive approach to the data. It was originally developed to explain intelligence test scores and has found wide application in the social and biological sciences. It is a tool for explaining observed relationships between large numbers of variables in terms of fewer variables. The new variables are linear combinations of the original ones. The reader should be aware that the description presented in this section is not designed to convey a complete understanding of the statistical principles and the mathematical derivation of the technique. To obtain a complete understanding of factor analysis and the computational procedures involved in its application, a standard text on the subject should be consulted.⁷⁹

Factor analysis may be useful in treating atmospheric data for the following reasons: first, it is not possible to control

the values of the variables experimentally, and second, the introduction of particulates into the atmosphere from a number of different sources and areas can be described by a variable which is a weighted sum of factor values.

Two models of factor analysis are described in the literature: the classical or common factor model,^{34,54,71,77,78} and the principal components model.^{73,80}

Classical factor analysis assumes that observed correlations between variables are the result of regularity in the data. Each variable is expressed as a linear combination of factors common to all variables and a unique factor not shared with the other variables. The values of the variables can be expressed as a set of n linear equations:

$$Z_i = \sum_{j=1}^m a_{ij} F_j + d_i U_i, \quad (m < n) \quad (\text{eq.8})$$

where Z_i = variable i in standardized form

F_j = common factors

U_i = unique factor for variable i

a_{ij} = factor loadings (correlation between a variable and the factor of which it is a part)

d_i = standard regression coefficient of variable i on unique factor U_i

m = number of factors

The common factors are chosen so that the correlations among the species can be reproduced as well as possible from only a few factors.

The factor analysis is begun by calculating the linear correlation coefficients for each of the possible pairs of variables. Next, the common factors must be found. The approach used in the literature is called the principal factor method. In this procedure, the matrix of correlation coefficients is modified so that the elements in the principal diagonal are the

estimated values of the communalities (variances of each variable that result from the common factors). The matrix is then called a communality matrix. There are several methods available to estimate the communalities. The usual methods involve the highest correlation coefficient for each variable or the squared multiple correlation.

The method found most frequently in the literature is the squared multiple correlation (SMC). The SMC is found from the inverse of the correlation matrix (with unity in each diagonal portion) by the formula:

$$SMC_j = 1 - \frac{1}{r_{jj}} \quad (\text{eq.9})$$

where r_{jj} = diagonal element for the j th variable in the inverse of the correlation matrix

The next steps in factor analysis are to diagonalize the communality matrix, determine the eigenvalues, and determine the corresponding orthogonal eigenvectors. The factor loadings can then be calculated from the following equation:

$$a_{ij} = \alpha_{ij} \lambda_i^{1/2} \quad (\text{eq.10})$$

where λ_i = eigenvalues

α_{ij} = components of the eigenvectors

a_{ij} = factor loadings

The factor loadings represent the correlations of a variable with the factor of which it is a part. In the factor matrix, each row of the matrix corresponds to one variable and each column represents one factor. There are as many factors as there are variables. However, normally only those factors with eigenvalues greater than or equal to 1.0 are used.

Using matrix algebra, factors are rotated to point a factor at a cluster of variables so the factor may be easier to interpret.

The object of rotation is to produce a factor matrix with high loadings in some rows and near zero values in the other rows. A number of methods of rotation are described in the literature, including varimax, quartimax, and equimax. The factor matrix is examined to determine which variables reveal high loadings (0.50 or greater).

An important aspect of factor analysis is that the solution is not unique. It is possible to produce equally valid sets of transformations through rotation from the same input data. The high loadings cannot be defined objectively. They require that other information be used to select appropriate descriptors for the factors.

The analyst does not preselect these factors. Rather, they are combinations of the original variables that explain the observed variance in the data. The first factor explains more of the variance than any other factor, the second more than any other except the first, and so on.

The principal component model mentioned earlier in this section ignores the unique factor of a variable. It does not require communality estimates, since it is interested in total variation among all variables instead of the common variance. The calculations of the component solution and the subsequent rotation are essentially the same as in the classical factor model.

The final step in factor analysis is the extraction of factor scores (the measure of each variable on each factor). These factor scores can be used as independent variables in regression analysis or as a new data set for further analysis.

6.6.2 Application of Technique

The application of this technique to the quantified species present in TSP samples leads to a characterization of the aerosol. The loadings on the factors give an indication of which species are linearly related. For example, one factor may contain high loadings on Pb and Br, indicating the species are related in the aerosol.

The factor analysis technique can be used to identify general source categories. By analyzing the high loadings in each factor, general emission categories can be inferred from the species having the high loadings. In an industrial area, for example, high loadings on Fe, Mn, and Ti for one factor could be expected to be associated with heavy industrial processes, such as steel making.⁷³

The technique does not lead to a quantification of the impact of a specific source of emissions, nor does it have any predictive capabilities. It cannot be used to trace changes in emissions over time.

6.6.3 Relationship to Other Techniques

The primary relationship of other techniques to factor analysis is in the interpretation of the results of the analysis. The factor loadings obtained for a particular factor need to be related to some aspect of the physical world. The cluster analysis, enrichment factor, microscopy, and interspecies correlation techniques can be used to aid in the interpretation of the results of factor analysis. In addition, knowledge of the existing emission inventory is necessary to attach meaning to the results.

Meteorological variables, such as wind speed, precipitation and wind direction, can be included as variables in a factor analysis.

A recent study combined the techniques of factor analysis and chemical element balance with a simplified diffusion model, Hanna-Gifford, into a factor model.⁵⁴ This model accounted for the observed means, standard deviations, and correlations of the primary chemical elements in the aerosol. Another study used the factor analysis technique to determine which types of sources were affecting sampling sites in St. Louis.⁸⁰

6.6.4 Resource Requirements

The mathematical formulation of factor analysis is most simply expressed in matrix notation. Therefore, because of the extensive computations necessary to reduce large matrices, factor analysis has become practical only with the ready availability of large digital computers. Programs from the Statistical Package for the Social Sciences (SPSS) or the BMDP Biomedical Computer Programs can be used to carry out the factor analysis computations described above. The resource requirements for factor analysis are:

Manpower	Moderate ^a
Skill	High ^a
Computer	Necessary
Data	Chemical/element concentrations; meteorological data optional

^a Does not consider the laboratory skills required to use the indicated instruments.

^b Specific choice of instrumentation depends upon the specific elements investigated.

6.6.5 Example Application

A good example of the application of factor analysis can be drawn from a recent analysis of air quality in Tucson.³⁴ Using the correlation matrix presented previously in Table 6-6, a factor analysis was performed. Table 6-7, presents the results of the factor analysis for an urban site. This six factor solution accounted for about 88 percent of the total variance of the species (see Table 6-8). Examination of the loadings of the corresponding factors resulted in the interpretation discussed below and summarized in Table 6-9. Recall that a factor loading is a correlation coefficient between a species and a factor, and that the results presented below are only one possible explanation for the factors calculated from the analysis. Different results could be obtained if data from another city were used or if a different rotation were performed.

Table 6-7. FACTOR LOADINGS FOR AN URBAN SITE
(VARIMAX ROTATION OF SIX FACTORS)

Species	Factor					
	1	2	3	4	5	6
Zn	.08	.27	.74	.27	-.09	.02
Pb	.04	.41	.10	.04	-.74	.05
Cu	-.03	.69	-.01	.17	-.04	.35
Cd	.02	.14	.20	.60	-.20	.02
NH ₄ ⁺	-.01	.86	.14	.07	-.20	-.19
SO ₄ ⁼	-.03	.93	.10	.02	-.16	-.06
NO ₃ ⁻	.17	.19	.02	.30	-.62	-.01
Ni	.52	.19	.12	.35	-.15	.36
Cr	.85	.02	.08	.24	.07	.29
Li	.94	-.03	-.09	.13	-.03	.18
Mn	.94	-.06	-.10	.13	-.01	.20
Sr	.94	-.06	-.05	.09	-.12	.03
Al	.97	-.05	-.07	.03	-.05	-.02
Si	.98	-.03	-.06	.07	.01	-.07
Rb	.87	.03	-.19	.21	-.14	-.05
Ti	.95	-.06	-.11	.05	.07	-.18
Mass	.84	.11	.01	-.04	-.03	-.27
Na	.82	-.05	.36	-.07	-.33	.18
Mg	.93	.01	.24	-.03	.01	.10
Fe	.91	-.03	.35	-.09	-.10	-.05
Ca	.87	.06	.37	-.09	-.08	-.10
K	.89	-.00	.39	-.10	-.17	.02

Source: Reference 34

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Table 6-8. EIGENVALUES OF CORRELATION MATRIX
FOR AN URBAN SITE

EIGENVALUE	% OF VARIANCE	CUMULATIVE % OF VARIANCE
13.2	55.17	55.17
3.46	14.43	69.60
1.43	5.94	75.54
1.21	5.03	80.57
1.06	4.41	84.98
0.719	3.00	87.98
0.607	2.53	90.51
0.517	2.15	92.66
0.375	1.56	94.23
0.338	1.41	95.64
0.239	1.00	96.63
0.168	0.70	97.33
0.136	0.57	97.90
0.102	0.43	98.33
0.094	0.39	98.72
0.085	0.36	99.08
0.080	0.33	99.41
0.040	0.16	99.57
0.035	0.15	99.72
0.026	0.11	99.83
0.022	0.09	99.92
0.011	0.04	99.97
0.005	0.02	99.99
0.004	0.01	100.00

Source: Reference 34

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Table 6-9. ELEMENT LOADINGS ON INDIVIDUAL FACTORS AND POSSIBLE EXPLANATIONS FOR FACTOR SIGNIFICANCE

	1	2	3	4	5
Elements					
Major Loading	Mass, Al Ti, Si Sr, Rb, Li, Mn, Ca, K, Fe, Mg, Na, Cr	NH_4^+ , $\text{SO}_4^{=}$, Cu	Zn	Pb	NO_3^-
(Minor Loading)	(Ni, Zn)	(Ni, Cd, Zn, Pb, NO_3^-)	(Mg, Na, Fe, Ca, K)		
Physical or Chemical Significance	Soil Large particles	Distant and/or diffuse source Small particles Gas- particle conversion	Common aliquot dilution variance	Automo- tive	Gas particle conversion

	6	7	8	9	10
Elements					
Major Loading	Cd				
(Minor Loading)	(Zn, Ni, Cu, Pb, NO_3^-)	(Li, Mn, Co, Cu)	(Ni, Cr, Cu)	(Rb)	(Na)
Physical or Chemical Significance	Unknown May have combustion and/ or transport sig- nificance	Urban only May sug- gest the signature of one or more urban sources	May sug- gest local mining or other anthropo- genic activity	Unknown	Background only Marine source

Source: Reference 34

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The first factor had high loadings on Na, K, Fe, Ca, Mg, Rb, Sr, Al, total mass, Ti, Si, Li, Mn, and Cr. The variance common to these species was probably due to their common source, soil.

Factor two has high loadings from SO_4 , NH_4 , and Cu while Pb, Zn, and a few other species had some variance in this factor. From these loadings, it was inferred that the factor corresponds to a background aerosol from nonlocal sources.

Factor three had a high loading from Zn and medium loadings from K, Ca, Na, Fe, and Mg. The medium loadings were accounted for by the chemical analysis procedures used to analyze these species. Zn does not cluster with these species as shown above in the dendrogram in Figure 6-6. Therefore, it appears that Zn originates from other sources.

Cd was the major contributor to factor four. Intermediate loadings were observed on Ni, NO_3 , and Zn. It was suggested that this factor is due to a high temperature or combustion process since these species have been identified as being enriched in fly ash.

Factor five is loaded from Pb and NO_3 and seems to reflect an automotive source.

Factor six has only intermediate loadings on Cr, Ni, and Al. One possible explanation for these factors is mining activity or local soil variations.

As indicated in the above analysis, the factor analysis took the results of correlation analysis a step further. A number of variables which contained portions of the variance common to several species were noted. However, interpretation of the factors by a researcher is the most critical part of the analysis.

6.6.6 References

34, 54, 71, 73, 77, 78, 79, 80

6.7 INTERPRETATION OF MORPHOLOGICAL DATA

Filter analysis by optical or electron microscopy to identify the probable generic origins of individual particles is becoming more frequently used in the assessment of TSP sources. The microscopy techniques are primarily laboratory analyses and are therefore not directly comparable with the data analysis techniques described in this digest. Data analysis following microscopy has generally been limited to a comparison of microscopy results (mass percent of particle types indicative of different source categories) with estimates of source contribution from other analyses such as emission inventory, X-ray fluorescence, or diffusion modeling.

The use of microscopy results to support or extend the findings of TSP data analyses depends on the confidence placed in quantitative results of microscopy. There are a number of divergent opinions on this topic. Therefore, the laboratory microscopy technique is discussed here in some detail to provide a basis for utilizing its output as part of TSP analyses.

6.7.1 Description of Technique

Filter analysis by microscopy is viewed by EPA as a semi-quantitative technique for identifying generic types of particulate in the >1-2 μm particle size mode. Particles are either viewed in situ through the use of immersion oil or removed and remounted using probes or an adhesive. The principal instrument used for routine analysis of the filters is the polarizing light microscope. Particles are examined with both transmitting and reflected light and characterized as to their generic type by observing morphology (structure and form), transparency, color, and other physical properties. Other analytical techniques such as electron microscopy are often used in conjunction with optical microscopy as a supplement to the morphological identification. The particles are usually grouped into categories such as minerals, combustion products, biological material, and miscellaneous, with further subcategorization where possible. A

detailed breakdown of particle categories and the average composition of 300 filter analyses performed by one laboratory are shown in Table 6-10.⁸¹

Particle counts and categorization must be made by size range in order to convert to mass percents. Also, the average density and shape factor for each particle category must be determined or assumed. In some cases, complex mathematical calculations have been made to derive values other than the size range midpoints to use in estimating the percent by mass for each particle category. However, estimates of relative contributions by source category are often limited to such classifiers as major, moderate, and trace. If percentages are assigned, the estimate should be rounded to 10 percentile values.

Several studies have been done to assess the reproducibility of microscopic analysis results. Such studies used reanalyses or blind replicate samples and results were compared. One study involved inter- and intra-laboratory analysis of blind replicate samples. The results showed a wide disparity (as much as a severalfold difference) among analysts, which suggests a potentially serious shortcoming of the technique. Factors such as analyst fatigue, misidentification, and misassignment of particles to source categories were suggested as causes for this lack of reproducibility.

Particles smaller than 1-2 μm are very difficult to identify microscopically. It has been shown that TSP is bimodally distributed with a minimum between the modes occurring in the 1 to 2 μm range. These modes are referred to as "fine" and "coarse," and the fine mode may include as much as 25 to 50 percent of the mass on the filter. Thus, any analysis which relies solely on microscopy may miss a large portion of the fine particle mode. The typical makeup of the fine and coarse modes have been characterized generically with the coarse mode consisting mostly of natural or fugitive particulates and the fine mode consisting mostly of stack emissions and secondary particles.

Table 6-10. COMPOSITE SUMMARY OF FILTER ANALYSES

Components	Quantity, percent	
	Average	Range
<u>Minerals</u>	(65)	3-99
Quartz	29	<1-84
Calcite	21	1-93
Feldspars	5	0-35
Hematite	10	0-65
Mica	<1	0-15
Other	<1	0-46
<u>Combustion Products</u>	(25)	1-89
Soot: Oil	7	0-86
Coal	5	0-52
Misc soot	5	0-88
Glassy fly ash	6	0-30
Incinerator fly ash	2	0-45
Burned wood	<1	0-10
Burned paper	<1	0-<1
Magnetite	<1	0-15
Carbon black	<1	0-19
Other	<1	0-15
<u>Biological Material</u>	(3)	0-90
Pollen	1	0-45
Spores	<1	0-2
Paper	<1	0-3
Starch	1	0-10
Misc plant tissue	1	0-8
Leaf trichomer	<1	0-18
<u>Miscellaneous</u>	(7)	0-50
Iron or steel	<1	0-25
Rubber	7	0-50
Other	<1	0-15

Source: Reference 81

Particle sizing by microscopic analysis can give some clues as to the size distribution in the large mode, but is hampered by agglomeration on the filter and also by the randomness of occurrence of very large particles within the field of view of the microscopist. Also, a heterogeneous distribution of particle sizes may be present through the filter such that the large particles and agglomerates may not penetrate as far into the filter mat as the fine particles. Unless fields at several focusing depths are analyzed in situ, this particle stratification may bias sizing and identification studies.

Microscopy has certain limitations in the accuracy by which particles can be identified. Crystalline compounds such as quartz or calcite can be easily identified but the inability to designate whether these constituents were entrained by the wind or resuspended by man's activity is a severe limitation. Some particles, such as rubber, are sometimes mistaken for combustion products. Many fine opaque particles, such as automotive, diesel fuel oil, and coal combustion products, cannot be easily distinguished from each other. Using optical and electron techniques in concert can provide more accurate and reliable particle identification, but some of the problems associated with particle sizing and sample handling may still exist.

Finally, sample preparation procedures may bias results. Removal of particles from the filter by manual manipulation (particle picking) or transfer (using an adhesive substance such as Aroclor) only samples the uppermost layers of particles. If any heterogeneous size stratification does exist, as suggested above, the removal procedure exacerbates this problem. Removal by ultrasonics is sometimes used, but this may disaggregate the agglomerates and solubilize some particles.

6.7.2 Applicability of Technique

Microscopy has had limited success in identifying specific sources of particulate matter from routinely collected ambient samples. It seems to have had the most success in either: (1)

very general qualification of the problem (e.g., mineral matter versus combustion as generic categories) or (2) very specific studies around a suspected source where the morphology of emitted particles is unique to the area. It is limited in capability by problems associated with particle sizing, sample preparation, the identification and assignment of particles to generic categories, and reproducibility of results, as discussed above.

Microscopy definitely characterizes the aerosol; it provides information on both the size distribution and physical properties of the particles. In conjunction with simultaneous analyses for elemental composition of the particles, these techniques provide more data on characteristics of the aerosol than any other available TSP analyses.

The technique has been employed most frequently to determine impacting source categories, as described in Section 6.7.1. However, its results have been overextended in some highly quantitative analyses of the contributions from single sources or source categories.

6.7.3 Relationship to Other Techniques

The most important relationships are with techniques that simultaneously analyze filter samples for elemental composition. These include X-ray diffraction, energy dispersive X-ray analyses (X-ray fluorescence), electron microprobe (electron spectroscopy for chemical analysis), and neutron activation. Most of these techniques give the elemental composition of the entire sample on the filter, but some (at least energy dispersive X-ray analysis) can determine the composition of individual particles for even greater resolution of morphological/elemental groupings.

In addition to being a particle sizing technique, microscopy is closely related to the cascade impactor sizing technique. Microscopic analyses have been performed on the individual filter stages of impactor samples to determine the morphology and probable generic origin of particles on each stage. This analysis also provides a direct comparison of the two sizing techniques.

Microscopy can be used as a supportive or confirmatory analysis with any other technique that determines impacting source categories.

6.7.4 Resource Requirements

Microscopic analysis of filter samples is quite time consuming and requires a high level of skill. Analysis by optical microscopy alone usually takes two to four hours per filter plus sample handling and preparation time. In contrast, requirements for data analysis of microscopy results are minimal. The resource requirements summarized below are for the microscopy work:

Manpower	High
Skill	High
Computer	Optional (for calculating mass percentages)
Instrumentation	Microscope and accessories

6.7.5 Example Application

In order to determine the feasibility and usefulness of routine microscopic examination of hi vol filters, the Texas Air Control Board (TACB) submitted 126 samples for optical microscopy analysis.⁸² These filters had previously been analyzed by X-ray fluorescence spectroscopy (XRF) to identify and quantify source contributions, so the microscopy data optimally would confirm the XRF findings and aid in source identification where XRF data were inconclusive or where particle types were not identifiable through XRF.

Particles were identified under the polarizing microscope by noting their color, shape, optical characteristics, crystal system, and other distinguishing properties. They were assigned to the same particle categories as listed in Table 6-10. Volume percentages were determined for each particle category, then an assumed density factor was applied to determine the weight percentages.

Additional sections of five filters were resubmitted for analysis. Three of the duplicate pairs had overall comparisons within the ± 8 percent precision claimed by the laboratory performing the work. However, the other two samples had errors in the range of 30 to 60 percent. Because of this discrepancy, TAC considered the microscopy results only as supportive evidence to the XRF and drew no conclusions on source contributions based on microscopic evidence alone.

In general, microscopic analysis proved to be supportive of the XRF results. In cases where industrial particles were identified microscopically, elemental analysis had shown the presence of significant amounts of the same material. For traffic-related emissions, filters with a high Pb content and a good Pb/Br correlation also had a significant percentage of tire rubber. Identification of local soil types and quantitative determination of fugitive dust impact was accomplished by XRF. Samples with high Si and Ca values had a large proportion of silicates and limestone identified by microscopy. Samples with high Al values had significant amounts of feldspars and clays microscopically. However, correlations between XRF values and particle mass percentages were occasionally difficult to show.

The results of microscopic analysis of filters from one highway site near the Houston ship channel are shown in Table 6-11.⁸⁴

Table 6-11. MICROSCOPIC ANALYSIS RESULTS, SITE 115004

<u>Date</u>	<u>TSP</u> <u>($\mu\text{g}/\text{m}^3$)</u>	<u>Minerals</u>	<u>Combustion</u>	<u>Iron</u>	<u>Other</u>	<u>Wind</u> <u>Direction</u>	<u>Comments</u>
12/09/71	268	47%	51%	2%	0%	SE	
1/22/72	317	27	0	8	65	S	Ship*
3/11/73	334	10	90	0	0	NE	Coke
12/30/73	473	65	0	3	32	SE	Ship
3/12/74	291	35	5	60	0	SE	Ship
8/27/74	276	14	24	2	60	SE	Ship*
1/30/75	138	52	21	26	0	SW	
2/23/75	204	4	90	0	0	NW	Coke
7/17/75	217	63	1	11	25	SE	Ship
12/08/75	332	13	87	0	0	N	Coke

Coke - Fugitive dust from coke storage pile.

Ship - Fugitive dust resulting from ship unloading activities.

Ship* - Unloading activities one or two days prior to actual sampling date.

Source: Reference 82

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6.7.6 References

59, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93

7.0 INTERPRETING PARTICLE SIZE DATA

Several different techniques for analyzing total suspended particulate and chemical species data have been presented in the preceding chapters. Used individually or interactively, they can tell the analyst much about the nature of the aerosol and its sources. But regardless of how well they are used, these techniques will always be limited by their blindness to the physical property of the aerosol that is most fundamentally determinative of the impact of that aerosol upon the health of the human organism: particle size. Particles in excess of ~ 15 μm do not normally become trapped in the respiratory system, hence they will not cause any adverse health consequences. Particles in excess of ~ 3.5 μm cannot normally intrude into the terminal bronchial and alveolar portions of the human lung. As a result, it is desirable for the analyst to have access to techniques which assess the particle size distribution of the measured aerosol.

There are several different methods currently being used to measure an aerosol's particle size distribution, among which are filter analysis by microscopy, cascade impactors, dichotomous samplers, diffusion batteries, optical counters, and electrical analyzers. It is not the purpose of this chapter to discuss the theory and construction of each of these methods, but some brief comments are warranted, given the methods potential use in providing data to which various analytical techniques can be applied.

Table 7-1 briefly summarizes the size ranges that are reportedly sampled by the above-mentioned methods:^{94,95,96}

Table 7-1. SIZE RANGES SAMPLED BY CURRENT METHODS

Method	Size range, μm
Diffusion battery	0.002 - 0.2
Electrical analyzer	0.0032 - 1.0
Cascade impactor	0.5 _b - 20 ^a
Dichotomous sampler	<16 ^b
Optical counter	>1.0
Microscopy	>1-2

^a Final filters collect a composite sample below some cutoff size; e.g., 1.1 μm for typical five-stage device.

^b One intermediate stage cutoff occurs at 3.5 μm .

Of these methods, only cascade impactors and microscopy are commonly used in ambient aerosol investigations. The other three have been used only in advanced research.^{19,97} Thus, currently available equipment indicates that only particles in excess of 0.5-1.0 μm can be successfully stratified into size classes.

This is a severe limitation. It has been shown that ambient aerosol size distributions are almost always bimodal in surface or volume, are frequently trimodal in surface area near sources of fresh combustion aerosols, and that the minimum between the two most common modes occurs in the 1 to 2 μm range (see Figure 7-1).⁹⁷ As a result, the most commonly used methods provide meaningful distribution data only for larger particles. Such distribution data are of limited utility, since it has also been shown that many sources emit particulate matter in the submicron range and that most secondary particulates also can be found in that range.⁹⁸

Given these limitations, the remainder of this chapter concentrates upon ways of analyzing particle sizing data that are reasonably available to air pollution control agencies. Two general techniques for analyzing such data have been commonly used:

- Frequency distribution comparisons
- Species-specific size distribution comparisons

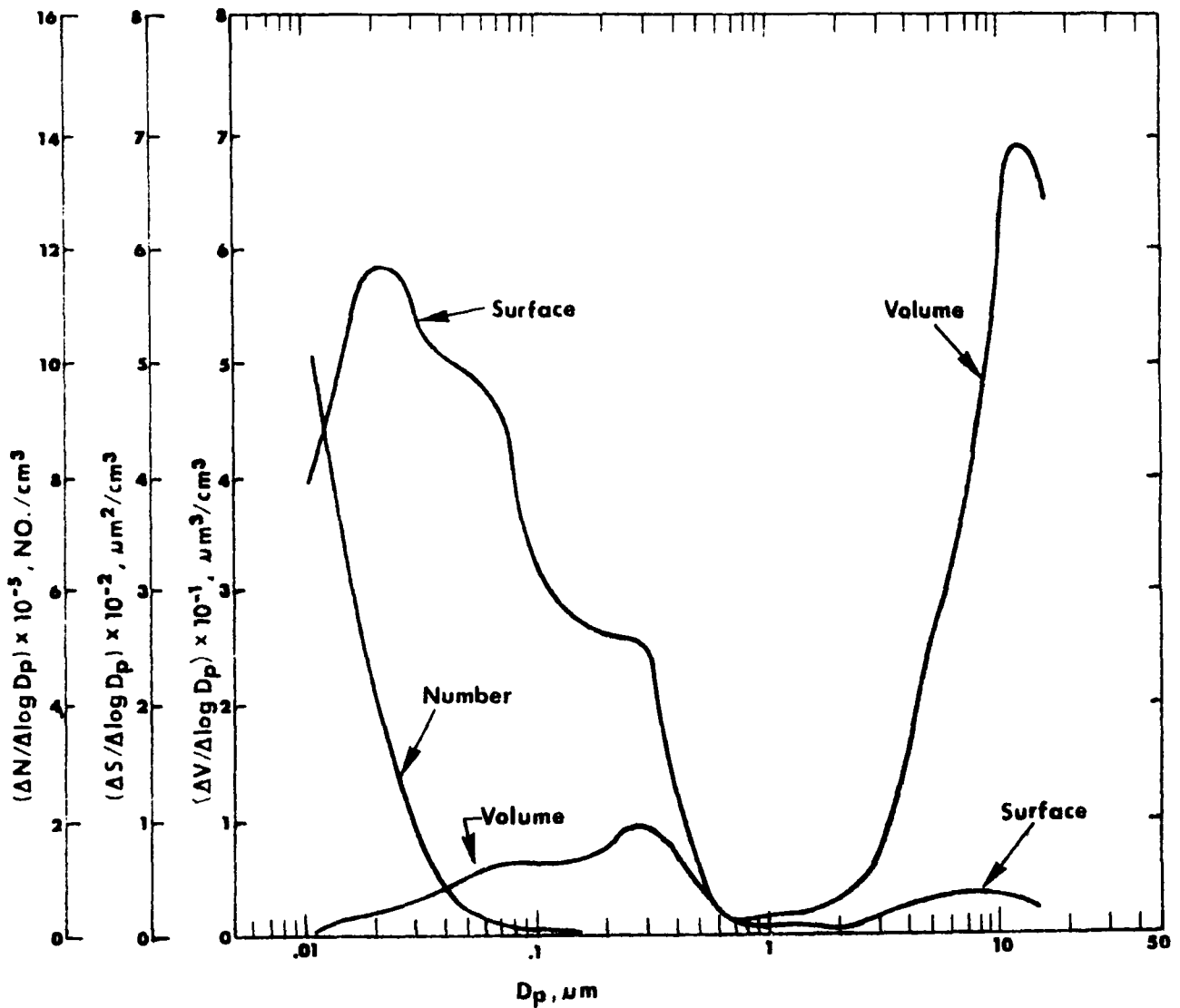


Figure 7-1. Normalized frequency plots of number, surface, and volume distributions for the grand average October 1977 measurements at Denver's City Maintenance Yard. Note the bimodal distribution of volume and the fact that each weighing shows features of the distribution not shown by the other plots.

Source: Reference 97

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7.1 FREQUENCY DISTRIBUTION COMPARISONS

7.1.1 Description of Technique

Particles are most often found to be lognormally distributed in aerosols sampled with cascade impactor type devices. As a result, the size distribution of such data can be completely described in terms of the geometric mean particle size (d_g) and the geometric standard deviation (σ_g). Two samples with the same d_g and σ_g values can be interpreted as being drawn from the same air mass. Therefore, this technique consists of comparing values obtained from two or more different sites, or from the same site for two different time periods or sets of meteorological conditions. Such a comparison can be performed numerically or graphically.

In those cases where smaller particle sizes can be sampled, their distribution is often found not to be lognormal. This results either from the fact that emissions from a dominant impacting source are bimodal or because more than one source (each with a different size distribution) is contributing to the aerosol. Size distributions can be investigated for evidence of such polymodality.

7.1.2 Applicability of Technique

Characterization of the aerosol is the primary use of this technique. Without it, the analyst cannot judge what portion may be respirable, hence potentially hazardous to the public health. Similarly, such data are necessary for investigating the relationship between visibility and suspended particulate concentrations.

The technique is useful as an indicator of the general types of sources that are impacting upon a sampler. Where coarse (i.e., $>3.5 \mu\text{m}$) particles dominate, abrasive sources such as soil dust, fugitive dust, or sea spray can be inferred. When fine particulates dominate, fuel combustion sources and/or secondary particulates may be inferred.

The quantitative impact of specific sources cannot be deduced with this technique. Use of refined instrumentation (diffusion battery, electrical analysis, etc.) can permit the analyst to deduce the quantitative impact of nearby fresh combustion sources.⁹⁷

7.1.3 Relationship to Other Techniques

Given an adequate number of samples, the temporal patterns in particle size distributions could be assessed. Similarly, spatial pattern techniques described in Chapter 3 and techniques assessing the effect of meteorological conditions could also be used in conjunction with particle size distribution data. A statistically adequate number of samples is rarely available to the analyst, however, and conclusions derivable from analysis of the data may be limited to the relative contribution of fugitive and windblown dust.

Source particle size distribution data are quite important in adapting diffusion models to account for deposition and fall-out effects. Certain recently developed models attempt to take these effects into account.²⁸ Furthermore, particle size distributions of emission data can be used to implicate certain source categories (e.g., windblown dust, automotive emissions).

Techniques for analyzing elemental data can also be adapted for use with this technique. Perhaps the most obvious inter-relationship involves the combination of interelement correlation data with particle size distribution data to investigate the relative contribution of soil and fugitive dust sources. Where the soil group (i.e., Al, Si, K, Ti, et al.) intercorrelation is high and the particle size distribution is greatly skewed toward the larger particle size ranges, then the predominance of such sources may be inferred. This is especially true if analysis of meteorological effects shows the expected impact of precipitation. Other chemical/elemental techniques such as chemical element balance and enrichment factor can be used in conjunction with this technique. No clear relationships with the microinventory, trajectory analysis, pattern recognition, and factor analysis techniques can be seen.

7.1.4 Resource Requirements

The resources the analyst needs in order to apply this technique are not severe unless advanced equipment is utilized. The following summary is based upon the use of cascade impactor data. Given in parentheses are estimates of resource requirements when more sophisticated equipment is used.

Manpower	Low (moderate)
Skill	Moderate (high)
Computer	Not required (necessary)
Data	TSP data stratified by size ranges down to 0.5 um (number, surface, and volume distribu- tions down to 0.01 um)

7.1.5 Example Application

Particle size distribution data were collected in Alaska using Andersen sizing head modifications (cascade impactor) to standard high volume samplers.⁹⁹ Totals of 23, 37, and 13 24-h samples were collected from Fairbanks, Anchorage, and Juneau monitoring sites, respectively. The resulting size distributions were then compared to similar data that had been collected for seven large urban areas (see Figure 7-2). Mean particle sizes were much larger for the Alaskan areas than for the cities addressed, thus leading the analysts to conclude that much of the Alaskan particulate matter was not of industrial origin.

7.1.6 References

19, 28, 85, 94, 95, 96, 97, 98, 99, 100

7.2 SPECIES-SPECIFIC SIZE DISTRIBUTION COMPARISONS

7.2.1 Description of Technique

Size distributions for elements and chemicals can be developed with cascade impaction-type devices and used in a number of ways. Urban/rural comparisons can be made, size distributions by wind direction can be produced, enrichment factors can be calculated, and the effect of meteorological conditions can be

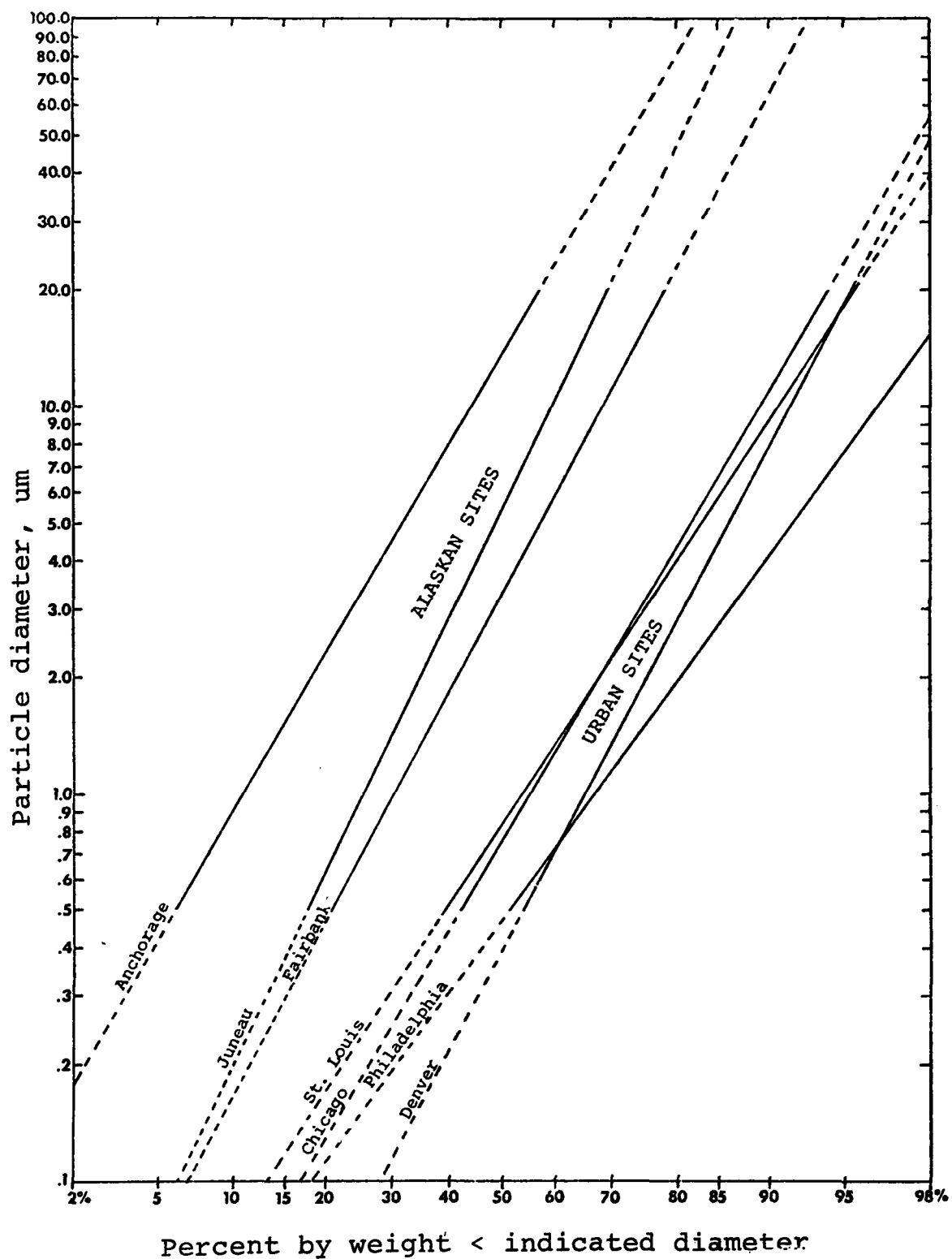


Figure 7-2. Particle size distribution for Alaskan and urban sites.

Source: Reference 99

assessed. Other combinations are possible as well. All that is required is for chemical/element measurements to be made for each particle size range of interest, and for resulting data to be stratified in a manner that appears useful to the analyst.

7.2.2 Applicability of Technique

This technique provides the most refined characterization of the aerosol of any discussed in this digest. Given sufficient data and taking meteorological conditions into account, the spatial and temporal variation in species concentrations by particle size can be described. Limitations imposed by currently available sampling techniques must be noted, however.

Major source categories impacting upon a sampler can be identified with this technique. For example, the impact of soil dust can be assessed by observing variations in Al, Si, and K concentrations in the coarse particulate mode under different precipitation conditions. Likewise, stratifying elemental size distribution by wind directions will permit one to judge whether fuel combustion or abrasive sources are prevalent in each wind direction. The elemental mix can then be related to with likely source types.

It is not yet possible to use this technique to quantify the TSP impact of a specific source or to assess the effect of changes in emissions or the location of a source.

7.2.3 Relationship to Other Techniques

It has already been noted that this technique can be used interactively with most of the others presented in this digest. Exceptions include emission inventorying, microinventorying, trajectory analysis, factor analysis, pattern recognition, and diffusion modeling techniques.

7.2.4 Resource Requirements

It is necessary to have access to a considerable amount of 12- to 24-h cascade impactor data, or to use more sophisticated

size distribution-related equipment. Further, the resources needed to produce species data can be burdensome. The following estimate assumes a 10-site network being analyzed for 20 elements and stratified into size ranges by cascade impactors:

Manpower	Moderate ^a
Skill	Moderate
Computer	Optional
Data	Same as for Section 7.1, plus chemical/element analysis; meteorological data optional

^a Manpower commitment is dependent upon whether the data are subsequently computerized and the number of interrelationships with other techniques that are developed. Laboratory manpower is not included.

7.2.5 Example Application

The example presented herein is drawn from a study of urban aerosols in Toronto, Ontario.⁶³ Concentrations of 25 elements at four different locations were determined with neutron- and photon-activation analysis. Size distributions were produced for most of those elements with a five stage Andersen sampler. Approximately 25 sampling periods of 24-h each were obtained at each site.

Most atmospheric concentrations did not vary from site to site. Exceptions noted were Sb, As, Cr, Cu, Pb, and Zn. The increase in Sb, As, and Pb at one site was attributed to a nearby battery recycling plant. The Cu increase was attributed to brush wear on the high volume sampler. Mass median diameters of each element were estimated by plotting the logarithms of the effective cutoff diameters for each stage versus the cumulative percent mass \leq each cutoff diameter on log probability paper. The elements Al, Ca, Co, La, Mg, Fe, Sm, Sc, Na, and Ti were found to be concentrated on larger particles, to have relatively low enrichment factors (see Table 7-2), and to be well correlated with each other. Other elements, particularly V, Pb, Cl, and Br, were found to be concentrated on smaller particles and, with the exception of V, to have relatively high EF's.

Table 7-2. CLASSIFICATION OF AIRBORNE TRACE ELEMENTS
ACCORDING TO SIZE

Element	Average conc in Toronto, ng/m ^{3a}	Enrichment factor, EF ^b	Approx mass median diam, um ^c	Percent of mass <1.1 um in diam
Aluminum	2100	1.0	8	12
Magnesium	1400	3.6	7	11
Calcium	5300	6.8	7	8
Samarium	0.33	1.9	6.5	21
Iron	2200	2.3	6	15
Scandium	0.27	0.7	6	12
Titanium	170	1.3	6	15
Lanthanum	2.4	2.0	4.7	15
Sodium	650	1.0	4	30
Cobalt	1.0	3.2	4	19
Potassium	870	1.1	2.5	27
Manganese	74	4.0	2.4	36
Copper ^d	<330	<420	1.5	42
Arsenic	12	260	1.5	37
Chromium	26	14	1.3	32
Iodine	<4	<320	1.3	41
Mercury	<5	<6300	1.3	45
Nickel	21	18	1.2	45
Zinc	320	200	1.2	47
Antimony	6.9	1300	1.2	40
Vanadium	14	5.7	0.9	54
Lead	970	2500	0.7	51
Chlorine	1200	150	0.6	55
Bromine	290	3700	0.3	71

^a Average of all Andersen and high volume samples, excluding concentrations of antimony, arsenic, and lead near industrial sources.

^b EF = atmos conc normalized to Al/crustal abundance normalized to Al.

^c Size data are for a typical summer aerosol, excluding samples influenced by local sources.

^d Cu concentration represents an upper limit because of contamination.

Source: Reference 63

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No specific conclusions were drawn as to the sources causing elevated element concentrations in the smaller particle size range, but combustion sources were implied.

7.2.6 References

11, 58, 62, 63, 101, 102

8.0 DESIGNING STUDY AND INTERPRETING RESULTS

A wide variety of individual particulate analysis techniques have been presented in the previous chapters. Each was discussed in terms of its general applicability, resource requirements, and interrelationships with other techniques. That information is necessary in order to permit the analyst to decide whether or not a certain technique applies to the problem he faces and whether or not he can afford to use it. However, more information is necessary for the analyst to integrate these techniques and synthesize the various findings that emerge. In this chapter, the analyst will be presented some guidance as to how to select, apply, and interpret the results of these techniques in light of the specific problem he faces.

8.1 SELECTING TECHNIQUES

Three general questions confront the analyst preparing to study a TSP problem:

- ° What is the general nature of the particulate problem that needs to be investigated?
- ° What techniques can be used to define this problem better and then to resolve it?
- ° How much effort can be expended?

The general nature of the problem at hand can vary considerably. The analyst might be interested in learning how the chemical composition of the aerosol changes over space and time. He might want to define the Pb emission rate from automobile exhaust in terms of g/VMT. He might want to develop a regional control strategy which will ensure the attainment and maintenance

of TSP NAAQS throughout a metropolitan area. Or he might want to investigate a host of other problems.

For the purposes of this section, it will be assumed that the analyst is primarily interested in developing a regional TSP control strategy. This is not meant to denigrate the importance of the other types of problems he faces. Rather, it is simply to focus attention upon the type of problem which will probably be uppermost in the mind of the typical user of this digest.

Once he has generally identified his problem, the analyst will need to define that problem as well as possible and to select those techniques which are most likely to help solve it. This initial decision as to which techniques to use can be aided by the early application of a simple screening procedure. This screening procedure consists of assessing the applicability and task effectiveness of the various techniques, and then assessing their applicability in terms of resource requirements and the sponsoring organization's resources.

The general applicability of each technique, as well as resource-related information which will be explained in the next few paragraphs, is summarized in Table 8-1. Each technique is ranked on a scale of 1 through 5 (where 5 indicates the greatest potential) for each of the general problem areas most pertinent to TSP control strategy development. As that table and Table 8-2 below indicate, different techniques are more effective at different tasks and none of the techniques is effective at all of the tasks.

Table 8-1. SUMMARY OF TECHNIQUES

Technique (Section)	Task rankings			Resource requirements				Cost-effectiveness		
	Charac- terize aerosol	Identify source categor.	Quantify specific src.imp.	Man- power	Skill	Compu- ter	Data	Charac- terize aerosol	Identify source categor.	Quantify specific src.imp.
Long term trends (2.1)	2	1	1	2	1	2	1	.3	.2	.2
Seasonal patterns (2.2)	2	1	1	2	1	2	1	.3	.2	.2
Daily patterns (2.3)	2	1	1	2	1	2	1	.3	.2	.2
Diurnal variation (2.4)	2	2	1	4	1	1	2	.3	.3	.1
Emission patterns (2.5)	1	2	2	4	1	1	2	.1	.3	.3
Site classification (3.1)	3	2	1	4	1	2	1	.4	.3	.1
Inter-site correlation (3.2)	4	1	1	2	2	2	1	.6	.1	.1
Pollution rose (3.3)	2	2	2	2	1	2	2	.3	.3	.3
Upwind/downwind (3.4)	2	1	3	2	1	2	2	.3	.2	.4
Correlation and regression (4.1)	1	2	1	6	3	3	2	.1	.1	.1
Decision-tree (4.2)	1	2	1	6	3	3	2	.1	.1	.1
Precipitation (4.3)	1	2	1	2	1	2	2	.1	.3	.1
Wind speed (4.4)	1	2	1	2	1	2	2	.1	.3	.1
Trajectory analysis (4.5)	2	2	1	6	3	3	2	.1	.1	.1
Emission inventory (5.1)	1	3	1	10	2	2	1	.1	.2	.1
Microinventory (5.2)	1	4	1	6	2	1	1	.1	.4	.1
Diffusion modeling (5.3)	4	4	5	4	4	3	1	.3	.3	.4
Temporal, spatial, and meteorological (6.1)	4	2	2	2	2	2	5	.4	.2	.2
Enrichment factor (6.2)	3	2	1	2	2	1	5	.3	.2	.1
Chemical element balance (6.3)	2	4	1	4	5	3	5	.1	.2	.1
Interspecies correlations (6.4)	4	2	1	4	3	2	5	.3	.1	.1
Pattern recognition (6.5)	4	3	1	4	4	3	5	.3	.2	.1
Factor analysis (6.6)	4	3	1	6	5	3	5	.2	.2	.1
Microscopy (6.7)	4	3	2	8	5	2	2	.2	.2	.1
Frequency distribution (7.1)	3 (4)	2	1 (3)	2 (4)	2 (3)	1 (3)	4	.3	.2 (.1)	.1 (.2)
Species-specific (7.2)	5	4	1	4	2	2	5	.4	.3	.1

Table 8-2. EFFECTIVENESS RANKINGS

Aerosol characterization	Source category identification	Specific source impact quantification
Species-specific size distribution	Diffusion modeling	Diffusion modeling
Intersite correlation	Microinventorying	Upwind/Downwind
Diffusion modeling	Chemical element balance	Emission patterns
Temporal, spatial, and meteorological variations of chemicals	Species-specific size distributions	Temporal, spatial, and meteorological variations of chemicals
Interspecies correla- tions	Emission inventorying	Pollution rose
Pattern recognition	Factor analysis	Microscopy
Factor analysis	Pattern recognition	
Microscopy		

It should be noted that these rankings are necessarily somewhat subjective due to the fact that there are no absolute measures of the relative accuracy of these techniques. One can assess the accuracy of each technique in its own terms. For example, the true correlation among TSP measurements at different sites can be assigned confidence limits which are dependent upon the sample correlation and the number of samples. Likewise, the accuracy of microscopy results can be assessed by analyzing replicate samples. But there is no accepted way of comparing the results of one technique with those of another.

Once he has conducted this initial screening, the analyst will want to know whether or not he can afford to implement those techniques which he considers most promising. To answer this question he can use the information presented in Table 8-1.

Table 8-1 summarizes resource requirements in terms of skill, computer, and data needs. It would be best to present

these data in common units (e.g., dollars) so that the overall resource needs could be calculated in a direct manner. Unfortunately, there can be so much variation in the cost of applying any one technique that this is not really possible. A diffusion model run may, for example, cost anywhere from \$20 to \$1,000+ depending on the complexity of the model and the situation to which it is being applied. As a surrogate for dollar cost, skill, computer, and data requirements are rated at 1 through 5 (5 being most resource intensive). Due to the greater cost that is normally associated with manpower activities, that resource is given twice the weight of the other three and is rated at 1 through 10. It has been assumed that the cumulative resources required to apply a given technique are represented by the sum of four individual resource areas (e.g., the combined resource requirement for the chemical element balance technique is 17).

The analyst must inventory his organization's resources to determine whether or not they are sufficient to apply the set of analytical techniques which he previously identified as being most promising. Those techniques which exceed his agency's resources must be excluded, unless the analyst is able to obtain financial or in-kind assistance from another organization.

In addition to excluding resource-excessive techniques, the analyst must also exclude techniques which are dependent upon those excluded techniques. Diffusion modeling is, for example, dependent upon emission inventorying. If an organization cannot afford to conduct an inventory, then it will not be able to apply a diffusion model.

The analyst will also be concerned about getting the most for his money. Thus, he will be concerned about cost-effectiveness as well as cost or effectiveness alone. By dividing individual task rankings by combined resource requirements, a general assessment of each technique's cost-effectiveness can be derived. The chemical element balance technique, for example, has been assigned task rankings of 2, 4, and 1 for aerosol characterization, source category identification, and source impact quantification, respectively, and a combined resource requirement of 17.

This results in cost-effectiveness ratings of 0.1, 0.2, and 0.1. As shown in Table 8-1, the techniques listed in Table 8-3 below are assessed as being most cost-effective:

Table 8-3. COST-EFFECTIVENESS RANKINGS

Aerosol characterization	Source category identification	Specific source impact quantification
Intersite correlation	Microinventory	Upwind/Downwind
Species-specific size distribution	Species-specific size distributions	Diffusion modeling
Site classification	Precipitation	Pollution rose
	Wind speed	Emission patterns
	Pollution rose	
	Emission patterns	
	Diffusion modeling	

At this point, the analyst will be left with a general set of cost-effective techniques that can provide answers to his specific questions without imposing excessive strain on his organization's resources. He will now want to apply those techniques to the specific problem at hand.

8.2 APPLYING TECHNIQUES AND INTERPRETING RESULTS

Two more specific questions confront the analyst at this point:

- ° In what sequence should the available techniques be applied?
- ° How should conflicting results be handled?

In line with the reasoning previously presented, it is recommended that the available techniques first be used to define the existing problem as explicitly as possible. The resulting definition should be in terms of the regional/local, short-term/long-term, improving/worsening/no change nature of the problem and should be discussed in terms of statistical significance.

Whether or not the problem is long-term and/or short-term should be assessed for each site and described in terms of probability. Hence, the analyst should calculate the probability that the primary annual NAAQS (75 ug/m^3) is exceeded for the entire year and the number of days that the secondary 24-h standard (150 ug/m^3) would be expected to be exceeded during the entire year.

The analyst should then assess whether or not the problem is region-wide or local. This can be accomplished by plotting the measured concentrations geographically, by determining intersite correlations, by applying a diffusion model, by using the site classification process described in Section 3.1, and/or by applying a trajectory analysis. Correlations should be qualified by also determining their true confidence limits (see Figure 1-1).

Thirdly, the trend of past data should be determined. The analyst should determine whether or not there has been a statistically significant trend in annual averages.

At this point, the analyst should have a pretty good definition of the specific problem he faces. For example, he should be able to say that there is a region-wide primary annual NAAQS problem which is worsening or a local short-term secondary 24-h NAAQS problem which is remaining generally stable. And he should be able to attach statistical significance to his definition. However, it may well be that there are not enough sampling sites in the region or number of samples at each site to do so. In this case, the analyst would conclude that he cannot define his problem clearly. If this occurs, he must weigh the consequences of developing a TSP control strategy with an inadequate problem

definition versus the consequences of delaying major control strategy implementation while he conducts a special study which has been designed to fill the gaps in available data.

Once he has defined the problem to his satisfaction, the analyst should determine whether that problem is primarily caused by a clearly identifiable source. To do so, he can compare the temporal patterns of the TSP data with the activity patterns of a suspected source, perform upwind/downwind analyses, apply a diffusion model, calculate a pollution rose, or--if agency resources are sufficient--calculate interspecies correlations, analyze filters microscopically, perform factor and cluster analyses, apply the chemical element balance technique, and/or evaluate the source contributions indicated by the previously applied diffusion model. If there is a major source significantly responsible for measured concentrations, each of these techniques should point out that source.

In some cases, however, results will conflict. When this occurs, the analyst should evaluate the techniques in terms of the statistical significance of their results. Of the techniques identified above, only the microscopy and chemical element balance technique cannot be assessed in terms of confidence limits about the mean. If, for example, the pollution rose fails to indicate a source identified by all of the other techniques, a statistical analysis may reveal that the shape of the rose may not identify that source simply because of the small number of samples upon which each directional mean is based. If the results of the various techniques are each statistically significant but still conflict, then the analyst should see if a clear majority of the techniques indicate one source. If that is the case, he should accept the results as sufficient evidence of that source's role. If there is no clear majority, he should attach more weight to the techniques ranked highest in task effectiveness (see Table 8-1).

If no single source can be clearly identified, the analyst should attempt to identify the source categories which have the

greatest effect upon TSP concentrations. Where the first stage of the study indicated a localized problem, a microinventory should be performed in the vicinity of the violating sampler. Where the problem was regional in nature or where additional evidence in support of the microinventory is desired, it is suggested that species-specific size distributions, the effects of precipitation and wind speed, and the results of pollution rose and emission pattern calculations should be analyzed. Where resources are sufficient, other techniques can be applied as well.

If no source category is shown to be the primary cause of violations, then a special study which takes the techniques discussed in this digest and the resources of the agency into consideration should be initiated.

The analyst may also consider applying some of the other meteorology-related techniques discussed in Chapter 4. Specifically, stepwise multiple linear regression or the AID decision tree technique can be used to determine the percent of variance in TSP concentrations that can be explained by meteorological variables.

In some cases, the analyst will conclude that he has not defined the problem clearly, found a culpable source or source category, or found that meteorological conditions are having an effect. In these cases, he should determine the reasons for this inability and initiate their correction if at all possible.

8.3 EXAMPLE APPLICATIONS

8.3.1 Small City/Simple Aerosol

The first example is a Rocky Mountain town of approximately 50,000 people, located at the juncture of two fairly broad river valleys. Two mountain ranges tightly surround the valley within which the city itself is located. Precipitation is normally rather slight (annual average of 14 in.), but the cool temperatures and low average wind speeds result in a climatic factor of

only about 10 (compared to 100 at Garden City, Kansas). There are large numbers of unpaved roads and lots in the city and surrounding countryside and three large wood products industry point sources located generally upwind of the city. Growth in population is expected to average 2-3 percent per year, but the growth in basic industry is uncertain.

There are two governmental agencies responsible for air pollution control activities in the city: a 2-man local agency, and a 24-man state agency. Given their sizes, these agencies are staffed with a broad range of skills and expertise. The state agency has ready access to computers required for sophisticated analyses. The data base for the area is somewhat limited, consisting primarily of the following:

- ° Four high volume sampler sites, each with more than three years of standard TSP data, and three other sites with less than one year of data
- ° Meteorological data from one site 5-10 miles upvalley of the city
- ° A 1974 area source emission inventory plus updated point source data
- ° No chemical, elemental, or particle size data are available; the agency does have a morphology capability

The two agencies seek to define their TSP problem clearly, to identify major contributing source categories, and to quantify the impact of specific sources. Hence, it is assumed that they would be generally interested in applying all the techniques discussed in this digest. As indicated previously, though, neither agency has sufficient resources to apply each individual technique without access to outside resources. It is difficult to say precisely whether any one technique exceeds the specific resources available to the specific agency in question. For the purpose of this discussion, it will be assumed that manpower resources are exceeded by those techniques ranked 8 or higher in that portion of Table 8-1, and that data resources are exceeded

by those techniques rated 4 or higher unless it has been explicitly noted that the agency has the resources necessary to implement that specific technique. Techniques which are cost-ineffective may also be excluded if the analyst so desires. For illustration purposes, it is assumed herein that cost-ineffective techniques are those which are rated no higher than 0.2 for any one task area and which are rated at 0.1 for the other two areas.

Figure 8-1 illustrates the process of screening out techniques which cannot be used in this example city. According to that figure, 13 techniques survived the exclusion process for the example city. At this point, the analyst must apply these techniques and interpret their results. Table 8-4 suggests how these techniques could be applied to the example city and suggests some qualifications that would probably be attached to the results of each of these techniques.

Creating a hypothetical situation, let it be assumed that the following conditions are found after applying the techniques presented in Table 8-4:

- ° Only one of the sites exceeds either NAAQS. It is located in the downtown area 10 miles down-valley from the major point sources.
- ° A good correlation is found between the two TSP sites which share a common wind direction from the wood products point sources and poor correlations are found for the other sites.
- ° There is a statistically significant slight decrease in annual TSP concentrations at two sites (those which correlated well).
- ° The activity patterns of the major point sources do not correlate well with temporal TSP patterns.
- ° A pollution rose at the downtown site shows no statistically significant pattern.
- ° TSP concentrations at the downtown site are slightly higher than those at the site lying between it and the point sources. The difference is not statistically significant.

Technique	Identify techniques desirable to solve problem	Exclude techniques that are resource-excessive	Exclude dependent techniques	Exclude cost-ineffective techniques	Select remaining techniques
2.1					→
2.2					→
2.3					→
2.4					→
2.5					→
3.1					→
3.2					→
3.3					→
3.4					→
4.1				→	
4.2				→	
4.3					→
4.4					→
4.5				→	
5.1		→			
5.2			→		
5.3			→		
6.1		→			
6.2		→			
6.3		→			
6.4		→			
6.5		→			
6.6		→			
6.7		→			
7.1		→			
7.2		→			

Figure 8-1. Screening techniques for example city No. 1.

Table 8-4. APPLYING TECHNIQUES TO EXAMPLE CITY NO. 1

Task	Applicable technique	Comments
Assess regionality	3.1	Probably not very useful; terrain is essentially the same at all sites
	3.2	Current data will be very precise
Determine direction of past data	2.1	Trends will be difficult to see due to relatively short time period available
Determine whether a specific source is clearly culpable	2.5	Dependent upon analysis of temporal patterns of TSP and some measure of emission patterns
	3.3	Wind direction data may not be representative
	3.4	Wind direction data may not be representative
Determine whether a source category is culpable	2.5	Same as above
	3.3	Same as above
	4.3	None
	4.4	None
Investigate meteorological effects	4.3	Same as above
	4.4	Same as above

- ° A three year old Air Quality Display Model (AQDM) run predicts concentrations fairly well with a correlation coefficient of +0.65. Based upon a four-site network, Figure 1-1 indicates that this represents a true correlation of -0.65 to +0.96.
- ° Since the above results are not conclusive, the effect of meteorological conditions are investigated. TSP concentrations at the downtown site are found to vary with days since precipitation. Wind speed is not shown to have a significant effect.

The cumulative impact of these techniques is to suggest that either fugitive dust or the wood products point sources are dominant causes of TSP violations but that it is not clear which. The initiation of a special study appears warranted.

8.3.2 Large City/Complex Aerosol

The second example is a Southeastern United States bistate metropolitan area of approximately 700,000 people, located along a major river valley characterized by gently rolling hills. Precipitation is somewhat greater than that for the first city, resulting in an annual average rainfall of 43 in. Most roads in the city are paved, but there is a large aggregation of chemical and power generation industrial facilities in the area. Overall, there are more than 100 point sources in the area which emit 25 ton/year or more. Growth in population is expected to average ~2 percent per year, and a large industrial park is planned for an area to the southwest of the city.

There are three governmental agencies responsible for air pollution control activities in the metropolitan area: a 37-man local agency, a 91-man state agency with heavy responsibilities in other portions of its state, and a 104-man state agency. Given their sizes, these agencies are staffed with a broad range of skills and expertise. All three agencies have ready access to computers required for sophisticated analyses. The data base for the area is considerably greater than that indicated for the first example city, consisting primarily of the following:

- Eighteen high volume sampler sites, most of which have more than five years of standard TSP data
- Meteorological data for at least three sites
- An emission inventory which has been updated annually
- Particle sizing data from a number of sites covering periods of six weeks each; monthly composites for SO_4 , NO_3 , and 13 metals at most sites, available from 1971 on; microscopy capabilities are also readily available

Using the same process described in subsection 8.3.1, the best techniques reasonably available for use in this city can be identified. The only exception from the process described in subsection 8.3.1 is that manpower constraints are not assigned due to the relatively large staffs available.

These reasonably available techniques are identified in Figure 8-2. Twenty-two techniques survived this screening process. Table 8-5 suggests how these techniques could be applied to the example city and suggests some qualifications that would probably be applicable to the specific situation described for this example city.

Creating a hypothetical situation, let it be assumed that the following conditions are found after applying the techniques identified in Table 8-5:

- Classifying sites by environment does not produce meaningful results.
- Of the five sites which are exceeding the primary annual NAAQS, two in the industrial area correlate very well together (based upon >50 samples). Correlations among the other sites are <+0.50.
- An AQDM run for the current year predicts TSP concentrations well and correlates with observed concentrations at +0.72, based upon 18 sites. This implies a true r of +0.35 to +0.87 and is considered to be good.
- A steady decrease in annual concentrations is shown at all sites until 1976, at which time a slight increase is observed.

Technique	Identify techniques desirable to solve problem	Exclude techniques that are resource-excessive	Exclude dependent techniques	Exclude cost-ineffective techniques	Select remaining techniques
2.1					➔
2.2					➔
2.3					➔
2.4					➔
2.5					➔
3.1					➔
3.2					➔
3.3					➔
3.4					➔
4.1				➔	
4.2				➔	
4.3					➔
4.4					➔
4.5				➔	
5.1					➔
5.2					➔
5.3					➔
6.1					➔
6.2					➔
6.3				➔	
6.4					➔
6.5					➔
6.6				➔	
6.7				➔	
7.1					➔
7.2					➔

Figure 8-2. Screening techniques for example city No. 2.

Table 8-5. APPLYING TECHNIQUES TO EXAMPLE CITY NO. 2

Task	Applicable technique	Comments
Assess regionality	3.1	May be useful due to river valley conditions
	3.2	None
	5.3	Relatively large number of sampling sites will permit tight calibration assessment
Determine direction of past data	2.1	Large time period will permit good assessment
Determine whether a specific source is clearly culpable	2.5	Dependent upon analysis of temporal TSP patterns and some measure of emission patterns
	3.3	Wind direction data may not be representative
	3.4	Wind direction data may not be representative
	5.3	Same as above
	6.4	Limited number of samples increase confidence limits
	6.5	Broad confidence limits; monthly composite data may create difficulties
	6.6	Same as for 6.5 above
Determine whether a source category is culpable	7.2	Limited number of samples increase confidence limits
	2.5	Same as above
	3.3	Same as above
	4.3	None
	4.4	None
Investigate meteorological effects	5.2	None
	4.3	Same as above
	4.4	Same as above

- ° Pollution roses for the two industrial area sites point in the general direction of those industrial facilities, but no other significant relationships are seen.
- ° One of the violating sites, in a semiurban, largely unpaved area, shows higher concentrations than surrounding upwind sites. Other relationships are not significant.
- ° Another of the violating sites, located downtown, shows a strong TSP versus daily traffic correlation (+0.87). No other significant patterns are revealed.
- ° Diffusion modeling indicates that area sources are the largest contributor to violating sites, but that the point source impact is greatest in the industrial area.
- ° Certain species are shown to co-vary strongly with TSP at various violating sites, but there is a strong probability that the correlation is still zero due to the small number of samples. The downtown site varies with Pb, the industrial sites with V, and a fourth site (located near a cement factory) varies strongly with Ca. No other sites show high correlations.
- ° Limited particle size/chemical data reveal no meaningful relationships.

Based upon these results, the analyst concludes that industrial sources probably cause observed violations at two sites. He feels that more detailed analyses are required for the remaining three violating sites. Subsequent analyses show that:

- ° Concentrations at the downtown and suburban site co-vary strongly with days since rainfall, but not with wind speed. The relationship is much weaker at the site near the cement factory.
- ° Microinventories of those three sites show that there are large amounts of fugitive dust emitted near the downtown and suburban sites, but no such results are shown for the cement plant site.

Based upon these results, the analyst concludes that fugitive dust sources are most probably causing violations at two of the sites. The third may be caused by emissions from the cement plant but any such conclusion would be based upon inadequate data. A special study of that site is recommended.

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APPENDIX A

TECHNIQUES
FOR
ELEMENTAL ANALYSIS

Table A-1. TECHNIQUES FOR ELEMENTAL ANALYSIS^a

Element	NA	PA	XRF	XRFS	XPA	PS	AA	ES
H						X		
He						X		
Li						X	X	
Be						X	X	X
B						X		
C						X		
N						X		
O						X		
F	X				X	X		
Ne	X					X		
Na	X		X	X	X	X	X	
Mg	X		X		X	X	X	
Al	X		X	X	X	X	X	X
Si		X	X		X	X	X	
P						X		
S	X	X	X			X		X
Cl	X	X	X		X	X		
Ar								
K	X	X	X				X	
Ca	X	X	X	X	X		X	
Sc	X							
Ti	X	X	X	X	X		X	
V	X	X	X	X	X		X	X
Cr	X	X	X	X	X		X	X
Mn	X	X	X	X			X	X
Fe	X	X	X	X	X		X	X
Co	X		X	X			X	
Ni	X	X	X	X	X		X	X
Cu	X	X	X	X	X		X	X
Zn	X	X	X	X	X		X	X
Ga	X		X		X			
Ge	X			X				
As	X		X	X	X			
Se	X		X		X			
Br	X	X	X	X	X			
Kr								
Rb	X		X		X		X	
Sr	X		X	X	X		X	
Y					X			
Zr			X	X	X			
Nb								
Mo	X			X				
Tc								
Ru	X							
Rh					X			
Pd			X					
Ag	X				X			
Cd	X		X		X		X	X
In	X				X			
Sn	X		X	X			X	X
Sb	X		X		X			
Te	X							
I	X				X			

Table A-1 (Continued). TECHNIQUES FOR ELEMENTAL ANALYSIS^a

Element	NA	PA	XRF	XRFS	XPA	PS	AA	ES
Xe								
Cs	X				X		X	
Ba	X		X		X		X	X
La	X							
Ce	X			X	X			
Pr	X							
Nd	X							
Pm								
Sm	X							
Eu	X							
Gd	X							
Tb	X							
Dy	X							
Ho								
Er	X				X			
Tm				X				
Yb	X				X			
Lu	X							
Hf	X				X			
Ta	X							
W	X			X	X			
Re	X							
Os	X				X			
Ir	X				X			
Pt	X				X			
Au	X				X			
Hg	X		X	X	X		X	
Tl								
Pb		X	X	X	X		X	X
Bi							X	
Po								
At								
Rn	X							
Fr								
Ra								
Ac								
Th	X							
Pa								
U	X		X					

^a Abbreviations:

- NA = Neutron activation
- PA = Proton activation
- XRF = X-ray fluorescence
- XRFS = X-ray radioactive source
- XPA = X-ray photon activation
- PS = Proton scattering
- AA = Atomic absorption
- ES = Emission spectroscopy

Source: Reference 22

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