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Linda Carol Hedstrom
The University of Montana

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PARTICULATE SOURCE APPORTIONMENT IN MISSOULA, MONTANA:

A Comparison of Two Winters, 1979-80 and 1982-83

by

Linda Carol Hedstrom

B.S. University of Montana, 1970

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Approved By:

Wayne R Van Meter
Chairman, Board of Examiners

Dean, Graduate School

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Date

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Particulate Source Apportionment in Missoula, Montana: A Comparison of Two Winters, 1979-80 and 1982-83

Director: Wayne VanMeter WVM

In 1980, the Montana State Department of Health's Air Quality Bureau conducted a source apportionment study to determine the sources and chemical nature of wintertime total suspended particulate collected from the ambient air in Missoula, Montana. That study was very limited in scope, and did not provide adequate information about particulates throughout the winter season. This project sought to conduct a similar, but more complete source apportionment study of Missoula's ambient air during the winter of 1982-83.

Using elemental data from X-ray fluorescence analysis of particulate samples collected every third day throughout the winter, chemical mass balance software apportioned particulate air pollution among three types of sources. These results were compared with the findings of the 1979-80 study using meteorological regime categorization to adjust for differences in weather during the two study winters.

Based on the conclusions drawn from these analyses, it is unclear whether there had been any measurable change in the particulate constituencies of Missoula's winter ambient air. This finding was most likely due to inadequacies in several aspects of the collection methodologies used prior to the analytical aspects of the project which this paper addresses. These inadequacies are discussed in relation to their effect on interpretation of results, and recommendations for further more complete studies are explored.

ACKNOWLEDGEMENTS

Many individuals supported and assisted me in this research. I wish to thank my committee members, Drs. Wayne Van Meter, Ron Erickson, and Tom Huff for their patience and valuable input and advice. Bob Stevens and other EPA staff and contractors supplied analytical results and advice, without which this project would not have been possible. The Montana Department of Health's Air Quality Bureau generously made available their computer and CMB software which were crucial to the analysis. Dr. John Cooper of NEA, Inc. assisted with the CMB procedure and interpretation. Elaine Bild and others at the Missoula City-County Health Department provided encouragement and moral support, as well as allowing me the time to complete this research. Finally, I wish to thank my friend and husband, Richard Steffel, who patiently typed and edited this work, offered valuable advice and criticism, and supplied inspiration and support when I needed them most.

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CHAPTER 1

INTRODUCTION

Under the rules of the Federal Clean Air Act, Missoula, Montana is a "nonattainment" area for total suspended particulates (TSP), and during winter months regularly exceeds State and Federal 24-hour ambient standards for TSP. In 1980, the Montana Department of Health and Environmental Sciences funded a source apportionment study to determine the primary contributors of Missoula's wintertime particulate pollution. Twelve samples collected at a central urban site were submitted for laboratory analysis using five procedures which provide data for models that allocate air pollution contributions among potential sources.

The results of the five analyses showed some variation, and in a few cases were difficult to compare because source categories were named differently and/or did not include exactly the same sources. Nonetheless, collectively, the analyses concluded that residential wood burning was the primary source of particulates in the fine or respirable range (i.e., < 2.5 microns in diameter), and that urban dust was the primary source of particulate matter in the coarse fraction (>2.5 μm).[1,2,3,4]

Between 1977 and 1983, the Missoula City-County Health Department conducted three surveys to measure residential wood use in terms of the number of area households burning wood and the amount of wood burned. The 1980 survey documented a significant expansion in residential wood burning since 1977 as measured by both parameters. In contrast, the 1983 wood-use survey of the Missoula urban area revealed slight decreases in the number of households burning wood, but also found significant increases in the number using wood as their primary heat source.[5,6] During the same period, sophisticated air monitoring devices showed that many of the high particulate episodes during winters since 1980 consisted mainly of particulate matter in the respirable size range. In fact, under certain meteorological conditions, the respirable particulate fraction comprised more than 90% of the TSP.

The new information about the nature of wintertime particulates and the increasing use of wood for primary residential space heating prompted local pollution control officials to conduct a second source apportionment study. Based on 1980 results and economic constraints, health officials decided to utilize only two of the five analytical procedures used in the 1980 study to provide data to be apportioned by a chemical mass balance (CMB) statistical procedure. This paper briefly describes the chemical analysis phase of this effort and reports the CMB apportionment stage in depth. Finally, it seeks to interpret the results.

The Source Apportionment Process

The source apportionment process includes the following: (1) identification of all probable contributing sources, (2) collection of source-specific materials (emissions) for analysis, (3) selection of one or more suitable receptor sites, (4) ambient particulate sample collection, (5) laboratory analysis of both source and receptor samples, and (6) apportionment among the various sources using chemical mass balance, factor analysis or some other statistical method. It is the final phase of this process which is the major topic of this thesis.

In source apportionment studies, results of a variety of procedures for analyzing particulate chemistry are used to develop representative source "fingerprints" based on the relative concentrations of chemical components of each source's emissions. These fingerprints can then be used in computer programs which "fit" ambient sample data to "known contributor" data. For example, this project uses chemical mass balance, which apportions pollutant concentrations at receptor sites among potential sources using a statistical least squares fitting procedure.

Once accomplished, source apportionment information can be used by regulatory agencies and/or pollutant contributors to develop pollution reduction strategies. Further, subsequent source apportionment studies can be used to evaluate the efficacy of pollution control programs. Since this project is the second of two source apportionment studies completed in the Missoula airshed, an important application of these results is a comparison with the 1980 findings. For this project, I accomplished such a comparison through application of a procedure called Meteorological Regime Categorization (MRC) which can "adjust" for meteorological variation within and between the two study periods.

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CHAPTER 2

LITERATURE REVIEW

This chapter examines the evolution of the source apportionment study utilizing receptor modelling or related techniques. Past works employing chemical mass balance are reviewed, and analytical (chemical) techniques which provide data to the models are described. One section surveys works which incorporate meteorological factors into the data analysis, and the final section details air pollution studies carried out in Missoula.

Receptor Model Source Apportionment

Receptor Versus Dispersion

Two mechanisms which can be used to estimate the effect(s) a specific pollution source has on ambient particulate levels at a monitoring (receptor) site are source (dispersion) models, and receptor models. Source models utilize pollution emission rates, pollutant dispersion patterns, and meteorological factors to project where air contaminants will go after emission by mathematically predicting concentrations at monitoring sites. In contrast, receptor models measure ambient levels at a receptor site and then work retrogressively to apportion contributions among suspected sources.

Receptor models were developed for use as an alternative to source models, which work adequately for stable gaseous pollutants when supported by accurate emission inventories and sufficient meteorological data.[1] However, Friedlander (1973) found that use of source models to predict the dispersion of particulate matter is complicated by atmospheric transformations (e.g., condensation and evaporation) which may alter pollutants after emission. Watson (1981) elucidated several other reasons why receptor models are preferable to source models for defining relationships between source contributions and ambient particulate levels. These include the following factors which cannot be taken into account using current source models: (1) particulate emissions, especially those from area sources, may be widely dispersed and therefore difficult to quantify; (2) differently sized particles remain airborne for varying amounts of time (e.g., large particles may settle out close to a source, while small particles may be transported long distances), and (3) minute particles may act as condensation nuclei or otherwise react with organic vapors to alter the chemistry and the dispersion characteristics of the pollutants. The contrasting approaches used by source and receptor models are illustrated in Figure 1.

Chemical Element Balance

Some of the first investigators to use a receptor oriented analysis were Miller, Friedlander, and Hidy, when in 1972, they estimated source contributions to ambient particulate concentrations in the Pasadena, California airshed

SOURCE IMPACT ASSESSMENT MODELS

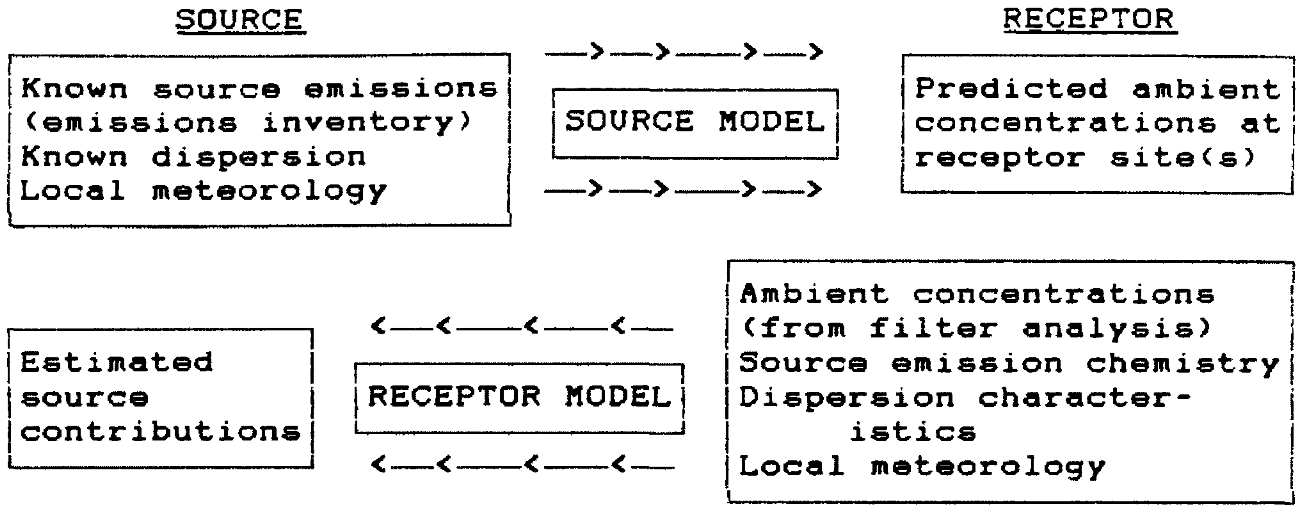

FIGURE 1

Diagrammatic representation of source and receptor models. Adapted from Watson (1979), and Cooper and Watson (1980)

using a procedure called chemical element balance (CEB).[2] Such early efforts were called "tracer studies" because they employed only one or two elements as chemical indicators of each potential contributing source. For instance, Friedlander and Hidy used lead and silicon as indicators for automobile exhaust and geological dust, respectively, in a Los Angeles pollution study.[3] In 1973, Friedlander established the CEB technique as an important tool in air pollution analysis when he improved the CEB methodology by developing a "source concentration matrix" which identified the major sources in a pollution equation using as many as 23 chemical species for each source.[4] This source matrix evolved into the source "fingerprint" used in subsequent CEB equations.

The fundamental assumption in the CEB method is that a source (or source type, e.g., kraft recovery boiler) emits specific chemical species in relatively static, characteristic proportions and concentrations.[5] CEB further assumes that this distinct fingerprint is conserved in the atmosphere from the point or points of emission to the receptor site. Based on these assumptions, CEB apportions the pollutants measured at a receptor site by solving simultaneous equations utilizing all of the following factors: (1) the total mass of the particles collected at a receptor site, (2) the mass concentration of each chemical species, (3) the number of contributing particulate sources, and (4) the identity and relative proportion of each of the chemical species emitted. That is, CEB mathematically "balances" the measured concentrations of chemical species at a receptor in relation to each other, and the estimated concentrations of those same species emitted at the different sources.[6] The mathematical computation which accomplishes this balance is called a least squares fit. While this statistical treatment does not reveal absolute percent contributions from each source, it does suggest the best combination of sources whose emission patterns come closest to explaining the chemical composition of particles collected at the receptor site.[7]

Other Models

During the period while Friedlander and his colleagues were perfecting the chemical element balance receptor model, at least four other types of pollution apportionment models were also being used. King et al. (1976) employed an "enrichment factor" model in which ambient "background" composition was determined, and then each element's relative enrichment (i.e., the amount contributed by pollution) was calculated. In a 1974 California study, Giauque et al. used a time series model based on the assumption that, ". . . chemical species originating from the same source will have the same time dependence when measured at a receptor."[8] For example, when the composition of ambient samples is tracked over time (e.g., hourly through several days, or daily through several years), they may show a definite pattern from which possible contributing sources may be inferred.

Neustadter et al. (1976) applied a spacial model which considered elemental compositions of ambient particles sampled at the same time but at various geographic locations. This method reveals possible sources through comparisons of the distribution of elemental concentrations at the receptors to the chemistry of emissions at each source, given reasonably constant wind direction patterns during the sampling period. This approach to receptor modeling is generally referred to as "spacial," but may also be called by the technique or tool used: cluster analysis or pollution wind rose.

CMB Critique and Response

Gordon (1980) found three major weaknesses in the chemical mass balance (CMB)* receptor model which could be avoided using yet another approach, multivariate analysis. He listed the CMB's weaknesses as follows: (1) only a few sources had (at that time) been thoroughly characterized chemically; (2) much of the point source sampling had been accomplished by collecting in-stack emissions which might not have reflected particulate composition once pollutants exited the stack; and, (3) similar sources were generally referred to in groups rather than as individual sources.

In 1980, the above criticisms were generally true. However, since then the bases for these problems have, in most instances, been eliminated. For example, one extensive CMB study resulted in the development of a source fingerprint library which is still being used in subsequent projects.[9] Further, new in-stack sampling techniques more closely approximate field conditions by diluting emissions with clean air at a ratio of 1:6, and by reducing the temperature of the sample.[10] Additionally, using results of extensive source samples, several investigators have gone so far as to be able to pinpoint individual stack emissions

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* Current literature refers to Chemical Element Balance or CEB as Chemical Mass Balance or CMB. I use the more modern term for the rest of this work.

and/or upset incidents from within large industrial complexes.[11,12] Finally, computer programs developed specifically for CMB analysis calculate not only source contributions, but also confidence intervals for results. Used together, these mitigate some of the uncertainties due to changing atmospheric conditions during sampling and unavoidable errors in sampling and analytical instrumentation and procedures.[13]

Gordon (1980) believed multivariate factor analysis to be a more useful tool than CMB analysis because investigators need not make "a priori" assumptions concerning the composition of emissions from contributing sources. Further, the factor analysis data set is not limited to ambient and source sample chemistry, but may also include meteorological data and/or other information potentially relating receptor site impacts to possible contributing sources. Factor analysis "normalizes" receptor site elemental concentrations relative to each species' mean and variance. These normalized values are then treated statistically in relation to other parameters to detect common factors which account for the observed variance and thereby indicate pollution sources.

Gordon acknowledged some weaknesses due to data normalization to include the following: (1) the investigator cannot work with real values, only variances, which do not reflect temporal or spacial differences in composition;

(2) the investigator cannot extract concentrations of individual species; and (3) the investigator cannot resolve differences among sources with emissions of similar chemistry. Given the improvements that have been made in recent years in CMB and the continuing weaknesses of factor analysis, CMB seems to be the most effective source apportionment method now available.

Receptor Applications

Numerous studies have utilized one or more of the above receptor models for apportioning particulate pollution among major potential sources. In addition to those already mentioned, other early source apportionment studies were conducted by Morrow and Brief (1971) in New York City, Martens et al. (1973) in San Francisco, Gladney et al. (1974) in Boston, Draftz (1975) in Chicago, and Scott Environmental (1975) in Philadelphia. Within the last ten years, source apportionment studies have been accomplished in Charleston, West Virginia, by Lewis and Macias (1976), St. Louis, by Gatz (1978), and Washington, D.C. by Kowalczyk (1978). Cooper and Watson (1979) conducted an extensive CMB study of the Portland area in which emissions from 28 sources were analyzed for 27 chemical species. This single study created a large source fingerprint library for use in subsequent studies.[14] Denver's "brown cloud" was thoroughly examined in the 1978-80 Denver Haze Study.[15,16] Finally, Cooper and DeCesar carried out a CMB study in Medford, Oregon using the methods perfected in the Portland study.[17] In Montana, source apportionment studies have been completed in Billings, Butte, Libby, East Helena, Columbia Falls, and Missoula.[18]

Laboratory Analyses for Receptor Studies

Four criteria by which to judge the suitability of analyses for source apportionment are as follows: " . . .

> 1) The analytical method must have been developed and tested specifically for analysis of suspended particulate matter;
>
> 2) [the method must be able to quantify concentrations at levels lower than expected from the ambient samples to be examined];
>
> 3) The method must be free of biases for all components to be quantified; and
>
> 4) The values [derived] by the method must be reproducible within defined and reasonable confidence intervals."[19]

Other desirable characteristics of an analysis technique include reasonable cost, low interference, small sample size requirement, good comparison with other methods, and conservation (i.e., non-destruction) of the sample. Table 1 summarizes the analytical methods used in source apportionment studies reviewed here.

Chemical Analyses In Source Apportionment

As shown in Table 1, instrumental neutron activation analysis (INA) is frequently used as an analytical procedure in source apportionment investigations. In INA analysis,

TABLE 1

Common Analytical Methods for Particulate Samples

Chemical Species Quantified	Analytical Method	Source	Studies Using Method	General Comments[a]
Elemental (atomic no. > 11)	INA[b]	Zoller et al. (1970) Dams et al. (1970)	1,2,5,6,10,12	Cost-effective; non-destructive; low detection limits; can quantify many "rare earth" elements other methods cannot
	AAS[c]	Ranweiler and Moyers (1972)	1,6,15	Destroys sample; good reproducibility
	XRF[d]	Giauque et al. (1974)	3,4,7,10, 11,12,13	Cost-effective; non-destructive; biased for some elements; specific collection media required
Carbon	VFI[e]	Johnson and Huntzicker (1978)	10,12	Reproducible; differentiates between elemental and organic carbon
	Serial Extraction	Appel et al. (1976)	8,9	Differentiates between primary, secondary, and insoluble carbon compounds[f]
Ionic	IC[g]	Small (1975)	7,10,12	Destroys sample; multi-element technique

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[a] Comments taken from Watson (1981), except where noted.

[b] Instrumental Neutron Activation

[c] Atomic Absorption Spectrophotometry

[d] X-ray Fluorescence

[e] Volatilization and Flame Ionization

[f] Comments from Appel et al. (1978)

[g] Ion Chromatography

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8. Kneip et al. (1978)
9. Appel et al. (1978)
10. Cooper and Watson (1979)
11. Dzubay (1980)
12. DeCesar and Cooper (1981)
13. Spengler and Thurston (1983)
14. Cass and McRae (1983)
15. Scheff et al. (1984)

elements in a particulate sample are quantified by placing the sample into a neutron flux from a nuclear reactor. The elements absorb neutrons and transmute into radioactive species. The number of elements that transmute is proportional to the number present on the filter. Each radioactive element then emits gamma radiation with an intensity proportional to the number of atoms present, and at a wavelength which is specific for that element. As the radioactivity decays, the gamma emissions are counted and compared with the spectrums of known elements.[20]

A major benefit of INA is that the procedure is non-destructive; and samples can be irradiated numerous times to verify results and may also be subjected to other analytical techniques. It has a distinct advantage over X-ray fluorescence (XRF) in being able to quantify sodium and magnesium. Further, aluminum presents special interference problems in XRF analyses which do not occur in INA. The chief disadvantage is the amount of time it takes to complete the procedure for some elements. For example, iron, cobalt, nickel, zinc, and several other elements require 20-30 days to complete the radiations counts.[21]

Atomic absorption spectrophotometry (AAS) is a relatively simple yet effective method for detecting trace metals in atmospheric particulate samples. Until 1974, the method had limited use in air pollutant analysis because the particle collection filters had high background amounts of some of the same elements being quantified; and, since sample preparation requires complete dissolution in an aqueous solution, relatively insoluble elements were not detected.[22] Ranweiler and Moyers (1974) revised an AAS procedure which allows 22 trace elements important in source apportionment studies to be quantified. In their version, particulate matter collected on polystyrene filters is dry-ashed at 400-425°C, and then dissolved in a mixture of HCl, HF, and HNO_3. At least five aliquots are prepared with different concentrations of the acids for detecting distinct elements. Each solution is then volatilized and analyzed by AAS using appropriate hollow cathode lamps as a radiation source. When the "atomized" elements are irradiated, many are excited to an elevated energy state, thus removing energy from the light beam. This loss of energy is detected and recorded by the spectrophotometer. While this technique gives excellent results which compare well with other methods, it destroys the sample and is therefore used less frequently than non-destructive analytical methods.

X-ray Fluorescence

Numerous source apportionment studies have employed X-ray fluorescence (XRF) to analyze thousands of particulate samples. This type of analysis is favored over others because it can detect very low levels of most elements with atomic numbers greater than 13, and it can produce precise results rapidly without damaging the sample. In general terms, the XRF procedure uses excitation radiation produced by X-rays to energize and eject inner (K shell) electrons in the elements of a particulate sample. Then, the replacement of K shell electrons with L shell electrons produces the emission of characteristic X-rays, which identifies the elements to a detecting device.[23,24,25,26,27] Additionally, many XRF instruments, such as the one used in this study, interface directly with computers which assist in correcting for interferences and which calculate elemental concentrations as well as their uncertainties.[28]

XRF analysis cannot quantify some elements and compounds which may be essential to a comprehensive source apportionment analysis. For example, concentrations of sodium and magnesium must be supplied by some other technique. Further, quantification of elemental and organic carbon, which is crucial in any study involving two or more carbon sources, cannot be accomplished by XRF. Consequently, in many source apportionment studies where XRF has been used as the main analytical technique, other methods have been used to supplement the necessary data base.

Carbon Analysis

Two accepted methods for quantifying the various types of carbon in particulate samples are volatization flame ionization (VFI) and serial extraction (see Table 1, above). Johnson and Huntzicker (1978) developed the VFI technique specifically to measure the carbonaceous content in ambient particulate samples. Their procedure involves volatilizing the organic portion of the total carbon in a helium atmosphere, oxidizing the carbon to CO_2, reducing it to CH_4, and then measuring the CH_4 with flame ionization. The elemental component of the total carbon is oxidized to CO_2 in an oxygen atmosphere, separated from the oxygen chromatographically, reduced to CH_4, and then quantified by flame ionization. At the time the Johnson and Huntzicker paper was published, the authors were in the process of modifying this method to facilitate the differentiation of carbon into three components: carbonate carbon, organic carbon, and elemental carbon.[29]

Carbon analysis of particulate samples by serial (solvent) extraction was developed by Appel et al. (1976) for a California air pollution study. In this method, the carbon is fractionated by a series of extractions into three components defined as primary carbon, secondary carbon, and elemental carbon. Primary carbon, extracted with cyclohexane, is that carbon which a source discharges directly into the atmosphere. Secondary carbon, extracted with benzene and methanol-chloroform, is the carbon in compounds which are formed by chemical reaction after emission. Elemental carbon is all the carbon that remains after the two previous extractions have been accomplished.

Serial extraction has the advantage over the VFI method in that extracted material can be further analyzed to identify individual compounds. However, extraction is time consuming and expensive, and requires large amounts of sample. Alternative methods of measuring carbon are being explored, and may utilize assessment of the light absorption coefficient of a filtered sample using laser transmission devices and photoacoustical techniques. Research in these areas has not yet been completed.[30]

Ion Analysis

In addition to elemental species, the ionic components of an aerosol (e.g., sulfates, nitrates, ammonium) should also be quantified in a comprehensive source study. This is especially true in the case of sulfates, because simply quantifying elemental sulfur in a particulate sample does not account for this element's chemical reactivity in the atmosphere. For instance, sulfur dioxide, under favorable conditions such as a particulate-laden atmosphere with warm temperatures and high relative humidity, quickly forms other sulfur compounds (e.g., sulfates, most of which occur in the fine particulate fraction).[31] These secondary products complicate the interpretation of the CMB results with respect to sulfur emissions. Further, source apportionment studies which overlook sulfur compounds ignore EPA findings that in the United States, sulfates comprise the predominant particulate species in the <1 micron size fraction.[32]

One technique for measuring the anion content of a particulate sample was developed by Small et al. (1975). This technique involves injecting an aqueous extract of the sample into a liquid ion exchange chromatograph which has been calibrated to quantify chloride, bromide, sulfite, nitrate, and sulfate. Measurement of cations, such as H^+ and ammonium, is usually accomplished with ion specific electrodes.[33]

Determination of Sample Mass

To conduct a CMB analysis, the particle mass must be determined for each sample. Mass measurements can be made gravimetrically using analytical balances which weigh to 10^{-6} gram, or by a technique called ß-attenuation, which deduces mass by observing the change in counting rate when a particulate-laden filter is inserted between a beta emitting source and a detector. Gravimetric methods require considerable handling of the sample which can result in errors due to mass loss and/or label or filing mistakes. In addition, in large studies with potentially thousands of specimens, handling and weighing individual samples is extremely tedious and time consuming. In contrast, ß-attenuation automates the mass determination procedure, eliminating the potential for human error due to handling and tedium. However, only films and membrane type filters with a thin layer of particles can be analyzed by this method.[34]

Courtney et al.(1980) examined mass measurements determined gravimetrically and by ß-attenuation and found them equivalent, provided the filter orientation in the ß-gauge and laboratory atmospheric conditions were carefully controlled. Dzubay (1980) and Spengler and Thurston (1983) utilized ß-attenuation for mass measurements in their respective studies.

Meteorology and Air Pollution

Meteorology affects air pollution quantitatively and qualitatively on both micro and macro scales. Local weather directly influences pollutant dispersion patterns, which in turn affect pollutant levels at a receptor.[35] Regional weather alters local pollutant chemistry and concentrations by transporting pollutant-laden air masses and by facilitating or retarding atmospheric transformations.[36] Weather also indirectly affects air pollution through its influence on human behavior (e.g., colder temperatures promote increased driving and increased heating demands).[37]

Dispersion (source) studies using standard models, such as the Box model and the Gaussian Plume model, have for many years recognized the importance of weather in air pollution equations. In this regard, most source models consider at least those meteorological factors such as wind velocity which directly affect a plume as it leaves a point source stack.[38] In contrast, few source-receptor studies have fully considered the air pollution/meteorological relationship.

Certain obstacles confront an investigator seeking to employ pertinent, local meteorological factors in an air pollution study, not the least of which is whether the data has even been collected. For example, Zeldin and Meisel (1978) found that while many large urban centers had meteorological stations nearby, receptor studies which included smaller cities or rural areas often suffered from a lack of a weather station sufficiently close to the monitoring site. This situation necessitates the use of regional weather data (which may or may not apply to a site, or may require adjustment) or the installation of a meteorological station at or near the study site.

A second concern is whether available meteorological data represents normal patterns for the study area (e.g., in terms of frequency distributions and averages).[39] Thirdly, in complex terrain (e.g., a mountain valley) or near a large body of water (which may affect weather over the land mass), the location of the meteorological station with respect to the particulate monitoring site becomes even more important.[40] If the meteorological station is not located at the pollutant monitoring site, the applicability of "distant" meteorological data to that site must be verified statistically.[41]

A fourth consideration in using meteorological factors in the statistical analysis of the data, is potential colinearity between or among meteorological factors. For example, temperature, relative humidity, and dew point all have some effect(s) on ambient particulate levels, but because there are also interrelationships among the three variables, they cannot be considered independently during statistical analysis.[42] Finally, the investigator must consider possible disparate effects a given meteorological factor may have on different pollutants. For instance, cold temperatures inhibit the transformation of SO_2 to sulfates, so during winter months, SO_2 levels appear to increase; oxidant concentrations, on the other hand, tend to decrease during cold weather.[43]

In addition to overcoming problems of the applicability and interpretation of meteorological data, a researcher must choose among a variety of ways for utilizing such information. For instance, in source model studies, meteorological data such as wind speed, wind direction, and atmospheric stability are included in standard models for predicting pollutant dispersion(s) and resulting ambient concentrations.[44] In contrast, there is no standard approach for incorporating weather factors into receptor studies. For example, Evans et al. (1981) examined the relationship between particulate levels and wind speed, and constructed several predictive receptor models using these and related factors. Cooke and Wadden (1981) demonstrated the consistent relationship between relative humidity and levels of SO_2 in developing a predictive model for sulfate levels in

Chicago. Other studies have simply used meteorological information to assist in explaining why pollutants appear in the levels they do at one or more receptor sites.[45,46,47]** For example, Robbins (1981) used meteorological variables in his factor analysis source apportionment study of Billings, Montana, while Scheff et al. (1984) employed wind direction to validate the results of a chemical mass balance source apportionment study in Chicago.

Other researchers have utilized meteorological information "prospectively," examining meteorological patterns in a study area in order to make decisions of when and how often to collect aerosol samples. For example, Cohen (1977) examined historical weather data in the Portland area and determined the frequency with which different weather patterns occurred. He then characterized those patterns into distinct classes based on air mass stability and wind direction. Using Cohen's work and other meteorological studies, contractors conducting the Portland Aerosol Characterization Study were reasonably assured that samples were collected on climatologically representative days.

In yet another application, Houck (1981) suggested a means for using meteorological classifications to adjust short-term, small sample projects in relation to "normal" weather in the study area. This is often necessary because,

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** These types of investigations are "relationship" studies which usually employ one or more statistical tools such as simple or multiple regression analyses.

in contrast to usual EPA approved sampling strategies,*** short-term (e.g., seasonal) sampling schedules can bias results due to meteorological aberration. Houck recommends adjusting mean pollutant concentrations based on the frequency of meteorological patterns (which have been segregated into distinct regimes) during the study period.[48] This creates a weighted average and eliminates sampling biases introduced by meteorological effects on a limited sampling schedule. This technique is discussed in depth in the Methods chapter.

Missoula Specific Studies

Emission Inventories

A document submitted to the Montana State Board of Health in the early 1960s recognized Missoula's distinctive topography as one which would increase the likelihood of frequent inversions and stagnant air conditions.[49] The same study reported an annual particulate average of 158 micrograms per cubic meter ($\mu g/m^3$) for Missoula which compared to an average of 104 $\mu g/m^3$ for all other Montana cities included in the study.**** The study included an emission inven-

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*** EPA allows a sampling schedule of once every three to six days to determine compliance with federal ambient standards. This type of sampling schedule, will, over a long period of time, randomly sample for pollutants in most, if not all, kinds of weather.

**** Only part of the original 1960s study, that part pertaining to Missoula, still exists. Therefore, information regarding sampling methods and frequency, is not availtory which identified major source groups for Missoula's combustion-caused pollutants and "quantified" each source group's contribution. These estimates are listed in Table 2.

TABLE 2

1961 Emission Inventory

Pollution Source	Tons/Day[a]	Percent of Total
Industrial and Commercial[b]	45.9	27
Domestic[c]	11.3	7
Vehicular[d]	111.4	66
TOTAL	168.5	

[a] Tons per day averaged over a year; includes gases and particulate matter

[b] Includes only the burning of fuels such as wood, fuel oil, natural gas, and wastes (no fugitive emissions)

[c] Includes only the burning of fuel oil, natural gas, coal, and wastes

[d] Includes gasoline and diesel

A second emission inventory compiled in 1974, expanded the list of pollution sources and examined particulate matter separately from gases.[50] Using this compilation as a base, Otis (1977) projected source contributions for 1980 and 1985 and, in addition, calculated a wintertime emission inventory. Finally, Church (1981) revised Otis' source contribution estimates based on new information on source emis-

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able. When daily monitoring for TSP began in Missoula in September, 1969, using the EPA reference method (high-volume sampling), monthly particulate averages were within the same range as those reported in the 1961-62 study.

sion factors and residential wood use. Table 3 presents the winter emission inventories compiled by Otis and Church.

TABLE 3

Winter Months Emission Inventories
(120 days)

	1974[a]		1980[b]	
Source	Tons/Day	% Total	Tons/Day	% Total
Fuel combustion[c]	1.08	11.1	1.24	12.1
Transportation	.49	5.0	.61	5.9
Unpaved roads	.75	7.7	.47	4.6
Paved roads	.51	5.2	1.70	16.5
Point sources	4.89	50.0	1.25	12.2
RWC[d]	2.05	21.0	5.01	48.7

[a] From Otis, 1977, based on estimates from PEDCo, 1975

[b] From Church, 1981, using recalculated numbers from Otis, 1977

[c] Does not include residential wood burning, or large point sources located outside the urban airshed; e.g., Champion Mills in Frenchtown and Bonner

[d] Residential wood combustion

Wood-Use Surveys

In its 1974 study, PEDCo estimated that residential wood combustion (RWC) contributed 246 tons of particulate matter per year, most of which was emitted during the winter months.[51] By 1976, Missoula's pollution control officials began to suspect that PEDCo's estimate was no longer valid because of rising particulate levels observed during the winter. In 1977, Otis conducted Missoula's first wood-use survey, and found that over 8,000 households produced 292 tons of particulate matter during the winter the survey was conducted.[52] In 1981 Church completed a second wood-use survey which was somewhat expanded, but similar in design to Otis'. The results of the second survey revealed substantial increases in the size of Missoula's wood-burning population as well as increases in the total amount of wood burned.[53] The most recent wood-use study (Steffel, 1983), which was greatly expanded over previous designs, found that while the number of wood-burning households had levelled off, the number of households using wood as their primary fuel source had increased significantly since 1980.[54] Steffel documented several instances where survey respondents gave false answers to some of the survey questions, necessitating upward revisions in some of his results by 10% (a "falsity factor").[55] Table 4 summarizes results of the three Missoula wood use surveys.

Particulate Source Studies

In 1979, the Montana Department of Health and Environmental Sciences funded a source apportionment study in the Missoula airshed to determine the source or sources of wintertime particulate matter. Four independent laboratories conducted different analyses of particulate matter collected during February and March 1980, at the Lion's Park monitoring station. XRF and carbon analysis were conducted by Cooper (1980); Moyers et al. (1980) analyzed particulate samples using wet chemistry methods; Davis (1980) used XRD to examine particulate matter; and optical analysis using a

TABLE 4

Changes in Wood Use for Residential Space Heating

Year	Number of Wood-Burning Households	Percent Change From '77	Number Primary Burners	Percent Change From '77	Tons of Wood Burned	Percent Change From '77	Tons of TSP Produced	Percent Change From'77
1977[a]	8,029	—	433	—	23,366	—	292	—
1980[b]	11,667	45.3	3,150	611.1	52,195	123.4	601	105.8
1983A[c]	11,568[d]	44.1	4,516	943.0	41,522[e]	77.7	1,316[f]	350.7
1990[g]	18,280	127.7	N.C.[h]	—	95,776	309.9	N.C.	—

[a] Source: Otis, 1977

[b] Source: Church, 1981

[c] Source: Steffel, 1983; All figures are thought to be conservative. The survey was conducted during a period of controversy concerning regulations on residential wood burning. This may have adversely affected responses to the survey.[56]

[d] Number adjusted upward by the 10% "Falsity Factor."

[e] This figure is thought to be lower than the 1980 figure because the 1982-83 winter was milder than the 1979-80 winter and due to the lack of "true" responses by some survey participants.[57]

[f] Steffel used emission factors (pounds of particulate matter produced per ton of wood burned) considerably higher than those used in previous surveys. This complicates strict comparison of these results. See Steffel, 1983, pp 35-38 for further information.

[g] Projections from Steffel, 1981, based on wood use survey by Church, 1981.

[h] Not calculated.

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polarized light microscope was conducted in the laboratory at the Missoula Health Department. Table 5 shows the results of these analyses.

TABLE 5

Results of Chemical Analyses in Missoula's 1980 Source Apportionment Study

PARTICULATE FRACTION	ANALYSIS	SOURCE CATEGORIES AND PERCENT CONTRIBUTIONS				
TSP	XRD[a]	Wood Combustion	Soils & St. Sanding		Sulfates	Unidentified
		53	31		3	13
	Optical Microscopy[b,c]	Combustion	Mineral		Biological	Unidentified
		26	56		16	2
COARSE FRACTION (>2.5µm)	XRF[d]	Wood Combustion	Mineral		———	Unidentified
		17	54		———	28
	Optical Microscopy[e]	Combustion	Mineral		Biological	Unidentified
		35	60		5	0
	Wet Chemistry[f]	Organic	Soil & Crustal		Ionic	Unidentified
		20	90		2	0
FINE FRACTION (<2.5µm)	XRF	Wood Combustion	Auto Exhaust	Hog Fuel	Mineral	Unidentified
		68	6	2	9	15
	Optical Microscopy[g]	Combustion	Mineral		Biological	Unidentified
		76	24		0.3	0
	Wet Chemistry	Organic	Mineral		Ionic	Unidentified
		75	9		8	8

[a] Source: Davis, 1980 - Analytical Method: Factor Analysis

[b] Source: Hedstrom, 1980

[c] Particles examined were ≥1.5µm in diameter

[d] Source: Cooper and DeCesar, 1980 - Analytical Method: CMB

[e] Particles examined were >3.5µm in diameter

[f] Source: Moyers et al., 1980 - Analytical Method: Bivariate Linear Regression; Enrichment

[g] Particles examined were ≥1.5µm ≤3.5µm in diameter

Air Pollution and Health Studies

Two health studies have been conducted in the Missoula area, both of which examined the potential health effects of air pollution. The first study, completed in the early 1970s by the University of Montana Student Environmental Research Center (SERC), investigated possible relationships between upper respiratory tract infections and mortality rates, due to various respiratory diseases, meteorology, visibility, and ambient particulate levels.[58] SERC tabulated statistics from hospital admission records, death certificates, National Weather Service records, and Health Department ambient particulate data. They found positive correlations between reduced visibility at the county airport due to smoke, haze, or fog; hospital admissions for upper respiratory tract infections; and average monthly particulate levels at the county courthouse.[59] SERC also found unusually high mortality rates due to pneumonia compared with the national average, but did not examine those rates in relation to environmental parameters. Investigators did, however, leave open the possibility that the two were related, and recommended further study.[60]

While the SERC study was retrospective in design, the 1978-80 Montana Air Pollution Study (MAPS) examined immediate and on-going human health effects as they related to daily levels of air pollution in several Montana communities. Meteorological and air pollution data were collected in seven communities for modeling purposes, and emission inventories were compiled.

Lung function tests of children in four Montana cities were compared, after accounting for physical differences among the children, differences attributable to the community (e.g., elevation), and social differences (e.g., smoking in the home). The lung function testing, referred to as the Acute Effects study, was complemented by a second investigation, the Chronic Effects study. This part examined the effects on individuals with emphysema, asthma, bronchitis, and other chronic lung dysfunctions caused by high particulate episode days.

Based on these analyses, Missoula exhibited the highest levels of airborne particulate of all communities studied, and Missoula school children demonstrated reduced lung functions compared with children in Great Falls, (which had the cleanest air of all the cities tested).[61] Further, lung-impaired residents had breathing difficulties and were less active on days when particulate levels were high.[62]

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CHAPTER 3

METHODS

Sample and Data Collection

Receptor Site

All ambient samples were collected at the Missoula Health Department's Rose Lawn Park monitoring station. This site is about eight city blocks from Lion's Park, the receptor site used in the 1980 source apportionment study. The two sites are similarly situated between a residential area and a commercial "strip," in the approximate geographic center of the urban area.

Ambient Samplers

Coarse and fine particulate samples were collected for XRF analysis using a virtual impactor Sierra dichotomous sampler (dichot) which, by use of calibrated orifices, aerodynamically separates particulate matter into two size fractions: $\leq 2.5\mu m$ and $> 2.5 \leq 15\mu m$ in diameter. The dichot has a pre-set flow rate of 16.7 liters per minute. Particles are collected on Teflon membrane filters (37 mm in diameter), suitable for automated ß-attenuation mass determinations and XRF analysis.

Total suspended particulate (TSP) samples were collected for carbon analysis with General Metals high volume samplers (hi-vols). These devices have variable flow rates ranging from 40-50 cubic feet per minute, and collect TSP less than about 75μm in diameter on 8 X 10 inch glass fiber filters. Such filters are acceptable for organic analysis.

Sampling Schedule

Beginning November 1, 1982, through February 28, 1983, two hi-vols, operating alternately, collected daily TSP samples over a 24-hour, midnight-to-midnight period. The dichot operated every third day collecting 24-hour samples, beginning November 17, 1982, and ending February 27, 1983.

Sample Handling and Records

The Environmental Protection Agency (EPA), Research Triangle Park, North Carolina provided four twenty pair lots of dichot filters, individually wrapped in aluminum foil. Pairs were consecutively numbered and each filter was designated as either coarse or fine by a letter preceding the number (C and F, respectively). The collection surface of each coarse filter came coated with a thin film of oil to prevent loss of particles during handling and storage.

All filters were enclosed in plastic, ring-shaped holders to facilitate installation into the dichot sampler and to insure proper alignment in later β-attenuation measurements. The first pair of filters in each lot was set aside to be used as blanks in subsequent mass and XRF analyses. After sampling, filters were returned to their foil packaging, and prior to shipping, were stored in plastic bags in a freezer to reduce possible mass loss from volatilization and to inhibit artifact formation.

Detailed field records were maintained during the study period to insure proper filter tracking and to record field conditions. These records included instrument calibrations, flow meter readings, and site and meteorological observations.

Meteorological Measurements

Some meteorological measurements were made at the receptor site, while others were taken by the National Weather Service (NWS). Continuous temperature, wind speed, and wind direction data were mechanically recorded at the Lion's Park station during the 1980 study, and temperature and wind speed were collected at Rose Park during the 1982-83 study. In addition to these study-specific records, the NWS station at the county airport (approximately six miles northwest of Rose Park) routinely measures temperature, wind speed, precipitation (water equivalent), and accumulated precipitation (snow) daily.

Other Data

Field operators observed and recorded road/soil conditions near the receptor site during dichot sampling periods. Road/soil conditions for each sampling day were also determined (predicted) by a computerized combination of daily meteorological factors. (The specific technique is fully described below.) Weather factors and road/soil conditions were then used to determine classification of each sampling day into meteorological regimes to facilitate comparison of the findings from the two separate studies. This analysis is detailed in its own section below.

Analytical Procedures

Mass Determinations

TSP mass was gravimetrically determined at the Missoula Health Department laboratory. Prior to use, the glass hivol filters were stored in a desiccator (desiccant: Drierite or $CaSO_4$) for 24 hours, then weighed to the nearest 0.1 mg on a Torbal AE analytical balance. After sampling, filters (and their TSP load) were again desiccated for 24 hours and reweighed. Mass was then determined by the following equation:

$$M = \frac{T - t}{f * m * c * k} * 10^6$$

Where:

M = mass concentration of the TSP ($\mu g/m^3$)
T = total weight of the filter and TSP (grams)
t = tare weight of the filter (grams)
f = calibrated air flow of hi-vol (ft^3/min)
m = total number of minutes sampled (24 hours ± 2)
c = conversion factor (.0283) for $feet^3$ to $meters^3$
k = adjustment for ambient temp to standard temp

Dichotomous sample masses were determined by β-attenuation (as described in Chapter 2) at EPA's laboratory, Research Triangle Park, North Carolina.

Chemical Analyses

After sample mass measurements were completed, EPA sent the dichot filters to the Lawrence Berkeley Laboratory in California for XRF analysis (as described in Chapter 2). EPA forwarded the TSP samples to the Oregon Graduate Center for FVI carbon analysis (also described in Chapter 2). EPA reviewed all laboratory results for quality assurance purposes, then sent the results to the Missoula Health Department for CMB analysis.

Chemical Mass Balance (CMB)

The CMB Model

The CMB analysis was conducted at the Montana Department of Health and Environmental Sciences' Air Quality Bureau, using an IBM-XT computer and CMB software developed and copyrighted by NEA Laboratories, Inc., Beaverton, OR.

The model is based on the assumption that if there are p sources contributing pollutants to the atmosphere and there is no change in the relative concentrations of the pollutant species between the source and the receptor site, the total mass concentration of particulate matter, C, measured at the receptor site will be the linear sum of the contributions of the individual sources, S_j: or,

$$C = \sum_{j=1}^{p} S_j$$

The mass concentration C_i of aerosol property i, will be

$$C_i = \sum_{j=1}^{p} a_{ij} S_j$$

where a_{ij} is the mass fraction of source contribution j possessing property i at the receptor.[1,2,3]

If C_i and a_{ij} are known for all sources p, and $p < n$ (where n is the number of chemical species), a set of simultaneous equations exists which, when solved, will reveal S_j, the contribution from each potential source. S_j is determined by solving the equations using a least squares method, the statistical procedure employed in NEA's CMB software package.

The CMB Procedure

There are four operational steps in the CMB procedure: (1) creating data input files, (2) creating an airahed-source library, (3) interactive CMB calculations, and (4) tabulation of results. Each phase is described briefly below.

Using various micro computer software to calculate and edit the data, input files containing all pertinent mass, XRF, carbon, and sampling information were compiled into the formats appropriate for use with the NEA CMB software. These files were then merged into binary computer language by the NEA software, and were then ready for analysis. Appendix A contains a list and description of these files.

Due to funding limitations, no Missoula-specific source samples were taken for this study. Instead, library fingerprints supplied with the software package were used as representations of Missoula sources. The same approach was used in the 1980 study (except that in 1980, a sample of Missoula road sanding material was analyzed to develop an "urban dust" source fingerprint). In an effort to make the results as comparable as possible, several of the same "point" and "area" pollutant sources were again utilized in this study. Further, several sources not included in the earlier study but recommended for use in subsequent works, were considered here.[4] Point sources included a hog fuel boiler, particle board dryer, kraft recovery boiler, veneer dryer, sawmill cyclone, and lime kiln. Area sources used were road dust, vehicle exhaust, distillate oil, and residential wood smoke. All these sources have been employed in source apportionment studies in Oregon, and six were developed in a community very similar to Missoula.[5*]

The interactive CMB procedure involves the operator and the computer working together to derive a "best fit" combination of ambient air data with potential pollution contributors. This is accomplished by adding and subtracting both sources and fitting elements in an attempt to arrive at a least squares fit which meets the following criteria:

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[*] These source fingerprints were developed in Medford Oregon. They included wood combustion, road dust, transportation, veneer dryer, particle board dryer, and the hog fuel boiler.

(1) it minimizes chi square; (2) it maximizes the degrees of freedom; (3) it creates a model which produces an acceptable "calculated-to-measured" mass ratio (as close to 1 as possible within a given uncertainty); and, (4) it explains as much of the measured mass as possible. Using this process, several CMB trials were run on the chemical analysis data for both fine and coarse particulates, both including and excluding the carbon results.

Tabulation of best fit results into tables and histograms constitutes the final phase of the CMB procedure. This was accomplished using the capabilities of the NEA software package. The Results chapter summarizes findings, and they are included in their entirety in Appendix B.

Meteorological Regime Categorization

Meteorological regime categorization (MRC) is a process for adjusting data to eliminate potential biases due to differences in weather during a short-term sampling period. This may be accomplished several ways, the most simple of which is to group days into regimes according to the surface weather conditions which occurred each day during sampling. This study used a fairly simple set of criteria to assign days into seven different regimes.

Selection of Meteorological Parameters

Two criteria were used to determine which meteorological factors would be employed in the creation of weather regimes: effect on airborne particulate levels, and availability of data. Weather factors which may directly influence both the kinds and levels of particulates at a receptor site include surface and upper level wind speed and direction, precipitation, temperature, relative humidity, dew point, cloud cover, and inversion height and strength. However, due to a lack of data, meteorological information used in this study was limited to wind speed, precipitation (in terms of moisture content), accumulated precipitation (snow on the ground), and temperature.

Wind speed and precipitation have obvious effects on particulate levels; when either increases, pollution tends to decrease. Effects due to snow accumulation and temperature are more indirect, but need to be considered because they have at least the following influences: (1) snow cover tends to prevent particulate matter stemming from road dust or bare soil from becoming airborne; (2) snow cover reflects solar radiation and may prolong inversions by reducing atmospheric warming; (3) measurable snow fall engenders distribution of road sanding material which may later become airborne; and (4) cycles of freezing and thawing lead to wet or dry roads, which retard or facilitate, respectively, re-entrainment of sanding materials. Therefore, the above factors were used in various combinations to derive a set of regimes to differentiate conditions which might cause increased or decreased particulate levels.

In meteorological regime studies of air pollution data it is preferable to use meteorological information collected at the receptor or monitoring site. This is especially true in studies of air pollution over complex terrain such as the Missoula valley airshed. As previously stated, however, only wind and temperature data were collected at the receptor sites during the two study winters, while complete and continuous meteorological measurements were made by the NWS about six miles from the site. Consequently, in constructing meteorological regimes for the two study winters it was necessary to choose among several options: (1) whether to use the data more accurately reflecting conditions at the monitoring site; (2) use the more complete information from the NWS station; or (3) use a combination of the two. To assist in making the decision, I used two statistical procedures to compare the data sets: (1) T-tests revealed whether the meteorological data from the two sites differed significantly; and (2) a simple linear regression analysis calculated correlation coefficients for the two data sets. Tables 6 and 7 show the results of these procedures. Based on these analyses showing a strong positive correlation between the wind speeds at the two sites, and no statistically significant difference in the average daily temperature, I chose to forgo the opportunity to use receptor-site-specific information in favor of more complete data from the NWS.

TABLE 6

Results of Linear Regression: Temperature and Wind Speed at Johnson Bell Field and Rose Lawn Park

	Regression Equation	R^2
Average Daily Temperature:	$y = 1.0849x - 2.7566$	0.9654
	where: y = avg temp at JBF x = avg temp at RLP	
Average Daily Wind Speed:	$y = 2.1338x + 1.2833$	0.6339
	where: y = avg wind speed at JBF x = avg wind speed at RLP	

TABLE 7

Comparison of Average Daily Temperatures and Average Daily Wind Speeds Monitored at Rose Lawn Park and Johnson Bell Airport (NWS)

Met Site	N	Mean	σ	Std. Error	T Value	Degrees Freedom	2-Tail Prob
Average Daily Wind Speed (mph)							
NWS	120	4.8	2.4	0.2	19.4	119	0.0[a]
Rose Park	120	1.65	0.9	0.08			
Average Daily Temperature (°F)							
NWS	120	28.9	9.9	0.9	-0.13	119	0.899[b]
Rose Park	120	29.2	9.5	0.867			

[a] Null Hypothesis: the means of the two data sets are equal; For wind speeds, the probability that the means are equal is 0.0

[b] With the same hypothesis, for average daily temperatures, the probability of the means being equal is 89.9%

Creation of Meteorological Regimes

Ideally, meteorological regimes could be developed using every weather parameter known to affect particulate levels. However, this would necessitate the collection of extensive weather data appropriate to the receptor site, and such was not the case with this study. Instead, available data were used to construct seven regimes based on prevailing weather patterns and the road conditions which such weather might cause.

In order to delineate which meteorological factors, or combination of factors led to low, moderate, high, or very high levels of particulates during the last seven winters (1978-79 through 1984-85) in Missoula, daily TSP values were grouped into four categories and meteorological averages calculated for each group. The categories were constructed using the following TSP values (micrograms per cubic meter): (1) less than or equal to 75; (2) greater than 75 and less than or equal to 100; (3) greater than 100 and less than or equal to 150; and (4) greater than 150.** Based on these groups, the SPSS^{x} procedure "Breakdown" was used to calculate average daily values for temperature, precipitation, and wind speed. The results of this analysis are shown in

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** These categorizations are more or less arbitrary, except that they correspond to the air quality conditions labeled "good," "fair," "poor," and "alert" by the Missoula Health Department when describing the relative "quality" of the air in public announcements.

Table 8. Approximations of some of these average values became the meteorological "trigger values" shown in Table 9.

TABLE 8

Meteorological Averages Over Seven Winters in Four Subjective Air Quality Categories

	GOOD: $\le 75\ \mu g/m^3$		FAIR: > 75 and $\le 100\ \mu g/m^3$	
	Mean	σ	Mean	σ
Temperature:	30.4	11.8	27.5	12.2
Wind Speed:	6.98	3.2	4.9	1.97
Precipitation:	0.069	0.092	0.046	0.072

	POOR: > 100 and $\le 150\ \mu g/m^3$		ALERT: $> 150\ \mu g/m^3$	
	Mean	σ	Mean	σ
Temperature:	23.7	14	25.3	12.4
Wind Speed:	3.8	2.1	3.1	1.4
Precipitation:	0.021	0.041	0.007	0.022

TABLE 9

Meteorological Regimes

Regime Number	24-Hour Avg. H_2O Equivalent Precipitation		24-Hour Average Wind Speed[a]	Road/Soil Condition
1	$\le$ .01 in.		$\le$ 3 mph	Wet
2	$\le$ .01 in.		$\ge$ 3 $\le$ 7 mph	Wet
3	$\le$ .01 in.		$\le$ 3 mph	Dry
4	$\le$ .01 in.		$\ge$ 3 $\le$ 7 mph	Dry
5	$>$ .01 $\le$ .1 in.		$\le$ 3 mph	Wet
6	$>$ .01 $\le$ .1 in.		$\ge$ 3 $\le$ 7 mph	Wet
7	$>$ .1 in.	or	$>$ 7 mph	Variable

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[a] Based on one minute observations each hour.

After regime definition, days were computer sorted into regimes based on the occurrence of the various weather factors. Wet and dry regimes (including road/soil conditions) were assessed by computerized combination and by consideration of the average daily temperature, total accumulated precipitation, and H_2O equivalent precipitation. Computer results were verified by comparison with study field logs. Each day was judged individually, but also in relation to the two days before it. Specifically, the wet/dry differentiation was as follows.

1) If there had been more than 0.01 inches of precipitation within the last three days, a day was wet.

2) If the average daily temperature was > 35°F and there were ≥ 3 inches of accumulated snow, a day was wet.

3) If average daily temperature was ≥ 33°F, the accumulated precipitation was < 1 inch, and the day was not already wet, a day was dry.

4) If the daily temperature was ≤ 32°F, the temperature the previous day was ≥ 33°F, snow cover was < 3 inches, and the day was not already wet, a day was dry.

5) If the previous day was wet and nothing occurred to make a day dry, it too was wet.

6) If the previous day was dry and nothing occurred to make a day wet, it too was dry.

Twenty-two sample days were classified as wet or dry using this method; comparison with field logs revealed only 4 days which did not agree with the computer classification.

Based on the regime constructs defined in Table 9, days from seven winters (1978-79 through 1984-85) were computer categorized, and mean and standard deviations of TSP levels calculated. Statistically significant differences between regimes based on testing this categorization using oneway analysis of variance generally verified this construct.

Application of the Meteorological Regimes

Meteorological regime categorization (MRC) was applied to CMB results in two distinct ways to "neutralize" potential biases caused by weather differences during the two study winters. First, the two study winters were compared by calculating a weighted seasonal average based on the frequency of occurrence of days in each regime; and second, results of the two source apportionment studies were compared one regime at a time. The weighted average and its standard deviation were calculated by the following formulas:

$$\log TSP_c = \sum_{i=1}^{m} \frac{N_i}{N} \log TSP_i$$

and,

$$\log \sigma TSP_c = \sum_{i=1}^{m} \frac{N_i}{N} \log \sigma TSP_i$$

where N_i = the total number of daily occurrences of regime i (of m total) during the experimental period

N = the total number of days in the experimental period

m = the number of regime categories

$\log TSP_c$ = the categorized geometric mean

$\log TSP_i$ = the geometric mean of TSP in regime i

$\log \sigma TSP_c$ = the geometric standard deviation of the categorized mean

$\log \sigma TSP_i$ = the geometric standard deviation of TSP in regime i

NOTE: Remove log function to compute arithmetic statistics

References Cited In Chapter 3

1. Watson, John G., Jr., 1979, "Chemical Element Balance Receptor Model Methodology for Assessing the Sources of Fine and Total Suspended Particulate Matter in Portland Oregon"

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3. Stevens, Robert K. 1983, "Analytical Measurements for Use in Source Apportionment Studies to Determine Impact of Wood Burning on Visibility and Fine Particle Mass"

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CHAPTER 4

RESULTS AND DISCUSSION

This chapter reports the results of the various analyses performed during the course of this project. In an attempt to present a complete discussion of these findings, it is necessary to approach the report of results in much the same way as the analyses progressed. To that end, this chapter presents the results by comparing the two study winters at each level of the analytical process.

Comparison Through Particle Mass Measurements

TSP Mass versus Coarse + Fine Particle Mass

Due to differences in the particulate sampling equipment used in the study, observations and conclusions made throughout this chapter necessarily refer to particulate data from both hi-vol and dichot samplers. In order to compare and contrast results from these two different devices, it is necessary to understand the relationship between the TSP as collected by high volume sampler, and TSP_{cf} (the sum of the coarse and fine fraction masses) as collected by the dichotomous sampler. I used a simple linear regression of data from both study winters to examine this relationship.

Initially, using all the valid sample pairs (7) from 1979-80 in the regression analysis, the result was a rather poor R^2 of 0.1208 and a regression equation of coarse + fine = 0.2066 * TSP + 73.6. By eliminating a single outlier, the

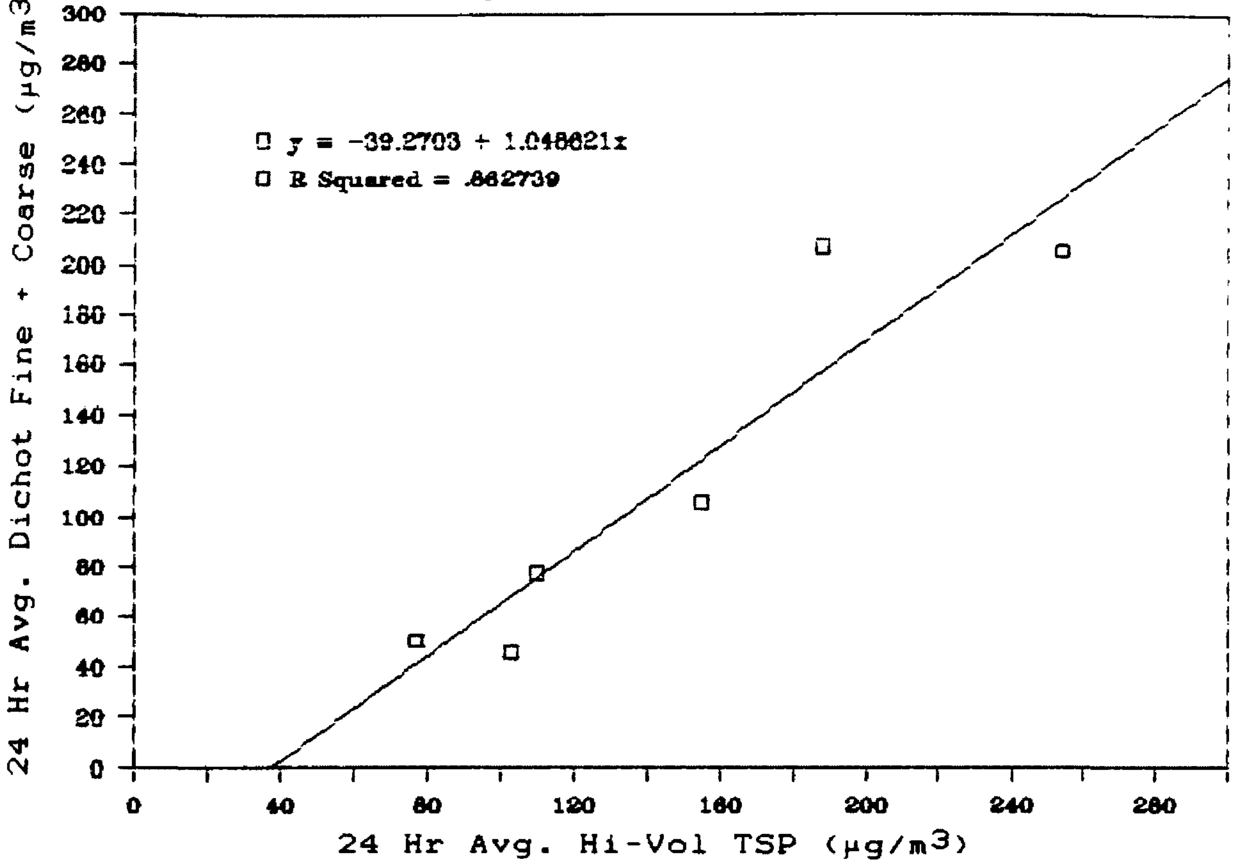
R^2 increased to a respectable 0.8627 and the regression equation became coarse + fine = 1.0486 * TSP - 39.27. The results of this analysis are graphically displayed in Figure 2A.

Results of a second linear regression using 1982-83 hi-vol TSP and the dichot TSP_{cf} data are graphed in Figure 2B. This analysis used 34 valid data sets and resulted in an R^2 of 0.930 and a regression equation of coarse + fine = 0.5534 * TSP + 0.7588. Of the two groups of "TSP" data points, the group from the 1982-83 study correlated more strongly than the group from the first study, and there were no outliers.

As can be seen in Figures 2A and 2B, the TSP_{cf} measured by the dichotomous sampler is always somewhat less than the TSP measured by the high volume sampler. This is to be expected for several reasons. First, the dichot collects particulate through a small orifice which prevents coarse fraction particles over about 15 µm in diameter from entering the sampler. This would contribute to the discrepancy in the two measurements of TSP, especially during periods when particulates consist primarily of large, higher mass particles, e.g., road dust. Second, the dichot operates at a lower flow rate than the hi-vol, which may favor the collection of particles in the smaller size and mass ranges since larger particles are not as easily deflected into the sampler at the slower flow rate. Finally, some of the difference may also be attributable to instrument error within

TSP vs Fine + Coarse Particle Mass

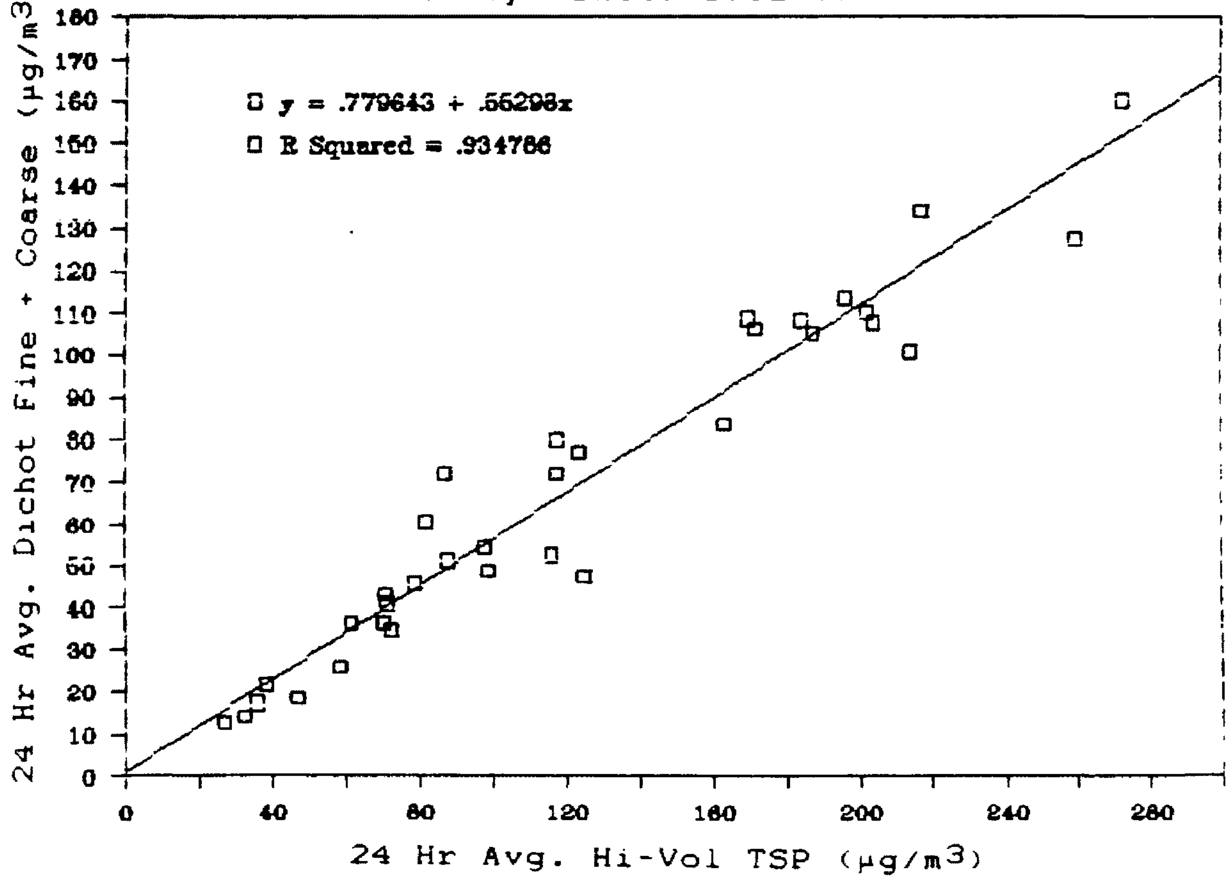
Study Winter 1979-80



FIGURES 2A & 2B

TSP vs Fine + Coarse Particle Mass

Study Winter 1982-83



operation specifications. However, in spite of these expected differences, the strong and consistent positive correlation between data from the two air monitoring devices demonstrates that analytical results based on measurements from one instrument can be reasonably assumed to apply proportionally to samples from the other.

Total Suspended Particulate During Both Study Winters

Tables 10 and 11 list the daily TSP mass measurements from the study winters 1979-80 and 1982-83, respectively. In both tables, dates marked by asterisks are days when the dichotomous sampler collected coarse and fine particulate samples for XRF and subsequent CMB analysis. Particulate mass was not determined for samples which field operators declared invalid due to technical problems (e.g., calibration errors) with the sampling equipment.

Comparing the two winters by using 150 micrograms per cubic meter ($\mu g/m^3$) as an indicator of "high particulate episodes," winter 1979-80 had a total of 47 episodes, while winter 1982-83 had 36. On a monthly basis, November 1979 had 12 episodes, November 1982 had 5; December 1979 had 11 episodes while December 1982 had 6; January 1980 experienced 9 episodes while January 1983 exceeded that with 18; and finally, February 1980 had 15 episodes while February 1983 had only 7.

Comparing the severity of air pollution episodes, winter 1979-80's highest 24 hour particulate average was 502.6

TABLE 10

Hi-Vol TSP Mass Measurements
Winter 1979-80 -- Lion's Park Site

NOVEMBER		DECEMBER		JANUARY		FEBRUARY	
Sample Date	Mass µg/m^3	Sample Date	Mass µg/m^3	Sample Date	Mass µg/m^3	Sample Date	Mass µg/m^3
1	106.0	1	162.2	1	174.6	1	349.3
2	167.6	2	60.3	2	122.6	2	397.0
3	127.8	3	124.0	3	112.6	3	246.7
4	41.0	4	30.2	4	110.3	4	82.1
5	50.2	5	111.1	5	58.4	5	134.7
6	75.8	6	162.2	6	29.2	6	270.4
7	75.3	7	265.4	7	81.9	7*	64.6
8	90.1	8	179.9	8	38.8	8	144.1
9	155.5	9	198.8	9	42.9	9	133.4
10	60.6	10	65.9	10	63.7	10	132.6
11	98.0	11	Inval	11	94.8	11	141.2
12	Inval	12	60.2	12	35.6	12	107.1
13	30.8	13	129.8	13	71.5	13	48.3
14	161.6	14	117.3	14	44.6	14	47.3
15	195.5	15	26.0	15	106.2	15	32.2
16	261.6	16	46.6	16	244.5	16	78.7
17	168.7	17	123.4	17	132.3	17	133.8
18	97.7	18	Inval	18	144.1	18*	155.0
19	100.7	19	183.5	19	281.5	19*	109.9
20	132.4	20	213.7	20	210.8	20*	187.9
21	169.2	21	154.9	21	192.0	21*	151.0
22	187.2	22	120.6	22	Inval	22*	416.5
23	165.6	23	53.9	23	320.7	23	254.6
24	71.7	24	135.7	24	143.3	24	260.1
25	80.8	25	107.1	25	84.8	25	410.2
26	71.3	26	134.9	26	54.2	26*	407.4
27	69.7	27	108.6	27	74.6	27*	502.6
28	167.1	28	205.8	28	136.4	28*	254.2
29	266.5	29	181.7	29	301.5	29*	193.4
30	178.4	30	140.8	30	273.9		
		31	173.1	31	260.0	March	
						1	300.5
						2	195.3
						3	155.8
						4	41.7
						5	38.5
						6*	77.0
						7*	102.9

---

* = Day sampled with dichot for XRF and CMB analysis
Inval = Invalid sample

TABLE 11

Hi-Vol TSP Mass Measurements
Winter 1982-83 -- Rose Park Site

NOVEMBER		DECEMBER		JANUARY		FEBRUARY	
Sample Date	Mass $\mu g/m^3$	Sample Date	Mass $\mu g/m^3$	Sample Date	Mass $\mu g/m^3$	Sample Date	Mass $\mu g/m^3$
1	54.0	1	72.5	1**	187.1	1	155.8
2	90.4	2**	96.8	2	156.2	2	107.7
3	90.1	3	37.9	3	195.9	3*	116.1
4	125.9	4	84.9	4*	117.1	4	158.4
5	57.7	5**	117.4	5	54.2	5	133.7
6	34.2	6	35.5	6	100.3	6**	123.5
7	48.6	7	40.2	7*	Inval	7	101.1
8	84.1	8**	87.7	8	60.8	8	90.7
9	71.3	9	71.2	9	143.2	9**	97.6
10	89.9	10	192.9	10*	32.2	10	74.6
11	58.1	11**	162.9	11	279.5	11	57.5
12	52.1	12	181.8	12	422.1	12*	46.8
13	76.0	13	168.2	13*	272.4	13	111.5
14	109.7	14**	169.3	14	346.4	14	143.2
15	159.8	15	50.9	15	236.2	15*	203.6
16	192.0	16	51.7	16*	259.3	16	122.7
17*	81.7	17*	26.6	17	239.1	17	36.0
18	51.6	18	80.8	18	153.5	18*	35.9
19	61.9	19	76.3	19*	214.0	19	111.1
20**	58.0	20**	71.4	20	155.2	20	193.2
21	37.4	21	60.7	21	264.4	21**	183.9
22	53.5	22	67.4	22**	86.6	22	162.8
23**	70.4	23**	38.4	23	Inval	23	147.3
24	123.9	24	54.1	24	87.0	24**	195.8
25	114.4	25	88.0	25*	171.6	25	93.6
26**	201.9	26**	71.0	26	Inval	26	77.7
27	173.9	27	90.1	27	122.4	27**	72.3
28	156.8	28	114.4	28*	124.8	28	74.3
29*	61.5	29**	78.8	29	210.5		
30	Inval	30	101.8	30	215.0		
		31	184.2	31**	217.1		

* = Days sampled with dichot for XRF analysis
** = Days with valid CMB fits
Inval = Invalid sample

$\mu g/m^3$; 1982-83's was 422.1 $\mu g/m^3$. In terms of the duration of episodes with 24-hour TSP levels above 150 $\mu g/m^3$, both winters had one extended event: the 1979-80 episode lasted 12 consecutive days (from February 20 through March 2), while the 1982-83 incident extended 11 consecutive days (January 11 through January 21). Based on these simple comparisons, it appears that Missoula experienced worse particulate pollution during the first study winter.

Table 12 contains a listing of the monthly particulate averages (as measured by hi-vol samplers) for both study winters, along with a subcategory of the means of those 24 hour periods for which valid samples were successfully analyzed using the CMB procedure. In this rudimentary fashion, the samples used for CMB analysis can be compared with the interval of the winter they are supposed to represent. For example, as shown in Table 12, during the first three months of winter 1979-80 the monthly TSP averages were about the same, although the large standard deviations indicate considerable variation on a daily basis. However, the monthly TSP average for February 1980, was considerably higher than the averages during the first three months of that winter. Further, the TSP average for the ten days during the month of February when samples were analyzed by CMB was even higher. From this compilation it is clear that the month during which CMB samples were collected had the highest particulate

TABLE 12

Comparison of Monthly TSP Averages for the Two Study Winters

DATE	PARK SITE	ARITHMETIC AVERAGE ($\mu g/m^3$)	STD. DEV. ($\mu g/m^3$)	GEOMETRIC AVERAGE ($\mu g/m^3$)	STD. DEV. ($\mu g/m^3$)
11/79	Lion's	125.0	61.7	109.8	1.7
12/79	Lion's	130.0	59.0	113.9	1.8
1/80	Lion's	134.7	87.6	108.3	2.0
2/80	Lion's	201.6	129.6	161.5	2.0
	CMB[a]	228.1	134.8	195.2	1.8
	Winter 79-80	136.6	72.3	118.0	1.8
	79-80 CMB[b]	206.0	132.5	173.2	1.8
11/82	Rose[c]	91.1	46.9	81.1	1.6
	CMB[d,e]	110.1	79.7	93.8	2.0
12/82	Rose	91.2	48.1	79.9	1.7
	CMB[f]	99.3	43.5	90.9	1.6
1/83	Rose	183.0	90.0	159.0	1.8
	CMB[g]	163.6	68.4	152.1	1.6
2/83	Rose	115.3	47.9	104.5	1.6
	CMB[h]	134.6	53.7	125.7	1.5
	Winter 82-83	118.1	70.5	100.7	1.8
	CMB[i]	119.4	56.0	107.0	1.6

[a] Sample days = 10

[b] Sample days = 12

[c] Rose = monthly TSP statistics

[d] CMB = TSP statistics for all days used in CMB analyses

[e] Sample days = 3

[f] Sample days = 9

[g] Sample days = 3

[h] Sample days = 5

[i] Sample days = 20

levels of the entire winter. Thus it is quite likely that February in general, and the days sampled for CMB analysis in particular, may not have been representative of the winter as a whole.

In contrast to winter 1979-80, winter 1982-83 had more variation in month-to-month particulate levels and in the occurrence of high particulate days. For example, only November and December of 1982 had similar monthly means, and both were considerably lower than the averages of the same months in 1979. Further, January 1983 had the highest TSP average of the winter, while February's mean was lower than January's, yet higher than the means of either November or December.

Continuing the comparison with the first study (as shown in Table 12), except for January, the 1982-83 TSP averages for those days used in the CMB analysis were all noticeably higher than the mean during the month in which collection of samples took place. For instance in December, the arithmetic mean for the nine "CMB analysis days" (i.e., the largest sample set for the entire study) was 8.1 $\mu g/m^3$ (≈9%) higher than the TSP average for all of December. However, in January, the month with the highest TSP mean, only three samples were successfully apportioned by the CMB procedure, and the TSP mean for those three days was approximately 19 $\mu g/m^3$ (≈10%) lower than the TSP average for the whole month. From this compilation, it appears that the samples used for CMB analysis during the second study winter also may not be a fair representation of the entire study winter. The possible reasons for the difficulty in achieving CMB apportionment are discussed in detail later in this chapter (pp.71-72 and 78-79).

TSP_{cf} on CMB Analysis Days

In terms of the relative severity of particulate pollution during the actual source apportionment studies, comparing and contrasting TSP_{cf} is somewhat indicative of the ability of CMB sampling days to represent the period during which they were collected. This information for both study winters is shown in the next two tables. In the first instance, while it is clear from the previous evaluation of TSP concentrations that 1979-80 had many high particulate days, as shown in Table 13, only three out of seven February 1980 TSP_{cf} samples used for CMB analysis had mass loadings greater than $100 \mu g/m^3$. This could well mean that the results of the first Missoula source apportionment analysis do not necessarily represent either the "worst case" or the "usual case" during that winter.

Table 14 lists the 35 valid sample pairs of coarse and fine particle mass concentrations collected during the second study winter. In contrast to the first study, these data include eleven high TSP_{cf} episodes, with one each in November and December, six in January, and three in Febru-

TABLE 13

Fine and Coarse Particle Mass
Study Winter 1979-80

Sample Date	Fine Mass ($\mu g/m^3$)	Percent of Total	Coarse Mass ($\mu g/m^3$)	Percent of Total
2-07	29	--	--	--
2-18*	106	100	<1	0
2-19	68	87	10	13
2-20*	164	79	43	21
2-21	55	--	--	--
2-23	37	--	--	--
2-26	46	52	42	48
2-27	--	--	89	--
2-28*	87	42	119	58
2-29	22	--	--	--
3-06	44	88	6	12
3-07	41	89	5	11

* Days with TSP_{cf} greater than 100 $\mu g/m^3$

ary. These samples should therefore result in a more accurate representation of the nature of high particulate air pollution across the entire winter.

The above comparisons raise the following several questions regarding the interpretation of results. First, are the CMB apportionment results for either study winter truly representative of the entire winter in which they were collected? Second, since sample collection for the 1979-80 study occurred only during February and the first few days in March, can the CMB results be meaningfully compared to those of the 1982-83 study which contained sample days from mid-November through February? And finally, can the 1979-80 study results which stem from analyses of twelve samples be

TABLE 14

Fine and Coarse Particle Mass
Study Winter 1982-83

Sample Date	Fine Mass (µg/m^3)	Percent of Total	Coarse Mass (µg/m^3)	Percent of Total
11-17	47.2	78.2	13.2	21.8
11-20	15.5	59.9	10.6	40.6
11-23	25.6	70.5	10.7	29.5
11-26*	79.7	72.6	30.1	27.4
11-29	24.2	66.5	12.2	33.5
AVERAGE:	38.4	71.4	15.4	28.6
12-2	29.6	60.7	19.2	39.3
12-5	57.9	80.8	13.8	19.2
12-8	37.3	72.9	13.9	27.1
12-11	74.3	88.2	8.9	11.8
12-14*	96.7	89.4	11.5	10.7
12-17	7.8	61.9	4.8	38.1
12-20	37.7	92.0	3.3	8.0
12-23	19.3	89.8	2.2	10.2
12-26	38.9	89.6	4.5	10.4
12-29	40.0	87.3	5.8	12.7
AVERAGE:	44.0	83.3	8.8	16.7
1-1*	86.2	81.5	19.6	18.5
1-4	76.1	95.4	3.7	4.6
1-7	61.7	93.3	4.4	6.7
1-10	11.5	80.4	2.8	19.6
1-13*	47.0	29.4	112.9	70.6
1-16*	52.9	41.6	74.3	58.4
1-19*	51.7	51.6	48.6	48.4
1-22	50.6	70.4	21.3	29.6
1-25*	70.9	67.0	35.0	33.0
1-28	13.5	28.6	33.7	71.4
1-31*	69.8	52.3	63.8	47.7
AVERAGE:	53.8	58.5	32.8	41.5
2-3	21.4	40.1	31.2	59.3
2-6	46.1	60.1	30.6	34.9
2-9	52.5	96.5	1.9	3.5
2-12	15.2	83.5	3.0	16.5
2-15*	39.5	36.9	67.7	63.1
2-18	17.0	98.3	0.3	0.7
2-21*	62.6	58.1	45.1	41.2
2-24*	47.1	41.6	66.1	58.4
2-27	15.9	45.7	18.9	54.3
AVERAGE:	32.3	52.4	29.4	47.6

* = Days with $TSP_{cf} \geq 100$ µg/m^3

effectively compared with results of the 1982-83 study which are based on twenty samples? The issues presented by these questions are addressed in the last section of this chapter.

Coarse versus Fine Particle Mass Distribution: Winter Trends

An important aspect of Missoula's wintertime pollution situation is the variation in the size and chemical nature of the particles during the course of the pollution season. Unfortunately, assessing the chemistry of wintertime particulates on a regular basis is beyond the financial means of the local control agency, which leaves the main means of assessing potential effects to analysis of gross particulate mass and size fractionation. Although this is not a complete means of analysis, it is an important aspect of assessing potential health and other environmental effects, and in developing control strategies.

One way to explore this aspect of the problem is to examine the changing size constituencies of the "total" suspended particulates in an attempt to understand sources of the various fractions. This too is revealed in the two immediately previous tables. Table 13 (page 64) displays the coarse and fine particle mass concentrations as measured with the dichot sampler during the 1979-80 study winter. Due to sample collection problems, determination of fine particle mass was limited to eleven samples out of the twelve collected, while coarse particle mass was calculable for only eight samples out of twelve. These errors resulted in collection of seven valid sample pairs out of a possible twelve, although unpaired coarse and fine samples were chemically analyzed and subjected to CMB analysis.

Considering only the seven valid sample pairs, an average of 71.2 percent of the TSP_{cf} was comprised of fine fraction particulates, leaving an average of 28.8 percent in the coarse fraction. However, the fine fraction varied from 100 percent on 18 February to 42 percent on 28 February.

In terms of size fractionation of particulates collected during the 1982-83 study (Table 14, page 65), in November 1982, an average of 71.4 percent of the TSP_{cf} mass occurred in the fine fraction. By December, the fine fraction accounted for 83.3 percent of the TSP_{cf}. During January and February, a major shift occurred in particulate composition, with the fine fraction making up just over half of the TSP_{cf} (58.5% and 52.4% respectively).

The dichot data for winter 1982-83 is dissimilar to the first study except with respect to the wide fluctuation in the fine fraction contribution to the TSP_{cf}. In November 1982, the fine fraction contributed a range of 59.9 to 78.2 percent; in December, the range extended from 60.7 to 92.0 percent; in January, the fine fraction contributed from 28.6 to 95.4 percent; and in February, the fine fraction varied from 36.9 to 98.3 percent of the TSP_{cf}. In order to assess possible control strategies, the reasons for these fluctuations in particulate size composition must be thoroughly considered.

The two factors with the greatest effect on the fine fraction composition of the TSP_{cf} are source emissions and weather. On a day-to-day or sample-to-sample basis, weather affects not only particulate concentrations, but also particle size proportions. Therefore, considering the winter as a whole, it is possible to postulate probable effects on the sources and nature of air pollution stemming from fluctuations in certain weather parameters. For example, the following scenario might explain the trends in the TSP_{cf} seen during the source apportionment study of 1982-83.

*) In November, temperatures drop low enough to foster residential wood burning for home heating, which generates mostly fine fraction particulates. At the same time, there is no measurable accumulation of snow, and little or no sanding material is spread on the roads. However, the streets dry out periodically, and small amounts of residual road dust becomes airborne as mostly coarse fraction particulate.

*) In December, temperatures become colder, and frequent precipitation in the form of snow causes accumulation which requires street sanding. Because the roads are either wet or snow-covered, they contribute little to airborne particulates, and consequently, most of the TSP remains in the fine fraction.

*) In early to mid-January, the valley experiences an unseasonable thaw which melts nearly all of the snow from the roads. With continuing moderate temperatures and little or no additional precipitation, while temperatures remain low enough to preclude street cleaning, roads dry and become dusty, contributing large amounts of coarse fraction particulates. Wood burning continues.

*) Through February, except when roads are wet from light snowfall and/or rain, much of the accumulated sanding material continues to be re-entrained by wind or vehicular traffic. In addition, as the sanding material is pulverized by traffic, resulting airborne particles

> become smaller in diameter. As a consequence, sanding materials comprise a progressively greater proportion of the fine fraction particulates. At the same time, cool temperatures at night result in the continuation of some residential wood burning.

The above scenario illustrates how weather patterns could affect particulate source contributions, and might explain how the monthly percentage of the fine fraction of the TSP_{cf} can fluctuate from 71 to 83 to 59 to 52 percent, during the course of a winter. While such a construct is speculative, it is also useful in beginning the assessment of the pollution situation and in developing potential control strategies. However, without more complete analysis using tools such as CMB and meteorological regime categorization (MRC), only preliminary control programs are possible. In turn, to reach the levels of precise analysis possible with CMB and MRC, it is first necessary to address several more types of data. These follow below.

Chemical Analysis of Particulates

X-ray Fluorescence of the Coarse and Fine Fractions

Tables 1 and 2 in Appendix C display the elemental concentrations and the calculated uncertainties of the fine and coarse fraction particulates sampled during study winters 1979-80 and 1982-83 respectively. These tables reveal several important characteristics of the studies they represent. For example, a noticeable feature of Table 1C is the four days with no results for the coarse fraction. Since the entire study consisted of only twelve sample days and four are missing, only eight samples are left to represent an entire winter. Clearly, this may affect the accuracy and applicability of results, while creating special difficulties in interpreting and comparing the results with other studies.

A second aspect of the 1979-80 study disclosed in Table 1C is that most of the sample days fall into "groups" of clustered sample days (and a single, isolated sampling day) rather than the samples being evenly spaced throughout the test period. This schedule was necessary because limited funding restricted the number of samples which could be collected and analyzed. For that reason, project coordinators tried to collect samples only on days when TSP levels were projected to be high enough to ensure that a sufficient quantity of particulate matter would be obtained for analysis. Unfortunately, this type of sampling can lead to biased results. For example, high particulate episodes during the month of February are most likely due to excessive quantities of airborne dust, causing results which will be biased toward this source. In contrast, during winter months with sufficient snow cover, sampling only on days with high particulate levels could lead to a bias toward other sources, e.g., residential wood combustion or vehicle exhaust.

In contrast to the 1979-80 study, the 1982-83 study was conducted with thorough sampling over the course of the entire winter. This difference is reflected in Table 2C, which contains a complete set of XRF results for both particulate fractions for the entire study period. As is evident in that table, the sample days were evenly spaced, with one sample collected every three days from mid-November until the end of February. Unfortunately, for reasons which are discussed in the following section, not all samples were able to be apportioned by CMB.

Organic Analysis Results

Table 3 in Appendix C discloses a major problem with the 1982-83 data: organic component results were derived from TSP samples collected by hi-volume sampler, instead of from analyses of the coarse and fine dichot samples as in the 1980 study. While the choice to use the TSP samples for organic analyses was based on economic constraints, it was unclear at the time that decision was made this option would result in the following negative consequences: (1) the organic analysis results could not be used in the CMB analysis*; (2) the lack of organic concentration data would cause errors in CMB results; and (3) because the raw data bases

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* The choice to use organic results from hi-vol filters led to carbonaceous concentrations expressed in micrograms per cm^2, while compositional data from the much smaller dichot filters were expressed in nanograms per m^3. With no means to adjust for the discrepancies in either filter size or instrument flow rates, this difference effectively precluded use of these data in the NEA CMB program.

differ, comparing CMB results for the two study winters would be much more difficult.

When the nature of the problem became apparent, I performed several kinds of data manipulations in an effort to make the organic analysis results useful in the CMB analysis. However, no method improved the data fits, and in most cases fewer data fits resulted. These manipulations included the following: (1) using only a portion of the organic concentration, i.e., an amount based on the ratio of the dichot airflow rate to the hi-vol airflow rate and respective filter sizes; (2) subtracting out of each sample an average "background" amount, based on the results of the carbon analyses conducted on three summertime filters; (3) using the same organic results for data fits for both coarse and fine CMB trials; and finally, (4) using no carbon data at all in any of the data fits. This final option allowed the largest number of acceptable fits in the fine fraction (20 out of 34), but no fits were found in any coarse fraction CMB trials.

CMB Results From Both Study Winters

Coarse Fraction

Table 15 contains the results of the CMB analysis of the coarse fraction of the 1979-80 study. Examination of these data reveals several discrepancies. For example, the total mass for the sample of 2-18-80 was less than 1$\mu g/m^3$,

TABLE 15

Source Contributions in the Coarse Fraction of TSP Study Winter 1979-80[a]

Sample Date	Mass ($\mu g/m^3$)	Urban Dust ($\mu g/m^3$)	Urban Dust %	Res Wood Comb ($\mu g/m^3$)	Res Wood Comb %	Percent Explained
2-18	<1	2	--	3	--	--
2-19	10	22	220[b]	--	--	220[b]
2-20	43	4	10	--	--	10
2-26	42	44	104	3	6	110
2-27	89	60	67	6	7	74
2-28	119	55	46	--	--	46
3-06	6	2	34	4	57	91
3-07	5	3	66	2	34	100
AVERAGE:			54		17	72
STD DEV:			32		23	38
STD ERROR:			13		12	16

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[a] Derived from Cooper, 1980

[b] Not included in calculating the average percent becauae of uncertainty in the mass

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but CMB attributed 2 and 3$\mu g/m^3$ of coarse particulate matter to urban dust and residential wood combustion sources, respectively. Also, the total mass for the sample dated 2-19-80 equals 10$\mu g/m^3$, yet 22$\mu g/m^3$ were apportioned to urban dust sources, accounting for 220 percent of the mass. In spite of these problems with the data, CMB analysis was able to apportion ambient particulates among known sources, but only with high associated uncertainties. The coarse fraction was attributed to 54 ± 13 percent from urban dust sources (e.g., paved and unpaved roads, sanding materials, and unpaved alleys and parking lots), and 17 ± 11.5 percent from residential wood combustion. Combined, dust and combustion sources accounted for only 72 ± 15.5 percent of the coarse fraction, leaving the remainder unexplained. That is, no additional particle mass could be attributed to other sources through the CMB data fit process.

As stated above, in the 1982-83 study, no acceptable data fits could be found through CMB analysis of the XRF coarse fraction, neither including nor excluding organic composition data in the analytical process. Attempted data fits using many combinations of the most likely sources included in the NEA source fingerprint library failed to produce satisfactory results. In repeated trials, the particulate mass was either underexplained (always less than 50 percent) or overexplained (always greater than 150 percent).

Based on the 1980 study which utilized a Missoula dust source sample, it is reasonable to conclude that the 1982-83 analysis failed to produce adequate apportionment of the coarse fraction due to the lack of Missoula-specific source fingerprints. I attempted CMB fits using not only the 1980 fingerprint, but also several others available in the NEA source library. None substantially improved the CMB fits. It is likely that the 1980 dust fingerprint was unsuitable for use in apportioning 1982-83 dust due to a major change in the type of street-sanding material which occurred between 1980 and 1982.** After numerous attempts, all resulting in unacceptable data fits, I abandoned further analysis in this area.

Fine Fraction

Table 16 displays the 11 valid data sets for fine fraction particulates as apportioned in the 1979-80 CMB study. The percent contributions from the various sources cover a wide range: residential wood combustion varied from a low of 21 percent to a high of 112 percent; auto exhaust contributed from 4 to 14 percent; urban dust percentages ranged from 0.8 to 36 percent; and finally, the hog fuel boiler added from 0.7 to 3 percent. Overall, residential wood combustion was the primary source of fine particulates, contributing an average of 68.3 ± 8.0 percent of the mass. Urban dust, auto exhaust, and the hog fuel boiler source contributed respectively smaller percentages of the fine fraction mass. An average of 14.6 ± 9.2 percent of the fine particle mass came from a source or sources which could not be determined by the CMB analysis.

** During winters prior to 1982, the Montana Department of Highways maintained Missoula street connectors to state highways using a combination of sand and salt to improve vehicle traction. The City of Missoula assumed this responsibility in 1982, and used a different source of sanding material than the State. Further, the City stopped adding salt to the sanding material mixture. This change could cause a major difference in the particles that would become airborne as sanding materials break down and are blown by the wind or entrained by vehicle tires.

TABLE 16

TSP Source Contributions in the Fine Fraction Study Winter 1979-80[a]

Sample Date	Mass (µg/m³)	Resid Wood Comb (µg/m³)	%	Auto Exhaust (µg/m³)	%	Urban Dust (µg/m³)	%	Hog Fuel Boiler (µg/m³)	%	Percent Explained
2-07	29	21	72	3	10	1	3	0.8	3	88
2-18	106	48	45	4	4	0.8	0.8	0.7	0.7	51
2-19	68	56	82	3	4	3	4	1.2	2	92
2-20	164	34	21	9	5	3	2	3.4	2	30
2-21	56	34	61	3	5	1	2	0.4	0.7	69
2-23	37	23	62	3	8	10	27	1.1	3	100
2-26	46	38	83	2	4	6	13	0.5	1	101
2-27	—	47	—	4	—	9	—	1.4	—	--
2-28	87	37	43	3	3	5	6	1.1	1	53
2-29	22	15	68	3	14	8	36	0.4	2	120
3-06	44	45	102	3	7	1	2	1.3	3	114
3-07	41	46	112	2	5	1	2	0.9	2	121
AVERAGE:			68.3		6.3		8.9		1.9	85
STD DEVIATION			26.4		3.3		11.8		0.9	31
STD ERROR			8.0		1.0		3.6		0.3	9

[a] Derived from Cooper and DeCesar, 1980

Table 17 contains the 1982-83 CMB apportionment of the fine fraction particulate. While inorganic data were available for thirty-five samples collected from mid-November through February, the CMB yielded acceptable fits for only twenty samples: three in November, nine in December, three in January, and five in February. In contrast to the 1979-80 CMB results, 1982-83 particulate mass was distributed among three sources: residential wood combustion, auto exhaust, and urban dust. No acceptable data fits included a point source (e.g., a hog fuel boiler).

TABLE 17

TSP Source Contributions in the Fine Fraction Study Winter 1982-83

Sample Date	Mass (µg/m³)	Resid Wood Comb (µg/m³)	%	Auto Exhaust (µg/m³)	%	Urban Dust (µg/m³)	%	Percent Explained
11-20	16	12.5	78	0.6	4	0.9	6	88
11-23	26	25.8	99	0.6	2	0.9	3	104
11-26	80	56.5	71	1.8	2	1.9	2	75
12-02	30	28.4	95	0.9	3	1.4	5	103
12-05	58	52.6	91	1.3	2	1.2	2	95
12-08	37	32.3	87	0.7	2	0.6	2	91
12-11	75	78.4	106	1.6	2	0.0	0	108
12-14	97	67.1	69	2.1	2	0.3	(1	71
12-20	38	35.2	93	1.1	3	0.2	1	97
12-23	19	17.9	94	0.7	4	0.2	1	99
12-26	39	41.8	107	0.7	2	0.3	(1	109
12-29	40	28.7	72	0.8	2	0.5	1	75
01-01	86	71.0	83	1.9	2	1.4	2	87
01-22	51	51.0	100	1.9	4	1.6	3	107
01-31	70	56.4	81	2.2	3	6.5	9	93
02-06	46	48.0	104	1.0	2	2.6	6	112
02-09	52	54.6	105	2.1	4	0.0	0	109
02-21	63	54.9	87	2.2	3	3.9	6	96
02-24	47	43.4	92	2.1	4	5.1	11	107
02-27	16	15.8	99	0.8	5	1.8	11	115
AVERAGE:			90.7		2.9		3.6	97
STD DEVIATION:			11.9		1.0		3.6	13
STD ERROR:			2.7		0.2		0.8	3

As in the 1979-80 analysis, residential wood combustion was the primary source of the fine fraction particle mass with percent contributions ranging from 69 to 107 percent. However, the average contribution from RWC was 33% higher in 1982-83 than during 1979-80. Again similarly to 1979-80, the second largest contributor was urban dust, with contributions ranging from 0 to 11 percent, but averaging 60% lower than the contribution found in 1979-80. Finally, vehicle exhaust contributions varied from 2 to 5 percent, with an average contribution 54% lower than in 1979-80. An overall average of 97.1 ± 2.9 percent of the particle mass was explained, that result being somewhat higher than the 85.4 ± 9.2 percent accounted for in 1979-80. These differences in the two CMB analyses are addressed in greater detail later in the final sections of this chapter.

Problems With Interpretation

The problems with the input data mentioned above raise questions about the reliability and applicability of the CMB results. For example, no carbon data could be incorporated into the CMB input of receptor concentrations, but nearly all source fingerprints contained some carbonaceous component. The lack of organic carbon and/or elemental carbon as fitting elements could eliminate one or more sources from the fitting process, and distribute some or all of the respective contributions of those missing sources among other sources. This could inflate some of the calculated source contributions (especially in the case of residential wood combustion).

In addition to the lack of complete receptor site chemical data, lack of Missoula-specific source data also complicates interpretation of CMB results. As with the coarse fraction, fine fraction data fits were derived using only NEA library fingerprints. Thus, the fine fraction CMB results may be in error, especially as regards the urban dust contribution. Since urban dust provided no acceptable data fits in the coarse fraction, it is reasonable to assume that the urban dust contribution in the fine fraction is somewhat underestimated.

Finally, the sample size and distribution of samples for the two study winters varied to a considerable degree. In winter 1979-80, ten of the twelve total samples were collected in the month of February, and nine of the ten provided "valid" CMB results. In winter 1982-83, nine of the thirty-five total samples were collected in February, but acceptable data fits were found for only five of these samples. With these additional differences in the study winters, comparisons of the two study winters become even more complex.

Appendix B contains the tabular and histogram results for all 1982-83 samples for which acceptable data fits were found. Two important observations can be made from these tables and graphs. First, elements lead and potassium were the only two elements used in all twenty data fits. This compares with 10 elements used in all fits in 1979-80, and again points to the weaknesses caused by the lack of Missoula-specific source fingerprints. Second, "source lead" consistently matched levels of "ambient lead" (indicated in the histograms by an asterisk approaching or intersecting the bar graph of a particular sample). This consistently good fit indicates there was most likely only one source of lead particulate to the air, in this case, emissions from vehicles using leaded gasoline as fuel. However, as implied above, the acceptability of the data fits does not necessarily accurately reflect the reality of a source's contribution.

Comparison Through Meteorological Regime Categorization

As emphasized previously, weather directly affects particulate levels and also indirectly affects the size fraction and chemical constituents by influencing particulate pollution sources. This section more fully explores this relationship by classifying certain data in terms of previously defined meteorological regimes, first by examining the overall levels of TSP which occurred, and finally, by reviewing the results of both source apportionment studies.

Total Suspended Particulate

Table 18 facilitates a comparison of TSP during the two study winters through meteorological categorization (See

TABLE 18

Study Winters' TSP Averages within Meteorological Regimes

1979-80

	Total	Sample	GEOMETRIC ($\mu g/m^3$)			ARITHMETIC ($\mu g/m^3$)	
Regime	Days	Days[a]	Mean	Std Dev	90% C.I.[b]	Mean	Std Dev
1	40	37	172.5	1.7	172-173	192.8	89.3
2	23	23	131.2	1.6	131-132	144.8	62.2
3	7	7	251.7	1.7	250-254	280.5	135.4
4	6	6	136.3	1.9	131-141	167.0	128.6
5	4	4	130.3	1.2	130-130	132.4	28.7
6	19	19	109.4	1.6	109-110	122.5	61.1
7	29	28	59.4	1.7	59-60	68.4	39.0
TOTAL	128	124					
UNADJUSTED MEAN:			120.3	1.9	120-121	146.8	93.1
ADJUSTED MEAN:			120.6	1.7	120-121	147.3	71.3

1982-83

	Total	Sample	GEOMETRIC ($\mu g/m^3$)			ARITHMETIC ($\mu g/m^3$)	
Regime	Days	Days[a]	Mean	Std Dev	90% C.I.[b]	Mean	Std Dev
1	16	16	177.8	1.3	177-178	186.1	68.8
2	31	28	96.2	1.6	96-97	107.8	57.1
3	11	11	178.0	1.5	177-179	190.9	74.8
4	17	16	128.3	1.5	128-129	138.2	56.3
5	4	4	119.0	1.3	119-119	121.9	32.9
6	22	22	73.4	1.5	73-74	79.3	35.0
7	21	21	55.9	1.7	55-57	66.6	54.0
TOTAL:	122	118					
UNADJUSTED MEAN:			109.6	1.5	109-110	127.3	54.1
ADJUSTED MEAN:			100.2	1.5	100-100	118.0	54.8

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[a] Sample Days = Days with valid TSP samples

[b] 90% C.I. = 90% Confidence Interval about the calculated mean

Table 9, page 50 for regime definitions). It thereby allows examination of the two winters in terms of both the frequencies of various weather factors and the TSP averages in identical regimes. Through such a compilation using nearly complete TSP data collected during the two study winters, it is possible to develop a more complete understanding of how predominant weather patterns influence seasonal pollution averages. This understanding can then be expanded to include the comparisons of the CMB results discussed later in this chapter (pp. 90-94).

For example, compared to 1982-83, winter 1979-80 was comprised of more than twice the number of Regime 1 days when weather patterns (i.e., winds < 3 mph, wet roads, no precipitation) are most conducive for accumulation of particulate, especially in the fine fraction. This preponderance of high particulate days is quite important because in spite of the fact that both winters had relatively close geometric means in Regime 1,*** this factor contributed to a higher seasonal TSP average during 1979-80.

In contrast, winter 1982-83 had over twice as many days in regimes conducive to high levels of coarse fraction particulates caused by dry roads during generally stagnant conditions (Regimes 3 and 4). At the same time, the occur-

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*** Both geometric and arithmetic means are shown in Table 18; however, the large variances associated with the small sample arithmetic means make useful comparisons impossible. Consequently, for the purposes of this work, only TSP geometric means will be specifically addressed.

rence of days in the "precipitation" regimes (5 and 6) was about the same during both winters. Finally, winter 1979-80 had 9 more days in the most unstable Regime 7, than did the second winter, and both winters had only four days of missing TSP data.

The real usefulness of this compilation becomes apparent through statistical comparison of the differences in average levels of particulates in each of the regimes. Such a comparison is displayed in Table 19, showing results of T-tests analyzing the hypothesis that the geometric means in both winters are equal. As shown in the table, this hypothesis is rejected for all seven regimes, demonstrating statistically significant differences that might not otherwise have been apparent. In addition, this comparison demonstrates the importance of fine (Regime 1) versus coarse (Regime 3) fraction particulate contributions to the seasonal mean, and allows for some speculation as to what might be the major particulate sources within the various distinct regimes. For example, assuming that Regime 1 is constructed to reflect accumulation of mostly fine fraction particulate, data in Table 19 suggests that there was a greater output from fine fraction sources during the second study winter. In contrast, using Regime 3 to indicate coarse fraction particulate, the first study winter showed a much larger impact from such sources. This points towards the importance of incorporating meteorological factors into analyses of chang-

TABLE 19

Results of T-Test Comparisons of Adjusted and Unadjusted TSP Means in Both Winters

Source/	1979-80		1982-83			
Regime	X[a]	σ	X	σ	t-value	Result[b]
1	172.5	1.7	177.8	1.3	11.12	Reject
2	131.2	1.6	96.2	1.6	77.74	Reject
3	251.7	1.7	178.0	1.5	96.59	Reject
4	136.3	1.9	128.3	1.5	10.38	Reject
5	130.3	1.2	119.0	1.3	12.77	Reject
6	109.4	1.6	73.4	1.5	74.30	Reject
7	59.4	1.7	55.9	1.7	7.13	Reject
UNADJUSTED MEAN						
	120.3	1.9	109.6	1.5	48.46	Reject
ADJUSTED MEAN						
	120.6	1.7	100.2	1.5	98.78	Reject

[a] All figures based on geometric statistics

[b] Reject (or Accept) the hypothesis that all means are equal at the .05 level of significance

ing air pollution situations and design of control strategies.

A final possible use for meteorological regime categorization is pointed out by the difference in the unadjusted and adjusted geometric means of winter 1982-83 (Table 18). Since the adjusted mean comprises both the known TSP concentration and the number of days occurring in each regime, the frequency of regime days affects the adjusted seasonal averages. For example, since three of the four missing days during 1979-80 were from the usually high particulate Regime 1, this inflates the adjusted seasonal mean. In contrast, since all four missing days from winter 1982-83 were in low to moderate particulate regimes, the adjusted weighting decreases the seasonal average. In the absence of complete ambient sampling data, this method facilitates a more complete analysis, and may, in fact, mean the difference between compliance and non-compliance with ambient air standards.

Reasonable Assumptions

Based on the above summation of the meteorological aspects of the two study winters, it would be reasonable to make certain predictions about the air pollution likely to occur during each winter. For example, in winter 1979-80 about 31 percent of the winter days had low wind speed, no precipitation, and wet or snow covered roads: conditions conducive to high levels of fine particulates, especially from combustion sources. In contrast, winter 1982-83 had only 13% of its days in this meteorological category. Therefore, it is likely that winter 1979-80 would show a greater contribution of fine particulate matter from combustion sources than would 1982-83. However, this was not the case. Further, it would be reasonable to expect 1982-83 to manifest somewhat higher levels of urban dust particulates since it had slightly more days with conditions conducive to this type source. However, 1979-80 had much higher levels of particulate in the "dust" regime, which apparently offset the greater occurrence of such days during 1982-83. These expectations are examined further after discussion of the meteorological categorization of the CMB results (pp.90-94).

CMB Results in Meteorological Regimes

Table 20 compares source contributions during the two study periods, by winter and by regime, for fine fraction particulate. Considering the uncategorized, seasonal means for RWC first, winter 1979-80 had an unadjusted mean 22.4 points lower than the same mean for winter 1982-83. After weighting the results based on the frequency of the weather categorized days, that difference decreased to 16.5 points, largely due to the increase in the adjusted mean for winter 1979-80.

Considering the uncategorized seasonal means of vehicle exhaust, winter 1979-80 had an unadjusted mean of 6.3% compared with 2.9% for winter 1982-83. After meteorological adjustment the mean for 1979-80 increased to 7.0% while the second winter mean increased slightly to 3.0%. In comparing both sets of seasonal means, winter 1982-83 showed a major decrease in contributions from vehicle emissions and from urban dust. The unadjusted mean contribution of urban dust for winter 1979-80 was 8.9% compared to an unadjusted mean of 3.6% in winter 1982-83; the adjusted means were 8.2% and 2.8% for 1979-80 and 1982-83, respectively.

TABLE 20

Percent Source Contributions in the Fine Fraction Within Meteorological Regimes

WINTER 1979-80

Regime	Total Days	Sample Days[a]	RESIDENTIAL WOOD COMB(%)			AUTO EXHAUST (%)			URBAN DUST (%)		
			Mean	StdDev	90% C.I.[b]	Mean	StdDev	90% C.I.	Mean	StdDev	90% C.I.
1	40	3[c]	91.7	27.0	46-137	5.7	1.0	5-7	2.0	0.0	---
2	23	2[d]	70.0	2.8	57-83	12.0	2.8	0-25	19.5	23.3	0-124
3	7	2[e]	72.5	14.8	6-139	6.0	2.8	0-19	20.0	9.9	0-64
6	19	4[f]	47.8	25.3	18-78	4.0	0.8	3-5	3.2	2.3	0-6
TOTAL	89	11									
UNADJUSTED MEAN:			68.3	26.4	55-81	6.3	3.3	5-8	8.9	11.8	3-15
ADJUSTED MEAN:			75.2	19.4	66-85	7.0	1.6	6-8	8.2	7.3	5-12

WINTER 1982-83

Regime	Total Days	Sample Days[a]	RESIDENTIAL WOOD COMB(%)			AUTO EXHAUST (%)			URBAN DUST (%)		
			Mean	StdDev	90% C.I.[b]	Mean	StdDev	90% C.I.	Mean	StdDev	90% C.I.
1	16	5[g]	84.0	15.2	70-98	2.0	0.0	2-2	1.2	1.1	0-2
2	31	5[h]	92.8	13.4	80-106	2.6	1.3	1-4	1.5	1.3	0-3
3	11	3[i]	92.3	11.5	73-112	3.0	1.0	2-4	8.7	2.5	4-13
4	17	2[j]	91.0	5.7	66-116	3.0	0.0	3-3	5.5	1.0	1-10
6	22	4[k]	94.0	11.7	80-108	3.5	0.6	3-4	2.5	2.6	0-6
7	21	1[l]	94.0	---	---	4.0	---	---	1.1	---	---
TOTAL	118	20									
UNADJUSTED MEAN:			90.7	11.9	86-95	2.9	1.0	3-3	3.6	3.6	2-5
ADJUSTED MEAN:			91.7	9.7	88-95	3.0	0.6	3-3	2.8	1.4	2-3

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[a] Sample days with acceptable CMB fits

[b] Calculated by the formula 90% C.I. = Mean ± (significance multiplier * Std Dev ÷ $\sqrt{N}$)

[c] Includes: Feb. 21, March 6 and 7

[d] Includes: Feb. 7 & 29

[e] Includes: Feb. 25,26,27

[f] Includes: Feb. 18,19,20,28

[g] Includes: Nov. 26, Dec. 5,11,14, and Jan. 1

[h] Includes: Nov. 23, Dec. 8,26,29, and Feb. 27

[i] Includes: Jan 31, Feb. 6,24

[j] Includes: Nov. 29, Dec. 2, Feb. 21

[k] Includes: Nov. 20, Dec. 20, Feb. 9

[l] Includes: Dec. 23

Differences In Source Contributions

In determining differences between the winters, it is necessary to conduct some statistical test to assess the "amount" of difference. For this I used T-tests, set at the 0.05 level to assess statistically significant differences between both unadjusted and adjusted means. This comparison is shown in Table 21. Based on this assessment, the contributions from residential wood combustion were significantly different for both set of means. Therefore, viewed as a seasonal whole, particulate composition during winter 1982-83 had a significantly higher percent contribution from residential wood combustion than winter 1979-80. This finding is the opposite of the prediction based strictly on the above meteorological regime categorization. In addition, it is also contrary to the expectations based on the findings of the 1980 and 1983 Missoula wood-use surveys which documented a slight decrease between 1980 and 1983 in the numbers of Missoula households that burned wood and in the tons of wood burned during the respective winters.

Considering vehicle exhaust, t-tests again demonstrated statistically significance differences, but only after meteorological adjustment. However, since Missoula's population of vehicles increased between 1980 and 1983,**** the

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**** Figures from the Montana Department of Motor Vehicles showed an increase in the number of registered passenger cars and pickup trucks from 57,391 in 1980 to 58,529 in 1983.

TABLE 21

Results of T-Test Comparisons of Source Contributions Using Meteorologically Adjusted and Unadjusted Means

	Unadjusted Contrib (%)				Adjusted Contrib (%)			
	1979-80		1982-83		1979-80		1982-83	
Source	X	σ	X	σ	X	σ	X	σ
RWC	68.3	26.4	90.7	11.9	75.2	19.4	91.7	9.7
	t = 3.23 (Reject)[a]				t = 3.14 (Reject)			
AUTO	6.3	3.3	2.9	1.0	7.0	1.6	3.0	0.6
	t = 1.69 (Accept)				t = 4.15 (Reject)			
DUST	8.9	11.8	3.6	3.6	8.2	7.3	2.8	1.4
	t = 1.86 (Reject)				t = 3.20 (Reject)			

[a] Reject (or Accept) the hypothesis that winter mean contributions are equal

absolute number of "sources" could not be a factor in the decrease.

As with the previous source, the average contribution of urban dust showed a statistically significant decrease from the first study winter to the second. Again, this is contrary to the prediction that winter 1982-83 might show a greater contribution of particulate from urban dust (assuming that the larger number of "dry" regime days in 1982-83 sufficiently offset the higher TSP means seen in the same regimes during 1979-80).

Reasons For Unexpected Results

The above findings are contrary to the expectations based on preliminary review of weather patterns and the contributions of pollution sources during the two study winters. This suggests that there are problems with either the meteorological regime techniques, with one or more aspects of the CMB analysis, or other facets not yet addressed. In search of possible explanations, consider the following.

Residential Wood Combustion

Looking at only Regime 1, with weather parameters "favorable" to particulates from sources other than urban dust (e.g., combustion particles from residential wood burning and vehicle exhaust), the RWC regime mean for winter 1979-80 appears to be higher than the mean of the same regime in winter 1982-83, as was expected. However, a t-test demonstrated no statistical difference between the two means (Table 22). Therefore, it appears that the residential wood combustion (RWC) contributions in the month of February in Regime 1 are statistically the same as those in Regime 1 for the second study winter. However, a factor affecting the results of the statistical test for difference is the rather large standard deviation for the RWC contribution in the first regime for winter 1979-80. The mean for Regime 1 was calculated from three sample days meant to represent the largest regime for the entire winter. This introduces the potential for a consequential error, especially since the

TABLE 22

Results of T-Test Comparisons Between Same Regimes in Both Study Winters

Source/	1979-80 (%)		1982-83 (%)			
Regime	X	σ	X	σ	t-value	Result[a]
RWC/						
1	91.7	27.0	84.0	15.2	0.53	Accept
2	70.0	2.8	92.8	13.4	2.25	Reject
3	72.5	14.8	92.3	11.5	1.71	Accept
6	47.8	25.3	94.0	11.7	3.30	Reject
AUTO/						
1	5.7	1.0	2.0	0.0	4.96	Reject
2	12.0	2.8	2.6	1.3	4.96	Reject
3	6.0	2.8	3.0	1.0	1.75	Accept
6	4.0	0.8	3.5	0.6	0.95	Accept
DUST/						
1	2.0	0.0	1.2	1.1	0.91	Accept
2	15.5	23.3	1.5	1.3	2.04	Reject
3	20.0	9.9	8.7	2.5	2.05	Accept
6	3.2	2.3	2.5	2.6	0.40	Accept

[a] Reject (or Accept) the hypothesis that regime means are equal

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three days constituting Regime 1 were in late February and early March, at the very end of the cold weather season when average temperatures are usually increasing. In contrast, Regime 1 in the second study winter was comprised of sampling days in late November, in December, and early January when average temperatures are usually quite low.

As mentioned earlier, the average seasonal RWC contributions for the two study winters were found to be statistically different at the .05 level. The difference is most easily explained by the fact that in the second study winter, sampling for all regimes was spread throughout the whole winter rather than just through February. Also mentioned earlier, wood use surveys conducted in 1980 and 1983 documented a slight decrease in the number of RWC sources in the valley. This decrease was not reflected in the CMB results, but whether that was due to the discrepancy in the sampling periods or in the questionable CMB results of the 1982-83 study is not possible to discern.

Auto Exhaust

As stated above, the relative size of the "vehicle exhaust" source increased between the two study periods, and therefore might have affected an increase in its contribution to fine fraction particulates. Instead there was an apparent decrease. A possible explanation lies in the fact that the CMB analysis relied heavily upon the use of lead as the primary indicator element for vehicle exhaust, especially since carbonaceous materials could not be used in the CMB fitting process. The number of vehicles using lead-free gasoline has been steadily increasing since the late 1970s[1], and the amount of lead in leaded gasoline has decreased during the same period.[2] Additionally, total emissions from vehicles may have decreased as newer models with more effective emission control equipment replaced older models. Further, the 1979-80 monitoring site was located within a few yards of an intersection on a busy city street, while the 1982-83 site was located at least one-half city block away from an intersection and was somewhat screened from that intersection by a house and various vegetation. Thus, the 1979-80 site was far more likely to be affected by vehicle emissions than the 1982-83 site.

Examining means from the individual regimes, it is possible to discern a discrepancy in the vehicle source contributions between the two winters: in winter 1979-80 nearly all means are close except in Regime 2 (light winds, wet roads, no precipitation) whose mean was over twice as high as those in any of the other regimes. In contrast, in winter 1982-83, all regimes had means which showed little variation. This difference could be explained by the same factors as those which affected the seasonal means, especially the difference in the location of the monitoring site.

Urban Dust

Contrary to the prediction that winter 1982-83 should show a higher contribution from urban dust, winter 1979-80 had the higher contribution both before and after adjustment for meteorology. This may have resulted from some of the same factors as affected the contribution from vehicle exhaust (e.g., the site's proximity to a dusty intersection). In addition, meteorological factors not considered by the structured regimes, may have also played a role in this apparent discrepancy.*****

As might be expected, urban dust was a major contributor to air pollution in both study winters during the dry regime (3), but no statisically significant difference was discernable between the two winters. However, dust was a primary contributor in a wet regime (2) only in the first study winter, and showed a statisically significant decline by the second winter.

Final Comments

For all three sources of fine particulates, hypotheses can be formulated which might explain why the CMB-derived contributions from those sources changed from winter 1979-80 to 1982-83. However, these hypotheses would be based on the fundamental assumption that the input data are essentially valid, and this chapter has elucidated many problems and questions concerning much of the data from both studies. In

---

***** For example, almost twice as much snow fell during the first study winter (39.7") compared with only 23.2" during the second winter. The greater snowfall in the first winter would almost certainly result in increased distribution of street-sanding material, but this likelihood cannot be verified because no records exist concerning the amount of sanding materials distributed on city streets.

Another complicating factor in this consideration is the fact that beginning in winter 1982-83 the City of Missoula assumed the responsibility for winter maintenance of arterial routes within the city limits which had previously been maintained by the State Highway Department which applied salt instead of sand. According to Joe Aldegarie, Director of Public Works for the City of Missoula, this change resulted in a greater potential for the application of sanding material on city streets than ever before, increasing the amounts of material that might eventually become airborne.

spite of this failing, the fact remains that this is the only Missoula-specific data available to local officials concerned with devising the means for reducing particulate pollution. Further, while both studies had major flaws which preclude the presentation of results in absolute terms, it is likely that a perfect study of the Missoula situation would result in data within the same range of the results of the studies examined here. In other words the "true" percentage contributions of particulates are probably somewhere within the bounds defined by these two studies.

During the first study winter, all samples were collected in late winter when contributions from residential wood combustion are normally expected to decrease and contributions from urban dust to increase. Therefore, the adjusted average of 75% contribution from RWC during 1979-80 could conceivably be a low estimate. For the 1982-83 study, the inability to use carbonaceous data may have inflated the apparent contribution for RWC, therefore the adjusted 92% contribution from this source may be a high estimate, although fine fraction particulates made up over 90% of the TSP during many days of both studies. Both numbers could therefore represent a likely range. Viewed in this context and with an understanding of their limitations, the results of both source apportionment studies are useful in developing strategies for controlling wintertime particulate pollution.

References Cited in Chapter 4

1. US Environmental Protection Agency, 1985, "Compilation of Air Pollutant Emission Factors," Volume II: Mobile Sources; Office of Air and Radiation, Office of Mobile Sources, Test and Evaluation Branch, Ann Arbor, MI AP-42, Fourth Edition, September 1985

2. Ibid.

CHAPTER 5

SUMMARY AND RECOMMENDATIONS

Summary

With the completion of this work, Missoula pollution control officials have results from two source apportionment studies of wintertime particulates. The first study, conducted during February and early March 1980, revealed the following information.

1) Residential wood combustion was the primary source of fine fraction particulates, averaging 68 ± 8 percent.

2) Urban dust contributed the second largest amount of fine fraction mass, producing 9 ± 4 percent.

3) Vehicle exhaust and a hog fuel boiler point source contributed 6 ± 1 and 2 ± 0.3 percent, respectively.

4) Urban dust was the primary source of coarse fraction particulates averaging 54 ± 3 percent; residential wood combustion contributed 17 ± 2 percent of the coarse fraction mass.

5) Approximately 85 ± 9 percent of the fine fraction and 72 ± 16 percent of the coarse fraction mass were explained by the CMB analytical process.

The second source apportionment study, conducted during November 1982, through February 1983, provided the following results.

1) Residential wood combustion was still the largest source of fine fraction particulate mass, averaging 91 ± 3 percent.

2) Urban dust was again the second largest contributor, producing 4 ± 1 percent.

3) Vehicle exhaust contributed about 3 percent of the fine particle mass and no other sources were found.

4) No coarse fraction source apportionment was possible.

5) The CMB procedure accounted for 97 ± 3 percent of the fine fraction particulate mass.

I compared the results of the two studies, beginning with the application of a technique called meteorological regime categorization to adjust for meteorological variation between the two study periods. Based on that initial examination of the weather and changes in the sources of Missoula's air pollution, I made the following predictions regarding possible differences in source contributions from 1980 to 1982-83.

1) Since the first study winter had over twice the number of days meteorologically favorable to both the production and accumulation of high levels of fine fraction particles, I predicted that 1979-80 should manifest higher source contributions from fine fraction combustion sources such as residential wood burning.

2) In contrast, 1982-83 was more meteorologically suited to production and accumulation of coarse fraction particulate, and I predicted that 1982-83 should manifest higher source contributions from coarse fraction sources like urban dust.

Contrary to these expectations, the source apportionment study analysis produced the opposite results. The reasons for these discrepancies became apparent with close examination of both the CMB input data and the meteorological parameters used in the analysis. These explanations are summarized below.

Coarse Fraction

No coarse fraction apportionment was possible in the 1982-83 study principally because there were no Missoula-specific source finger prints available explicitly for urban dust.

Fine Fraction

In the 1982-83 study, it was possible to successfully apportion only 20 out of 35 fine fraction samples using CMB. I believe this result was due to three reasons. First, it was not possible to incorporate organic composition data from hi-vol TSP samples into the CMB analyses using elemental data from dichot fine fraction filters. Consequently, the entire CMB process utilized only XRF inorganic composition data. Second, there were no Missoula-specific source fingerprints, leaving the process dependent on library fingerprints of similar, but not necessarily identical sources. Finally, with no organic data available for use in the CMB analysis, it was not possible to include or identify effectively any point sources of fine fraction particulate, which probably inflated the apparent contributions from at least one other source: residential wood combustion.

Judging from the results of the two source apportionment studies, the contribution of residential wood combustion to the fine particulate increased dramatically from 1979-80 to 1982-83. However, this apparent difference is likely the result of one or more of the following reasons. First, the 1979-80 study actually represents the end of that winter when fine particle source contributions were lower than during much of the rest of the winter. Second, as mentioned above, the 1982-83 results may have been inflated. And finally, the 1982-83 study depended on a majority of samples from the month of December, which is likely to be a time with highest combustion-source contributions and lowest urban dust impacts. Consequently, this high figure does not necessarily represent the entire winter situation any better than does the 1979-80 study.

At the same time residential wood burning contributions appeared to be increasing, the contribution from auto exhaust was apparently declining significantly. However, this change is likely the result of a combination of the following factors. First, the 1982-83 receptor site was further from an intersection of city streets; and second, the CMB source fingerprint for auto exhaust depended greatly on lead as an indicator element, and from 1979-80 to 1982-83, both the number of leaded-gas vehicles and the amount of lead remaining in leaded gas declined.

Urban dust contributions to the fine fraction also apparently decreased between the two studies. This may be explained by one or more of the following reasons: (1) the 1982-83 receptor site was farther from an intersection and somewhat screened from this dust source by vegetation; (2) no Missoula-specific source fingerprint was available for the 1982-83 CMB analysis; (3) less sanding material was probably spread on Missoula streets during the second study winter because half as much snow fell during the winter and almost no snow accumulated after mid-January 1983; and (4) the preponderance of December samples in the second CMB analysis would likely underrepresent winter long urban dust contributions, while the first study may well overrepresent this source.

Recommendations

Because both Missoula source apportionment studies are restricted in their applicability by either limitations or defects, it is essential that a third study be conducted. Such a study should incorporate the best elements of the two previous works, and insofar as financially possible, eliminate their problems. Only with such an effort will pollution control officials and the public finally have a definitive analysis of the nature and sources of Missoula's wintertime particulates.

Further, I recommend that the study be designed to include the following components:

1) sample collection every other day throughout an entire winter, from mid-October through mid-March;

2) collection of two complete sets of fractionated samples; one set for inorganic analysis (including XRF, ionic, and instrumental neutron activation for sodium and magnesium), and the other for organic analysis;

3) complete identification and collection of Missoula source samples (especially urban dust);

4) meteorological regime analysis for the entire sampling period to assist final selection of samples for chemical and CMB analysis, and assist in final interpretation; and

5) analysis by chemical mass balance.

APPENDIX A

DESCRIPTION OF CMB INPUT FILES

Receptor Data Set Files

1. MASS.DAT - containing the sample ID, sample mass and associated uncertainty, and a mass flag if needed
2. FIELD.DAT - containing the sample ID, a collection site code, sample collection start time and duration, sample size, volume, and uncertainty, and collection flow ratio if needed
3. METHODS.DAT - for specifying what analytical methods were used to derive the raw elemental data
4. METHODnm - containing the analytical results, uncertainties, and any needed conversion factors

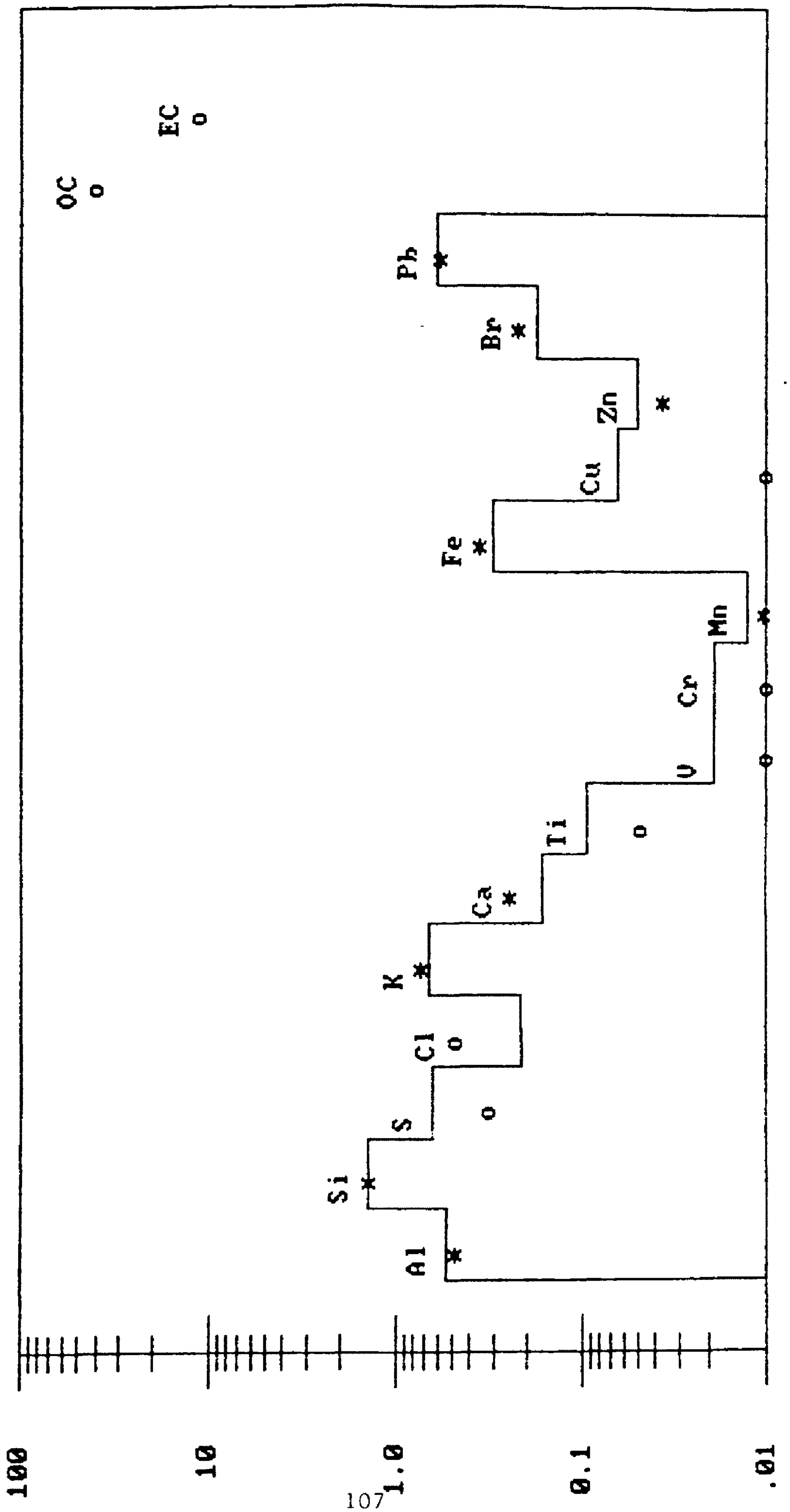
APPENDIX B

TABLES AND HISTOGRAMS OF CMB FITS

```
SAMPLE ID: MMF38                              PARTICLE SIZE: FINE
FIELD FLAG:    MASS FLAG:    ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 830227  START TIME:   .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:    .598  DEGREES OF FREEDOM:  4

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  15.830+- 2.206   98.939+-17.617
          5017  ROADS  F   1.838+-  .216   11.490+- 1.857
          5018  TRANS  F    .862+-  .129    5.386+- 1.004
          ----------------------------------------------
               TOTAL:     18.530+- 2.220  115.815+-18.905

---SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.-------
   Al   *     .170+-  .052   1.063      .153+-  .006    .903+- .277  Al
   Si   *     .443+-  .070   2.769      .443+-  .024    .999+- .166  Si
   S          .117+-  .008    .731      .070+-  .043    .598+- .368  S
   Cl         .023+-  .002    .144      .102+-  .008   4.416+- .572  Cl
   K    *     .153+-  .011    .956      .153+-  .015   1.000+- .127  K
   Ca         .047+-  .003    .294      .067+-  .024   1.418+- .524  Ca
   Ti   *     .020+-  .003    .125      .015+-  .004    .775+- .222  Ti
   V          .005+-  .002    .031      .000+-  .000    .085+- .052  V
   Cr         .005+-  .002    .031      .000+-  .000    .096+- .052  Cr
   Mn         .005+-  .001    .031      .002+-  .000    .404+- .096  Mn
   Fe   *     .094+-  .007    .588      .106+-  .017   1.128+- .200  Fe
   Cu         .014+-  .001    .087      .001+-  .001    .053+- .041  Cu
   Zn         .010+-  .001    .063      .008+-  .001    .755+- .121  Zn
   Br   *     .034+-  .003    .212      .046+-  .016   1.348+- .470  Br
   Pb   *     .129+-  .009    .806      .121+-  .019    .940+- .153  Pb
   OC                                  8.075+- 2.186                 OC
   EC                                  2.277+- 1.332                 EC
-----------------------------------------------------------------------
   MASS     16.0  +- 1.8          * FITTING SPECIES
```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1.0
0.1
.01
Al
Si
S
Cl
K
Ca
Ti
V
Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
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FIELD FLAG:    MASS FLAG:    ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 830224  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .908  DEGREES OF FREEDOM:  7

         --SOURCE----SIZE----UG/M3------------PERCENT---
         5015  WSMOKE F  43.384+- 4.562   92.307+-14.179
         5017  ROADS  F   5.068+-  .535   10.783+- 1.659
         5018  TRANS  F   2.082+-  .312    4.431+-  .828
         -----------------------------------------------
             TOTAL:      50.535+- 4.604  107.520+-15.520

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .690+-  .210   1.468      .423+-  .016    .612+- .188  Al
   Si   *    1.470+-  .232   3.128     1.218+-  .066    .829+- .138  Si
   S          .545+-  .038   1.160      .181+-  .104    .332+- .191  S
   Cl         .066+-  .004    .140      .272+-  .021   4.118+- .424  Cl
   K    *     .398+-  .028    .847      .420+-  .042   1.055+- .129  K
   Ca   *     .106+-  .007    .226      .180+-  .064   1.699+- .619  Ca
   Ti   *     .028+-  .003    .060      .040+-  .010   1.414+- .379  Ti
   V                  .007              .001+-  .001    .000+- .000  V
   Cr         .003+-  .002    .006      .001+-  .000    .439+- .334  Cr
   Mn   *     .005+-  .001    .011      .006+-  .001   1.115+- .266  Mn
   Fe   *     .290+-  .021    .617      .287+-  .042    .989+- .163  Fe
   Cu         .018+-  .001    .038      .002+-  .002    .113+- .088  Cu
   Zn   *     .022+-  .001    .047      .020+-  .002    .930+- .116  Zn
   Br   *     .095+-  .007    .202      .111+-  .038   1.166+- .406  Br
   Pb   *     .309+-  .022    .657      .294+-  .043    .951+- .155  Pb
   OC                                 22.012+- 5.991                 OC
   EC                                  6.168+- 3.650                 EC
----------------------------------------------------------------------
   MASS     47.0  +- 5.3            * FITTING SPECIES
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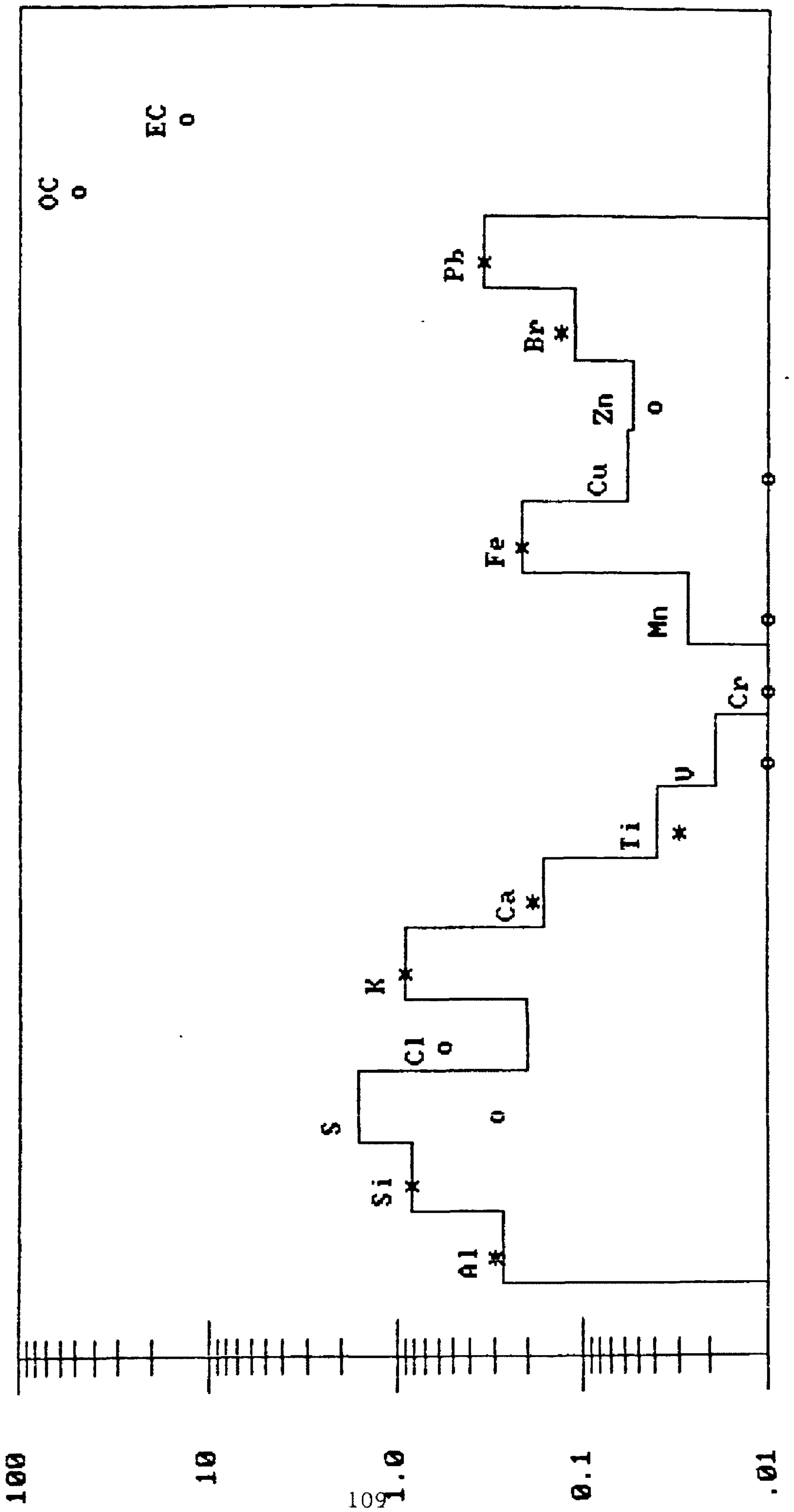
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S
Cl
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Ca
Ti
V
Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF18                               PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821229  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .269  DEGREES OF FREEDOM:  3

          --SOURCE-----SIZE----UG/M3-------------PERCENT---
          5015  WSMOKE F  28.743+- 3.834   71.859+-12.508
          5017  ROADS  F     .496+-  .077    1.239+-  .237
          5018  TRANS  F     .802+-  .120    2.004+-  .375
          -----------------------------------------------
                TOTAL:    30.041+- 3.837   75.102+-12.749

--SPECIES-----MEAS. UG/M3------%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .057+-  .018     .142      .047+-  .003    .830+- .272   Al
   Si   *     .122+-  .019     .305      .123+-  .007  1.006+- .166   Si
   S          .538+-  .038    1.345      .086+-  .040    .159+- .075   S
   Cl         .084+-  .006     .210      .165+-  .011  1.968+- .188   Cl
   K    *     .251+-  .018     .628      .252+-  .028  1.003+- .132   K
   Ca         .028+-  .002     .070      .041+-  .014  1.465+- .496   Ca
   Ti         .004+-  .003     .010      .006+-  .003  1.447+-1.325   Ti
   V              <   .006               .000+-  .000    .000+- .000   V
   Cr             <   .005               .000+-  .000    .000+- .000   Cr
   Mn         .002+-  .001     .005      .001+-  .000    .273+- .200   Mn
   Fe   *     .032+-  .003     .080      .039+-  .014  1.222+- .463   Fe
   Cu         .026+-  .002     .065      .000+-  .000    .008+- .012   Cu
   Zn         .019+-  .001     .047      .012+-  .001    .611+- .067   Zn
   Br   *     .036+-  .003     .090      .043+-  .015  1.185+- .413   Br
   Pb   *     .117+-  .008     .293      .111+-  .017    .951+- .157   Pb
   OC                                  14.048+- 3.967                  OC
   EC                                   3.896+- 2.416                  EC
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   MASS     40.0  +-  4.5            * FITTING SPECIES
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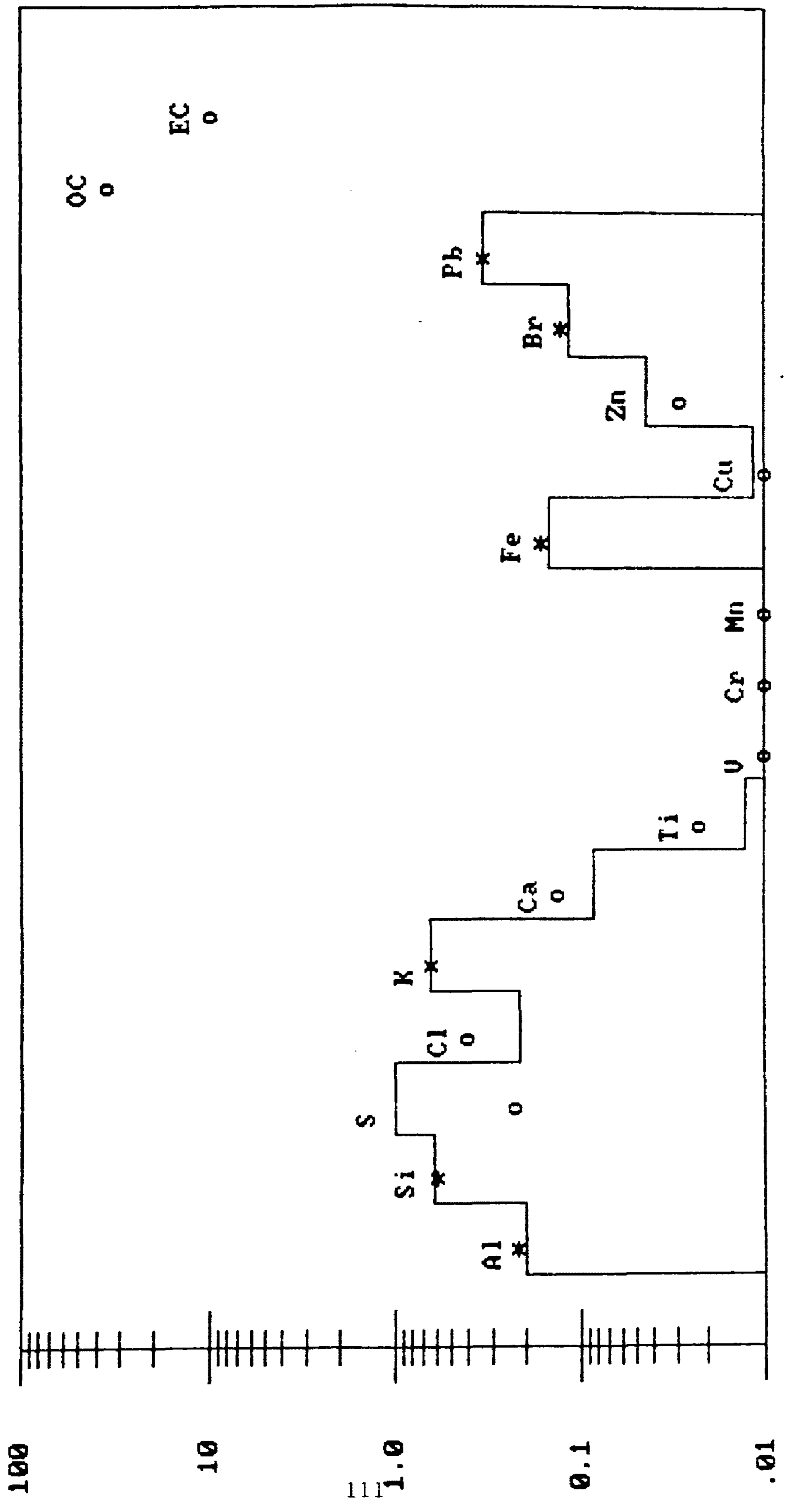
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1.0
0.1
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Si
S
Cl
K
Ca
Ti
V
Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF19                              PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 830101  START TIME:   .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:    .628  DEGREES OF FREEDOM:  5

          --SOURCE-----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  70.985+- 6.497   82.540+-11.915
          5017  ROADS  F   1.431+-  .215    1.664+-  .311
          5018  TRANS  F   1.886+-  .280    2.194+-  .407
          -----------------------------------------------
               TOTAL:     74.302+- 6.506   86.398+-12.258

--SPECIES----MEAS. UG/M3------%--------CALC. UG/M3---CALC./MEAS.------
   Al   *      .177+-  .054     .206       .133+-  .009   .754+- .234   Al
   Si   *      .345+-  .055     .401       .352+-  .019  1.019+- .171   Si
   S           .759+-  .054     .883       .209+-  .094   .275+- .126   S
   Cl          .190+-  .014     .221       .406+-  .026  2.138+- .209   Cl
   K    *      .549+-  .039     .638       .624+-  .068  1.136+- .148   K
   Ca          .049+-  .003     .057       .105+-  .033  2.151+- .697   Ca
   Ti          .007+-  .003     .008       .015+-  .007  2.191+-1.396   Ti
   V                   .007                .000+-  .001   .000+- .000   V
   Cr                  .005                .000+-  .001   .000+- .000   Cr
   Mn   *      .002+-  .001     .002       .002+-  .001   .787+- .536   Mn
   Fe   *      .081+-  .006     .094       .105+-  .034  1.296+- .433   Fe
   Cu          .033+-  .003     .038       .001+-  .001   .017+- .025   Cu
   Zn   *      .032+-  .003     .037       .029+-  .002   .896+- .101   Zn
   Br   *      .095+-  .007     .110       .100+-  .034  1.056+- .368   Br
   Pb   *      .267+-  .019     .310       .262+-  .039   .982+- .162   Pb
   OC                                    34.674+- 9.797                 OC
   EC                                     9.599+- 5.965                 EC
---------------------------------------------------------------------
   MASS     86.0   +-  9.6           * FITTING SPECIES
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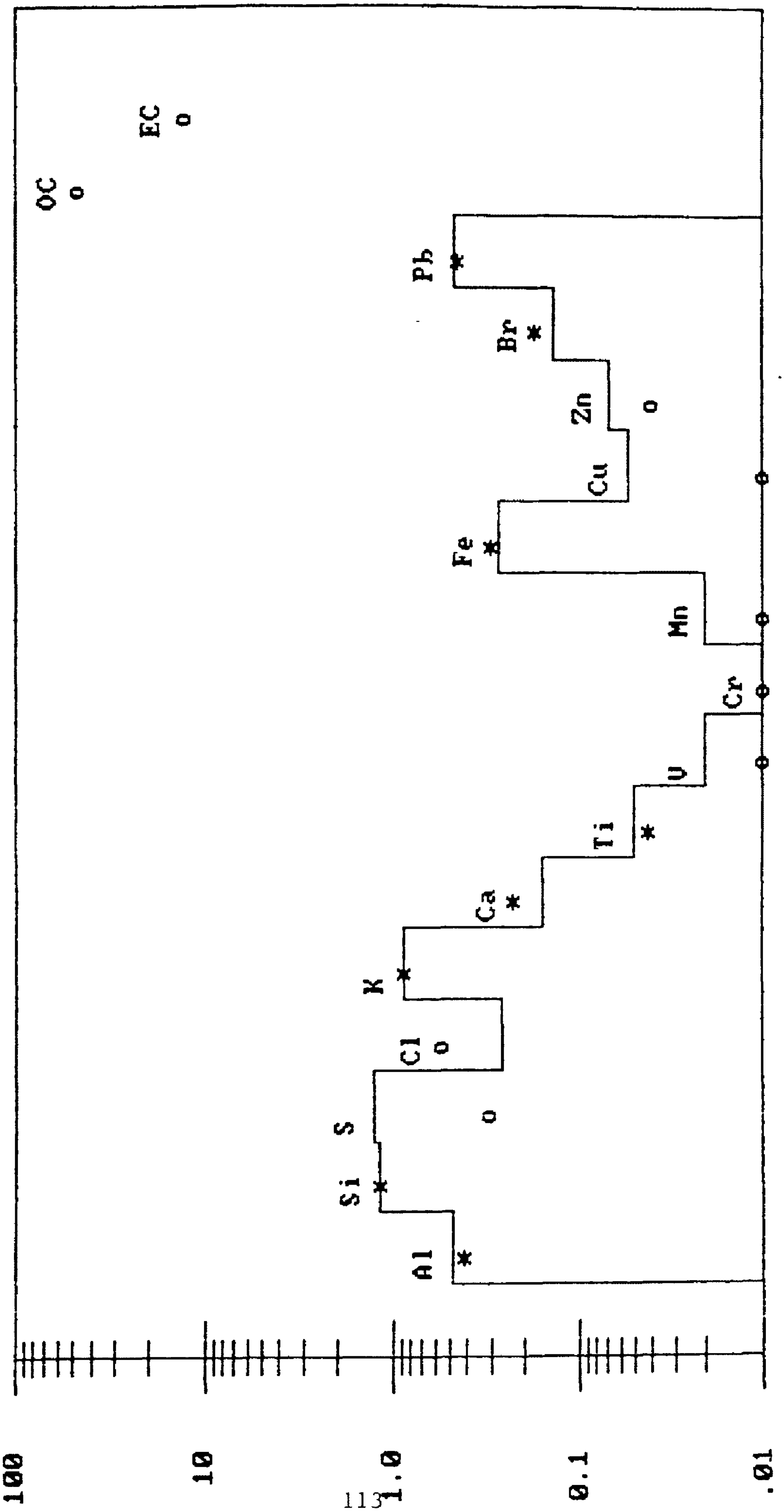
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Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
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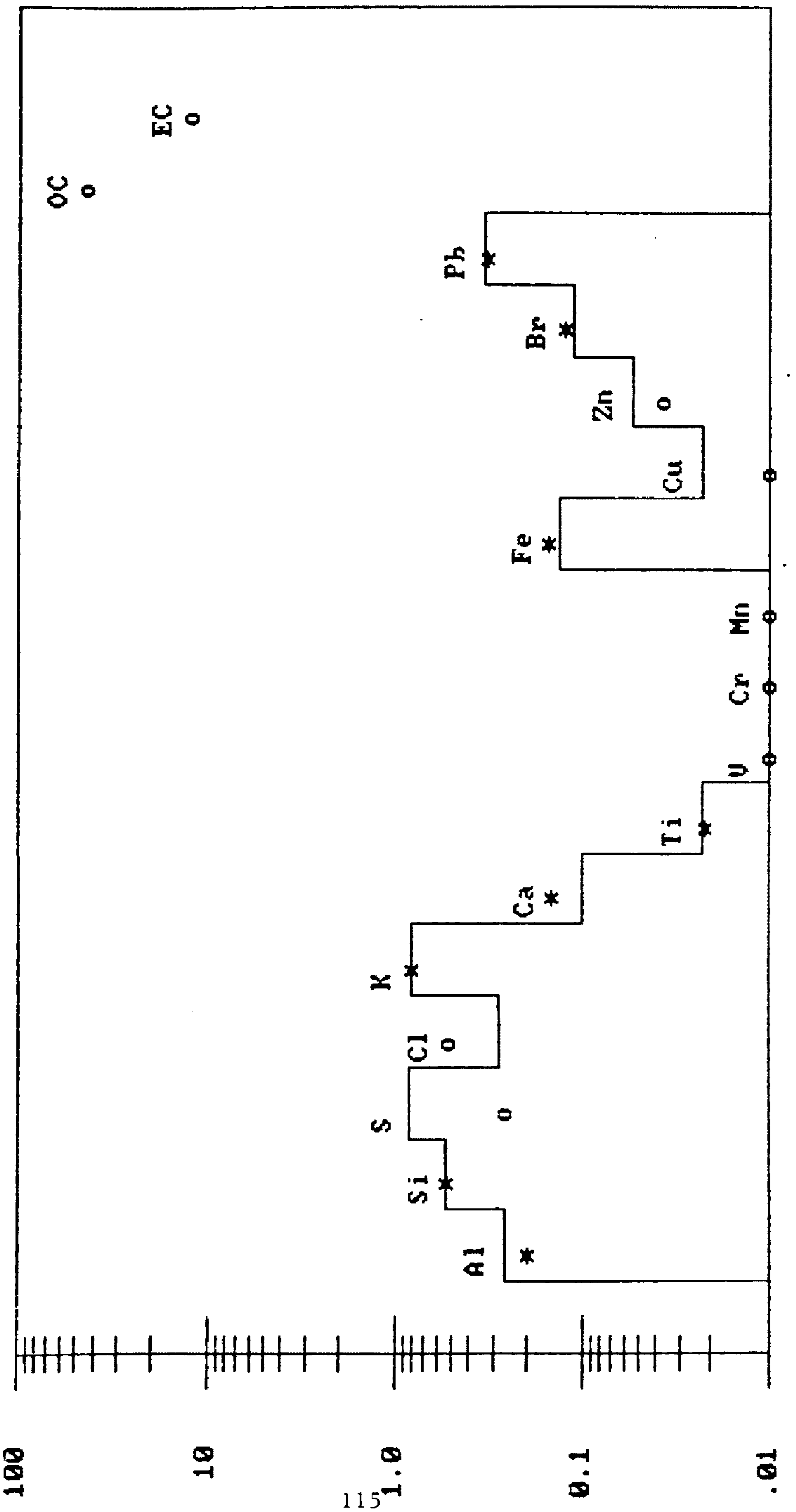
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 830122  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .318  DEGREES OF FREEDOM:   4

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  51.013+- 4.801  100.025+-14.595
          5017  ROADS  F   1.555+-  .231    3.049+-  .567
          5018  TRANS  F   1.914+-  .283    3.752+-  .695
          ------------------------------------------------
               TOTAL:     54.481+- 4.815  106.826+-15.200

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al  *     .122+-  .037     .239     .139+-  .007  1.142+-  .356  Al
   Si  *     .408+-  .064     .800     .381+-  .021   .935+-  .156  Si
   S         .642+-  .045    1.259     .174+-  .095   .271+-  .150  S
   Cl        .132+-  .010     .259     .305+-  .022  2.312+-  .236  Cl
   K   *     .418+-  .030     .820     .453+-  .049  1.084+-  .140  K
   Ca        .056+-  .004     .110     .095+-  .034  1.704+-  .626  Ca
   Ti        .011+-  .004     .022     .016+-  .007  1.476+-  .857  Ti
   V         .007+-  .003     .014     .000+-  .001   .051+-  .077  V
   Cr        .003+-  .002     .006     .000+-  .001   .135+-  .194  Cr
   Mn        .007+-  .002     .014     .002+-  .001   .244+-  .104  Mn
   Fe  *     .100+-  .007     .196     .112+-  .035  1.116+-  .357  Fe
   Cu        .020+-  .001     .039     .001+-  .001   .031+-  .035  Cu
   Zn  *     .023+-  .002     .045     .021+-  .002   .929+-  .108  Zn
   Br  *     .097+-  .007     .190     .102+-  .035  1.050+-  .365  Br
   Pb  *     .268+-  .019     .525     .266+-  .040   .993+-  .167  Pb
   OC                                25.212+- 7.042                 OC
   EC                                 7.051+- 4.289                 EC
----------------------------------------------------------------------
   MASS    51.0  +- 5.7          * FITTING SPECIES
```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Mn
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Zn
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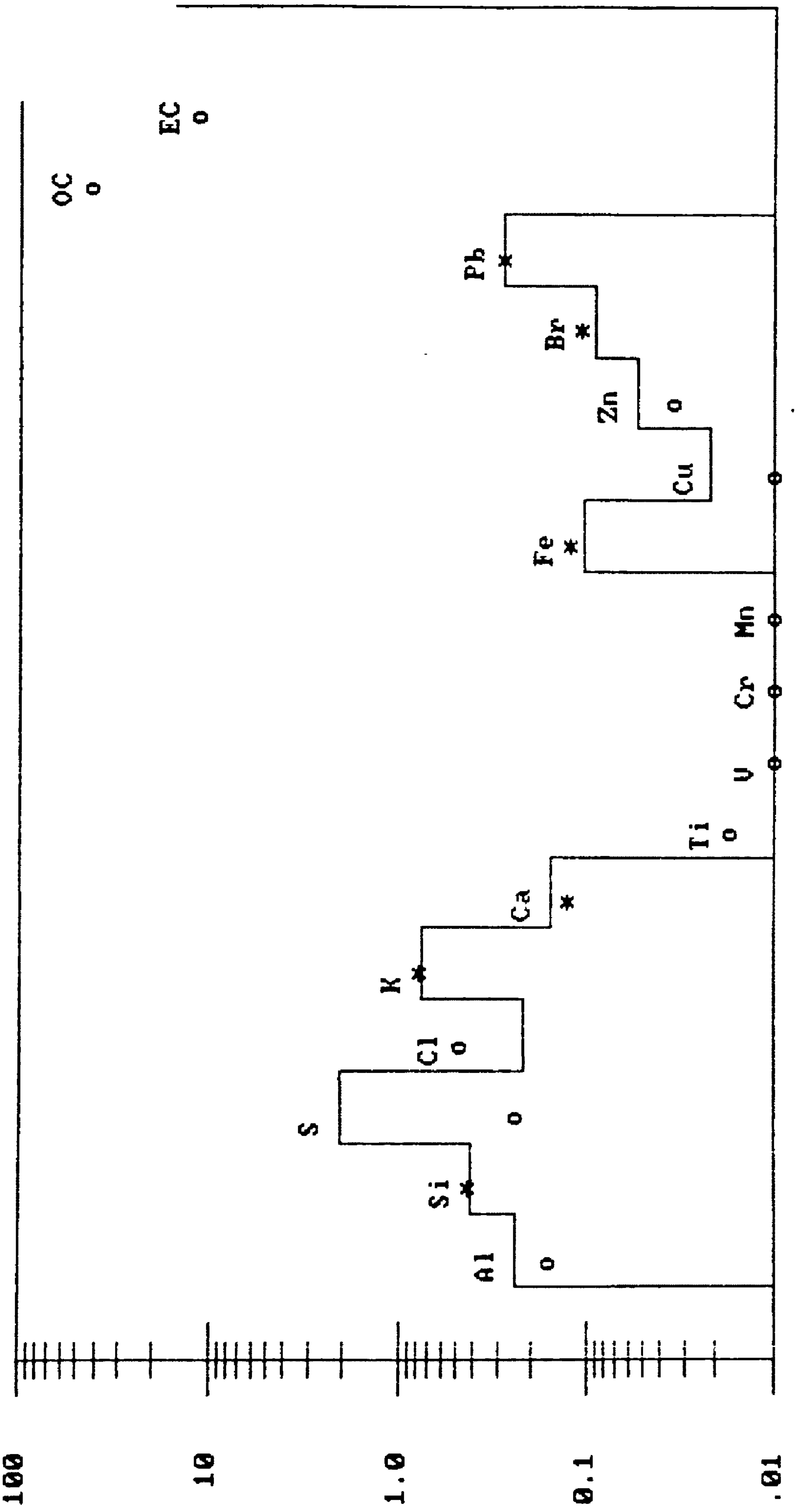
Pb
OC
EC

```
SAMPLE ID: MMF29                                 PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 830131  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .252  DEGREES OF FREEDOM:  5

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  56.398+- 5.557   80.569+-12.007
          5017  ROADS  F   6.513+-  .674    9.305+- 1.417
          5018  TRANS  F   2.183+-  .328    3.119+-  .584
          -----------------------------------------------
              TOTAL:      65.095+- 5.608   92.992+-13.125

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al   *     .607+-  .185     .867       .542+-  .021   .894+- .274  Al
   Si   *    1.480+-  .232    2.114      1.563+-  .085  1.056+- .175  Si
   S          .651+-  .046     .930       .214+-  .109   .329+- .169  S
   Cl         .159+-  .011     .227       .341+-  .024  2.144+- .216  Cl
   K    *     .524+-  .037     .749       .545+-  .054  1.040+- .127  K
   Ca         .105+-  .007     .150       .226+-  .079  2.154+- .770  Ca
   Ti         .027+-  .004     .039       .049+-  .011  1.827+- .492  Ti
   V                  .003                .001+-  .001   .499+- .550  V
   Cr         .004+-  .002     .006       .002+-  .001   .423+- .263  Cr
   Mn   *     .009+-  .002     .013       .007+-  .001   .796+- .205  Mn
   Fe   *     .346+-  .024     .494       .360+-  .047  1.039+- .154  Fe
   Cu         .034+-  .003     .049       .003+-  .002   .077+- .060  Cu
   Zn   *     .027+-  .002     .039       .026+-  .002   .966+- .107  Zn
   Br   *     .106+-  .007     .151       .116+-  .040  1.096+- .380  Br
   Pb   *     .316+-  .022     .451       .310+-  .045   .980+- .159  Pb
   OC                                   28.382+- 7.786                OC
   EC                                    7.880+- 4.742                EC
---------------------------------------------------------------------
   MASS    70.0  +- 7.8           * FITTING SPECIES
```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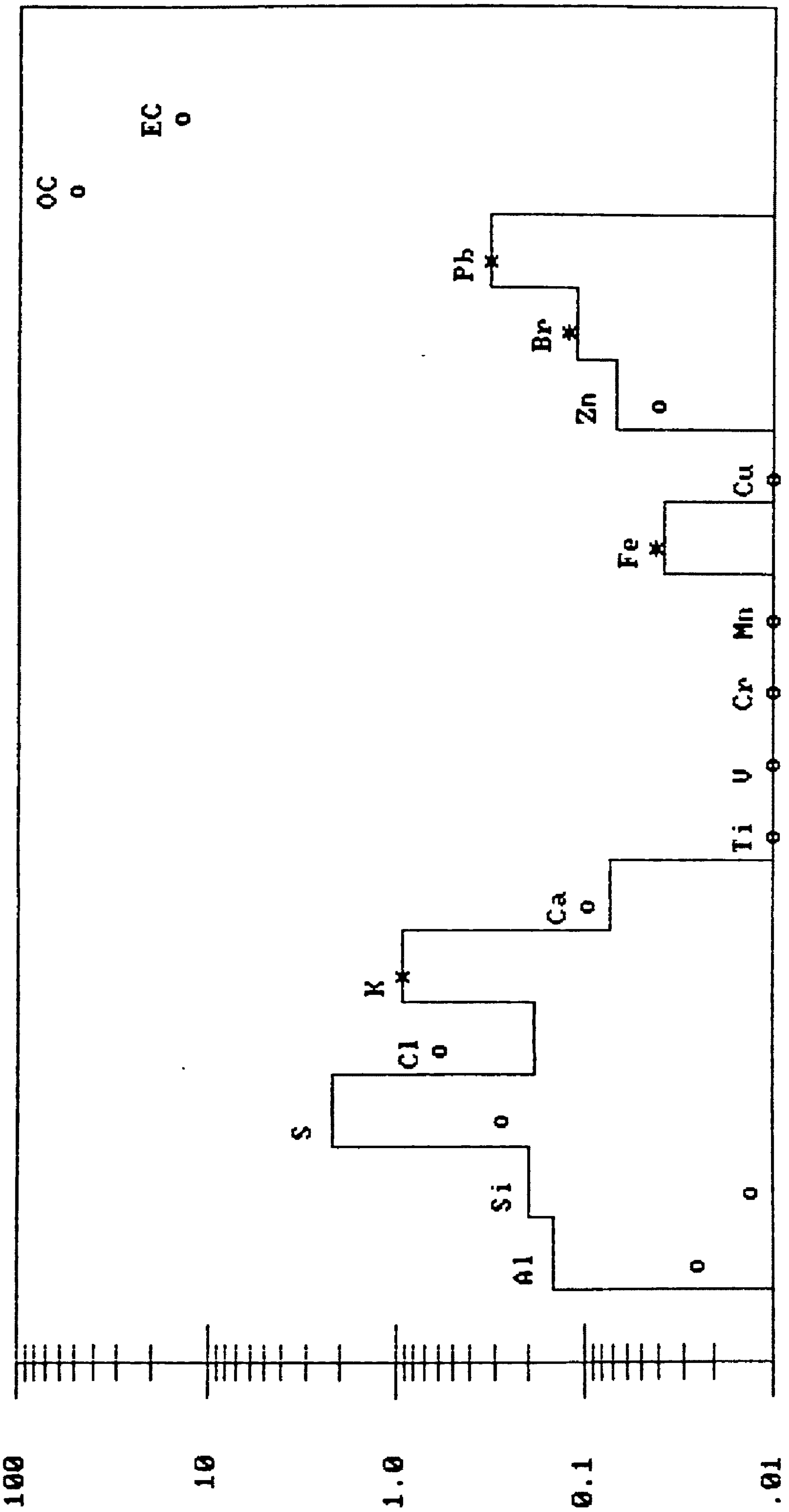
OC
EC

```
SAMPLE ID: MMF31                                PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 830206  START TIME:   .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .114  DEGREES OF FREEDOM:  4

         --SOURCE----SIZE----UG/M3------------PERCENT---
         5015  WSMOKE F  48.045+- 6.478  104.445+-18.273
         5017  ROADS  F   2.647+-  .293    5.754+-  .905
         5018  TRANS  F    .984+-  .148    2.139+-  .400
         -----------------------------------------------
              TOTAL:     51.676+- 6.487  112.339+-18.860

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al  *     .202+-  .062     .439      .226+-  .009  1.118+- .345  Al
   Si  *     .679+-  .108    1.476      .636+-  .034   .936+- .157  Si
   S         .459+-  .032     .998      .137+-  .050   .298+- .110  S
   Cl        .094+-  .007     .204      .269+-  .017  2.859+- .274  Cl
   K   *     .438+-  .031     .952      .438+-  .046   .999+- .127  K
   Ca        .091+-  .007     .198      .110+-  .033  1.207+- .373  Ca
   Ti  *     .019+-  .004     .041      .020+-  .005  1.071+- .338  Ti
   V            <    .003               .001+-  .001   .203+- .264  V
   Cr        .004+-  .002     .009      .001+-  .000   .172+- .150  Cr
   Mn        .007+-  .002     .015      .003+-  .001   .416+- .144  Mn
   Fe  *     .147+-  .010     .320      .148+-  .021  1.006+- .156  Fe
   Cu        .026+-  .002     .057      .001+-  .001   .041+- .036  Cu
   Zn        .021+-  .001     .046      .020+-  .001   .952+- .093  Zn
   Br  *     .047+-  .003     .102      .052+-  .018  1.114+- .386  Br
   Pb  *     .143+-  .010     .311      .139+-  .020   .974+- .158  Pb
   OC                                 23.510+- 6.631                OC
   EC                                  6.444+- 4.037                EC
---------------------------------------------------------------------
   MASS    46.0  +- 5.1          * FITTING SPECIES
```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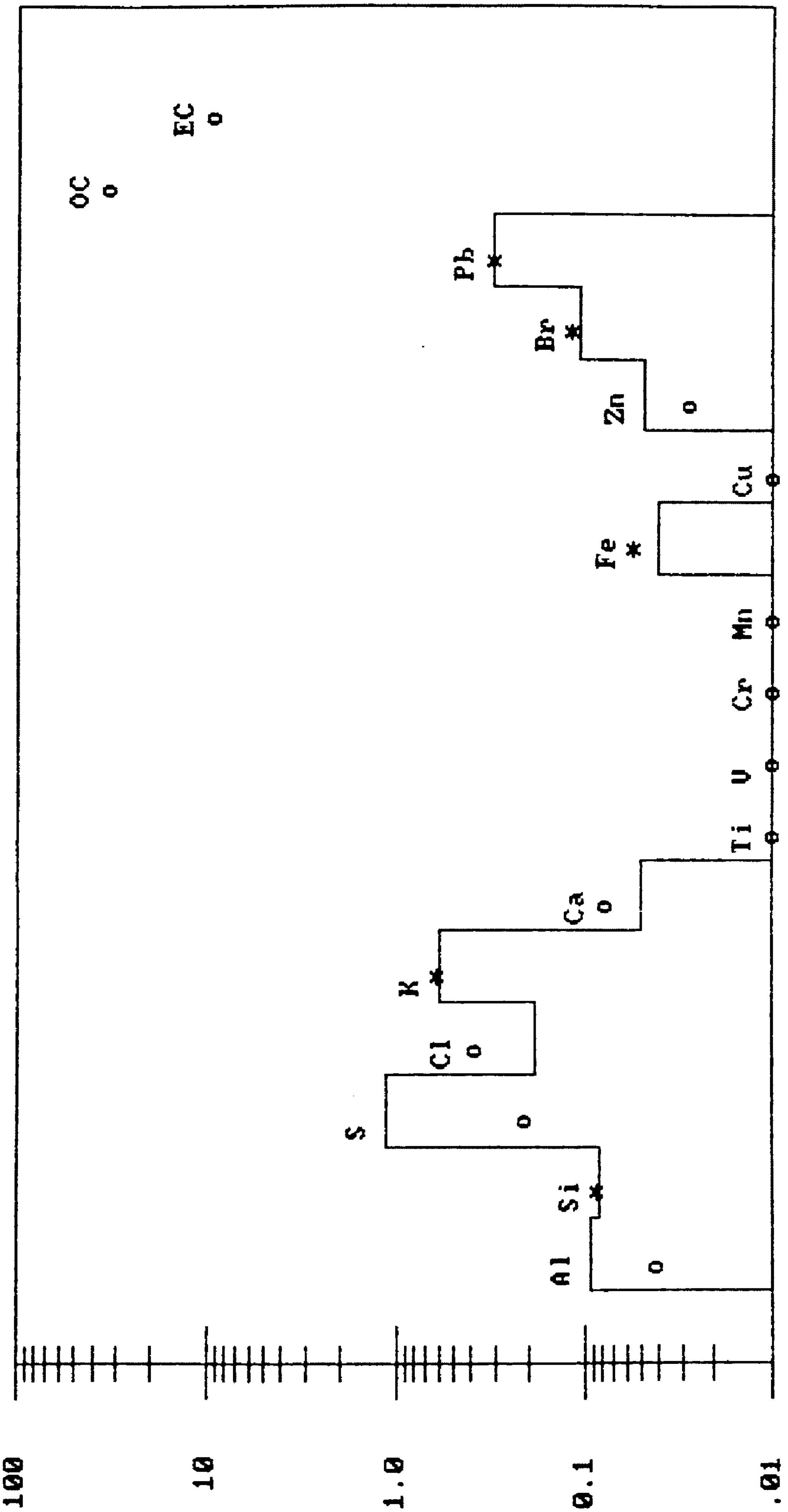
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF32                                 PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 830209  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:      .159  DEGREES OF FREEDOM:   3

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  54.616+- 7.195  105.032+-18.157
          5018  TRANS  F   2.117+-  .317    4.071+-  .761
          ---------------------------------------------
               TOTAL:     56.733+- 7.202  109.103+-18.466

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al  *         <    .067             .014+-  .006   .000+- .000  Al
   Si  *         <    .025             .012+-  .005   .000+- .000  Si
   S       1.050+-   .073   2.019      .182+-  .105   .174+- .101  S
   Cl       .099+-   .007    .190      .328+-  .024  3.311+- .335  Cl
   K   *    .470+-   .033    .904      .470+-  .052   .999+- .132  K
   Ca       .020+-   .001    .038      .061+-  .033  3.068+-1.662  Ca
   Ti            <    .011             .007+-  .008   .000+- .000  Ti
   V             <    .008             .000+-  .001   .000+- .000  V
   Cr            <    .006             .000+-  .001   .000+- .000  Cr
   Mn       .003+-   .002    .006      .000+-  .001   .000+- .182  Mn
   Fe       .025+-   .002    .048      .039+-  .038  1.567+-1.523  Fe
   Cu       .022+-   .001    .042      .000+-  .001   .000+- .025  Cu
   Zn       .030+-   .003    .058      .022+-  .002   .737+- .096  Zn
   Br  *    .098+-   .007    .188      .113+-  .038  1.149+- .400  Br
   Pb  *    .303+-   .021    .583      .292+-  .044   .964+- .160  Pb
   OC                                26.856+- 7.539                OC
   EC                                 7.544+- 4.592                EC
---------------------------------------------------------------------
   MASS     52.0  +- 5.8           * FITTING SPECIES
```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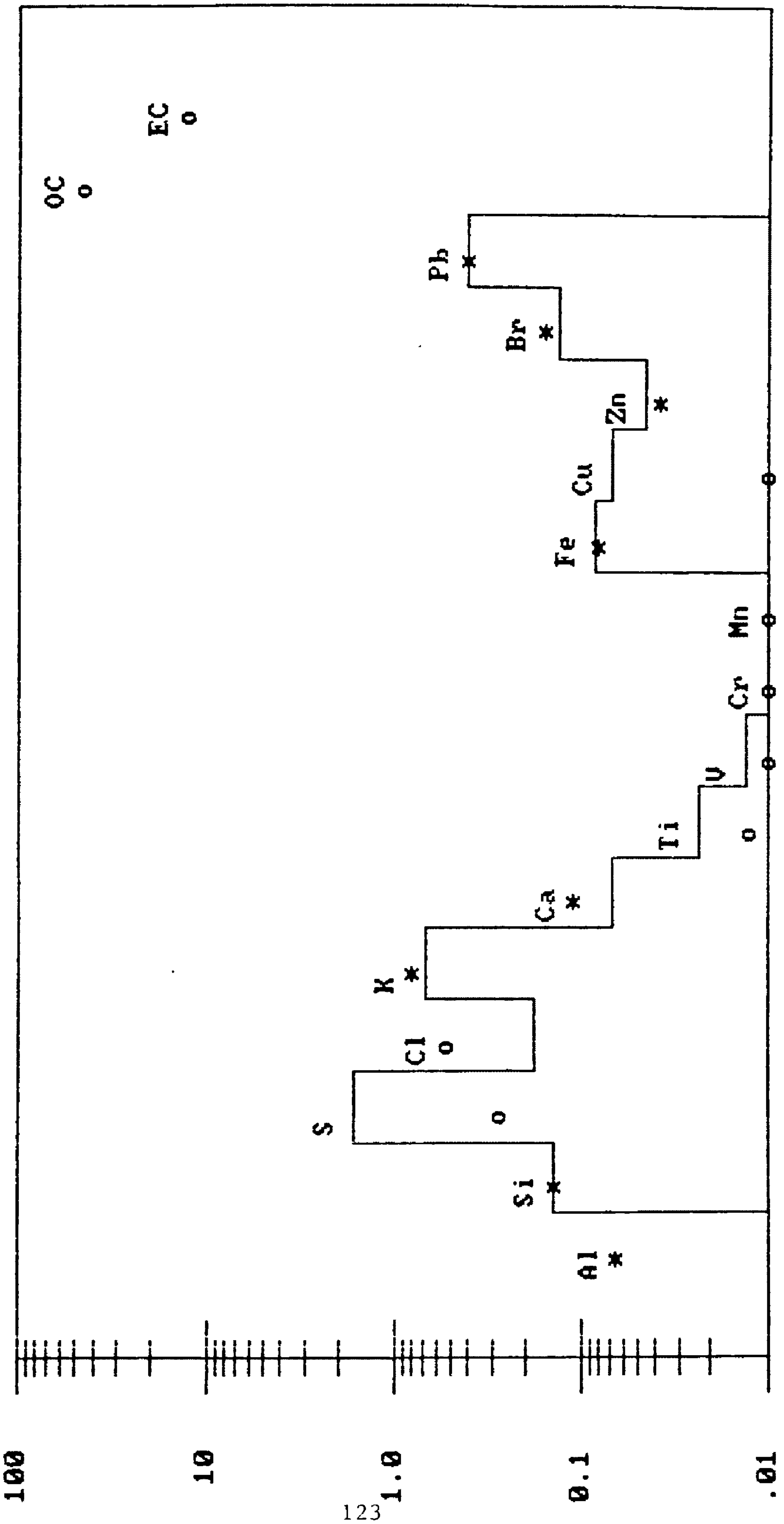
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF36                           PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 830221  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:      .566  DEGREES OF FREEDOM:  7

       --SOURCE----SIZE----UG/M3------------PERCENT---
       5015  WSMOKE F  54.909+- 5.339   87.157+-12.906
       5017  ROADS  F   3.876+-  .450    6.152+-  .991
       5018  TRANS  F   2.200+-  .328    3.492+-  .651
       -----------------------------------------------
           TOTAL:      60.984+- 5.368   96.800+-13.765

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al  *     .442+-  .135    .702       .329+-  .013    .743+- .229  Al
   Si  *    1.000+-  .158   1.587       .935+-  .051    .935+- .156  Si
   S         .411+-  .029    .652       .202+-  .109    .491+- .269  S
   Cl        .154+-  .011    .244       .333+-  .024   2.161+- .221  Cl
   K   *     .497+-  .035    .789       .508+-  .053   1.022+- .129  K
   Ca  *     .098+-  .007    .156       .159+-  .055   1.627+- .570  Ca
   Ti  *     .022+-  .003    .035       .032+-  .009   1.465+- .469  Ti
   V             <   .007               .001+-  .001    .000+- .000  V
   Cr            <   .002               .001+-  .001    .504+- .580  Cr
   Mn  *     .005+-  .001    .008       .004+-  .001    .853+- .221  Mn
   Fe  *     .214+-  .015    .340       .231+-  .042   1.078+- .212  Fe
   Cu        .022+-  .001    .035       .002+-  .001    .070+- .059  Cu
   Zn  *     .025+-  .002    .040       .024+-  .002    .969+- .112  Zn
   Br  *     .108+-  .007    .171       .117+-  .040   1.084+- .376  Br
   Pb  *     .326+-  .023    .517       .309+-  .046    .947+- .155  Pb
   OC                                 27.418+- 7.580                 OC
   EC                                  7.657+- 4.617                 EC
---------------------------------------------------------------------
   MASS     63.0  +- 7.0          * FITTING SPECIES
```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Fe
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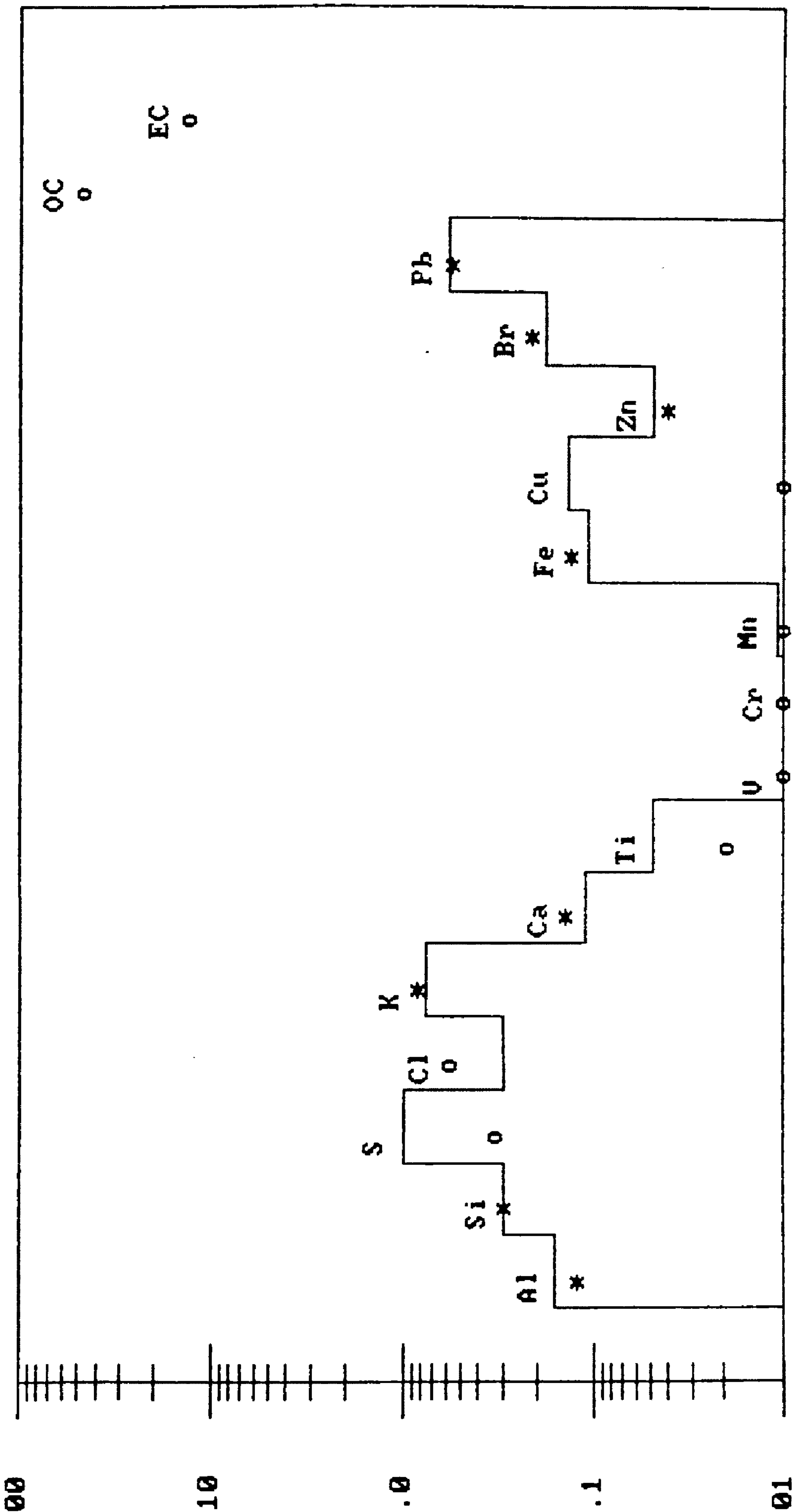
Zn
Br
Pb

```
SAMPLE ID: MMF16                             PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 821223  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .521  DEGREES OF FREEDOM:   5

           --SOURCE-----SIZE----UG/M3------------PERCENT---
           5015  WSMOKE F  17.926+- 1.942   94.346+-14.739
           5017  ROADS  F     .217+-  .037    1.143+-  .234
           5018  TRANS  F     .746+-  .110    3.925+-  .728
           ------------------------------------------------
                 TOTAL:    18.889+- 1.945   99.414+-15.168

---SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .030+-  .015    .158     .022+-  .002   .747+- .382  Al
   Si   *     .056+-  .008    .295     .056+-  .003  1.002+- .162  Si
   S          .191+-  .014   1.005     .063+-  .037   .328+- .196  S
   Cl         .056+-  .004    .295     .109+-  .008  1.944+- .202  Cl
   K    *     .143+-  .010    .753     .156+-  .017  1.092+- .143  K
   Ca   *     .021+-  .001    .111     .026+-  .012  1.246+- .571  Ca
   Ti         .009+-  .003    .047     .004+-  .003   .422+- .340  Ti
   V                  .006             .000+-  .000   .000+- .000  V
   Cr                 .005             .000+-  .000   .000+- .000  Cr
   Mn         .002+-  .001    .011     .000+-  .000   .119+- .108  Mn
   Fe   *     .020+-  .001    .105     .024+-  .013  1.222+- .674  Fe
   Cu         .026+-  .002    .137     .000+-  .000   .003+- .007  Cu
   Zn   *     .009+-  .001    .047     .007+-  .001   .824+- .132  Zn
   Br   *     .034+-  .003    .179     .040+-  .013  1.167+- .407  Br
   Pb   *     .107+-  .007    .563     .103+-  .015   .964+- .159  Pb
   OC                                 8.858+- 2.475                OC
   EC                                 2.492+- 1.507                EC
-----------------------------------------------------------------------
   MASS    19.0  +- 2.1            * FITTING SPECIES
```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Ti
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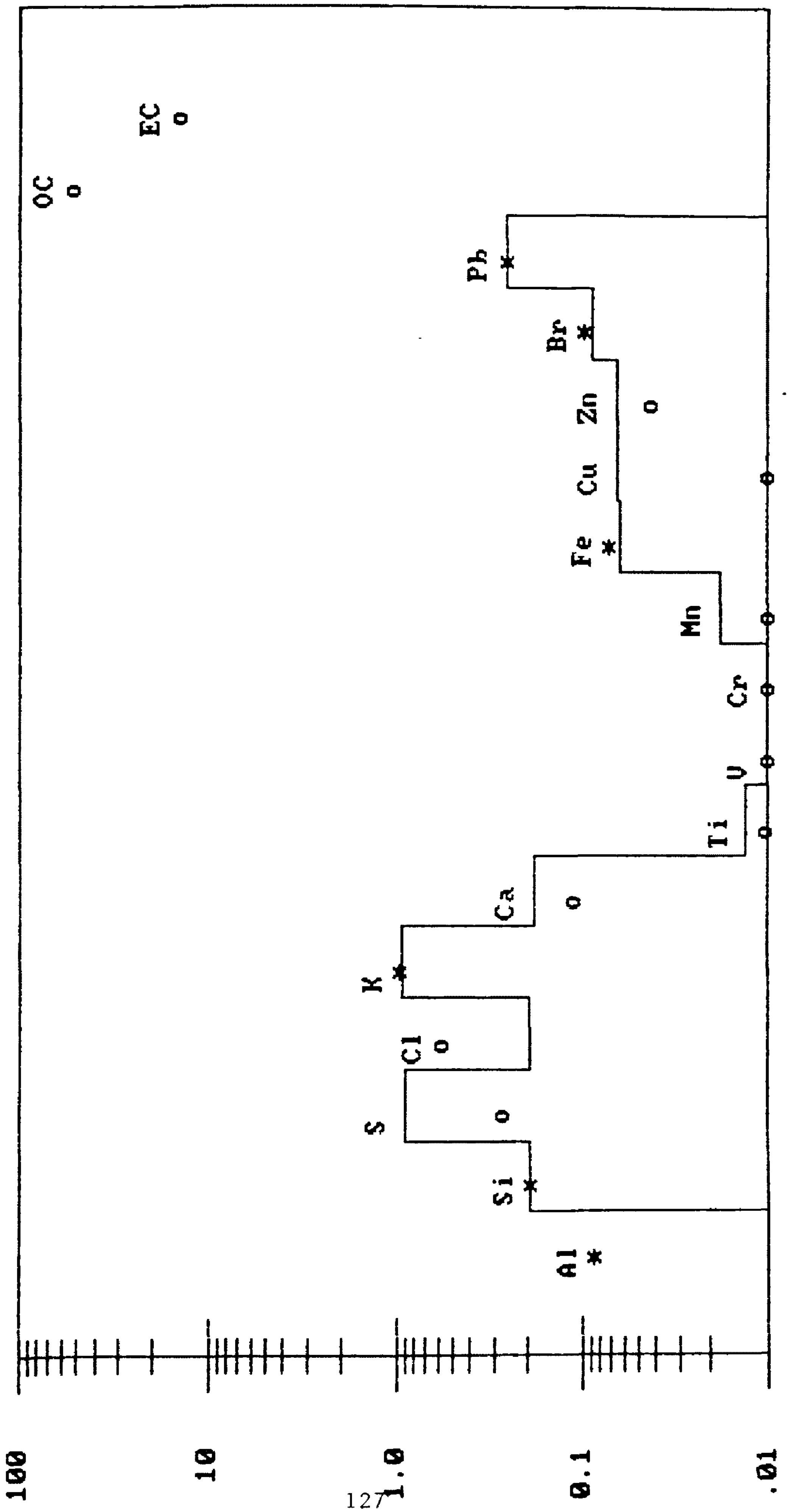
Fe
Cu
Zn
Br
Pb

```
SAMPLE ID: MMF17                              PARTICLE SIZE: FINE
FIELD FLAG:    MASS FLAG:    ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821226  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .143  DEGREES OF FREEDOM:  3

         --SOURCE----SIZE----UG/M3------------PERCENT---
         5015  WSMOKE F  41.844+- 5.536  107.291+-18.611
         5017  ROADS  F     .285+-  .051      .731+-  .154
         5018  TRANS  F     .691+-  .103     1.773+-  .331
         -----------------------------------------------
              TOTAL:      42.820+- 5.537  109.795+-18.796

---SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.-------
   Al   *          <   .059              .033+-   .004    .000+-  .000   Al
   Si   *     .073+-   .012     .187     .072+-   .004    .986+-  .166   Si
   S          .355+-   .025     .910     .104+-   .035    .294+-  .101   S
   Cl         .075+-   .005     .192     .229+-   .014   3.058+-  .292   Cl
   K    *     .363+-   .026     .931     .362+-   .040    .999+-  .131   K
   Ca         .069+-   .005     .177     .043+-   .011    .627+-  .171   Ca
   Ti         .005+-   .004     .013     .004+-   .003    .813+-  .835   Ti
   V               <   .007              .000+-   .000    .000+-  .000   V
   Cr              <   .006              .000+-   .000    .000+-  .000   Cr
   Mn         .007+-   .002     .018     .000+-   .000    .045+-  .061   Mn
   Fe   *     .024+-   .002     .062     .027+-   .012   1.115+-  .529   Fe
   Cu         .025+-   .002     .064     .000+-   .000    .005+-  .017   Cu
   Zn         .025+-   .002     .064     .016+-   .001    .650+-  .060   Zn
   Br   *     .034+-   .003     .087     .037+-   .013   1.082+-  .378   Br
   Pb   *     .098+-   .007     .251     .096+-   .014    .977+-  .162   Pb
   OC                                  20.202+- 5.775                    OC
   EC                                   5.541+- 3.515                    EC
-----------------------------------------------------------------------
   MASS     39.0  +- 4.4             * FITTING SPECIES
```



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Zn
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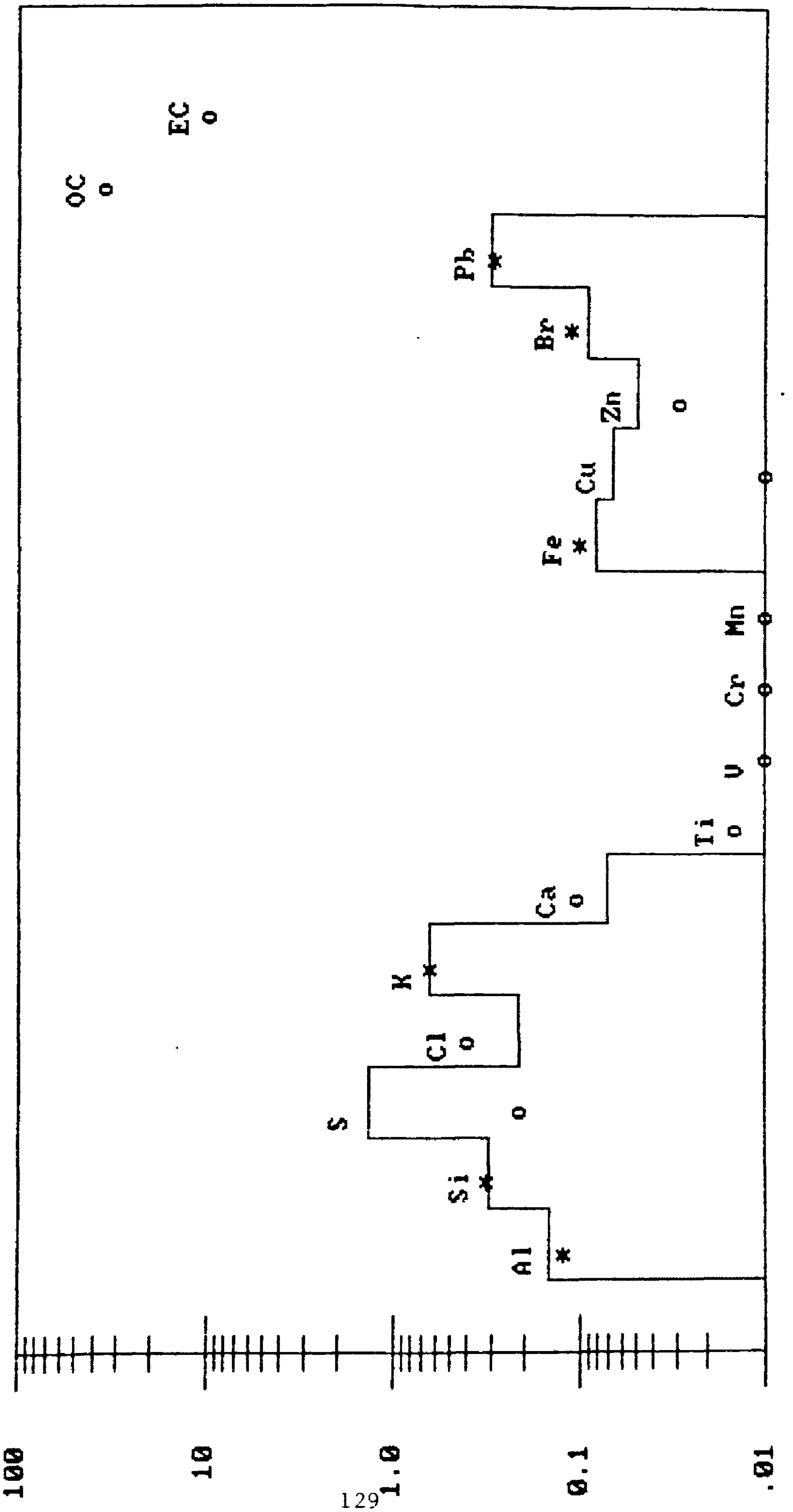
EC

```
SAMPLE ID: MMF15                                  PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821220  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:   1.090  DEGREES OF FREEDOM:  5

         --SOURCE----SIZE----UG/M3------------PERCENT---
         5015  WSMOKE F  35.181+- 3.271   92.582+-13.448
         5017  ROADS  F    .193+-  .038     .509+-  .116
         5018  TRANS  F   1.098+-  .163    2.891+-  .536
         -----------------------------------------------
              TOTAL:     36.473+- 3.275   95.982+-13.749

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al  *          <  .059               .025+-  .004   .000+- .000  Al
   Si  *     .053+-  .008    .139       .053+-  .004   .991+- .171  Si
   S         .636+-  .045   1.674       .108+-  .055   .170+- .087  S
   Cl        .067+-  .004    .176       .205+-  .014  3.059+- .292  Cl
   K   *     .255+-  .018    .671       .304+-  .034  1.193+- .157  K
   Ca  *     .026+-  .002    .068       .041+-  .017  1.587+- .672  Ca
   Ti        .009+-  .004    .024       .005+-  .004   .530+- .512  Ti
   V         .005+-  .002    .013       .000+-  .000   .009+- .071  V
   Cr        .003+-  .002    .008       .000+-  .000   .017+- .118  Cr
   Mn        .002+-  .001    .005       .000+-  .000   .106+- .184  Mn
   Fe  *     .032+-  .003    .084       .030+-  .020   .931+- .620  Fe
   Cu        .026+-  .002    .068       .000+-  .000   .003+- .014  Cu
   Zn  *     .017+-  .001    .045       .014+-  .001   .830+- .097  Zn
   Br  *     .049+-  .003    .129       .058+-  .020  1.193+- .413  Br
   Pb  *     .156+-  .011    .411       .152+-  .023   .973+- .162  Pb
   OC                                 17.204+- 4.856                OC
   EC                                  4.793+- 2.957                EC
---------------------------------------------------------------------
   MASS     38.0  +- 4.2          * FITTING SPECIES
```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Br
Pb
OC
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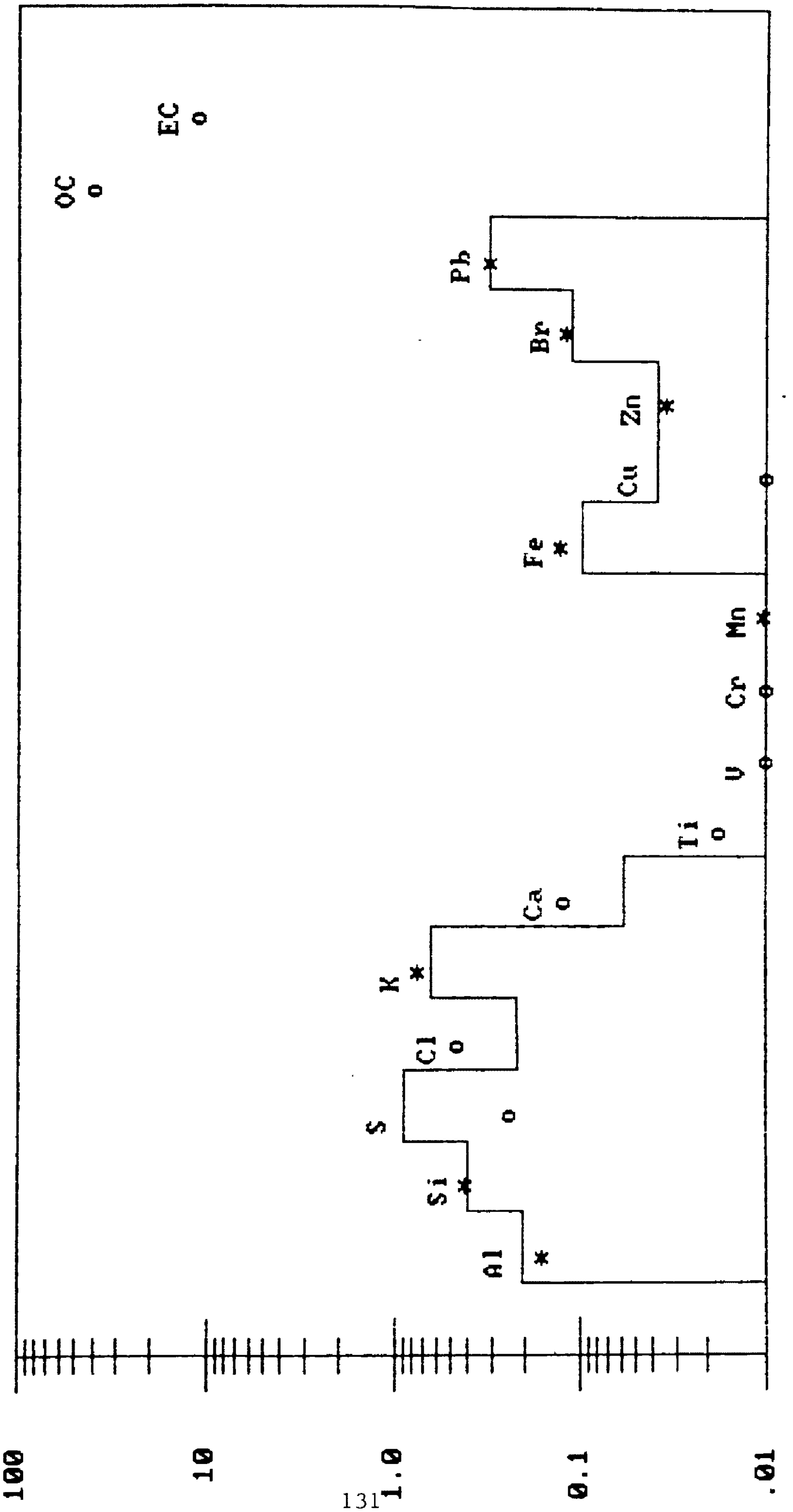
```
SAMPLE ID: MMF05                                 PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 821120  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:    1.194  DEGREES OF FREEDOM:  6

          --SOURCE-----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  12.493+- 1.475   78.081+-12.646
          5017  ROADS  F     .889+-  .121    5.559+-  .978
          5018  TRANS  F     .649+-  .097    4.056+-  .757
          ----------------------------------------------
               TOTAL:     14.031+- 1.483   87.696+-13.433

--SPECIES----MEAS. UG/M3------%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .086+-  .026     .538      .076+-  .003    .879+- .272  Al
   Si   *     .226+-  .036    1.413      .216+-  .012    .954+- .160  Si
   S          .103+-  .007     .644      .052+-  .032    .502+- .315  S
   Cl         .034+-  .003     .212      .079+-  .006   2.329+- .260  Cl
   K    *     .104+-  .007     .650      .116+-  .012   1.112+- .139  K
   Ca   *     .026+-  .002     .163      .038+-  .014   1.469+- .548  Ca
   Ti         .015+-  .003     .094      .008+-  .003    .524+- .204  Ti
   V          .003+-  .002     .019      .000+-  .000    .068+- .064  V
   Cr         .003+-  .002     .019      .000+-  .000    .077+- .067  Cr
   Mn   *     .002+-  .001     .013      .001+-  .000    .489+- .258  Mn
   Fe   *     .047+-  .003     .294      .056+-  .012   1.183+- .270  Fe
   Cu         .010+-  .001     .063      .000+-  .000    .036+- .030  Cu
   Zn   *     .008+-  .001     .050      .006+-  .001    .706+- .125  Zn
   Br   *     .027+-  .002     .169      .035+-  .012   1.279+- .442  Br
   Pb   *     .096+-  .007     .600      .091+-  .013    .945+- .156  Pb
   OC                                   6.303+- 1.725                 OC
   EC                                   1.781+- 1.051                 EC
---------------------------------------------------------------------
   MASS     16.0  +- 1.8           * FITTING SPECIES
```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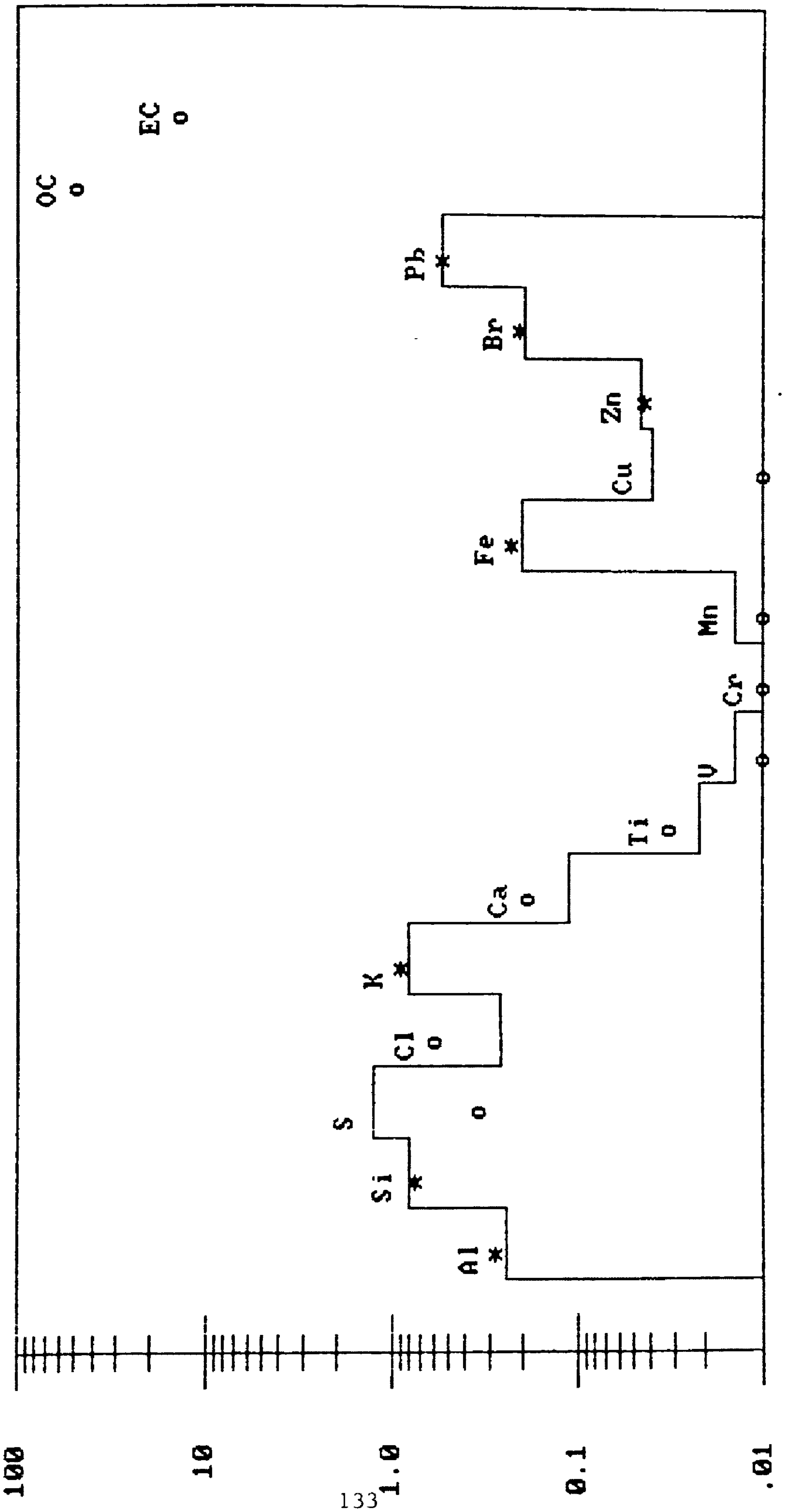
```
SAMPLE ID: MMF06                               PARTICLE SIZE: FINE
FIELD FLAG:    MASS FLAG:    ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821123  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .162  DEGREES OF FREEDOM:  5

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  25.775+- 3.433   99.135+-17.204
          5017  ROADS  F    .869+-  .113    3.343+-  .571
          5018  TRANS  F    .620+-  .092    2.386+-  .442
          -----------------------------------------------
               TOTAL:     27.265+- 3.437  104.865+-17.628

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .069+-  .021     .265      .077+-  .004  1.112+- .347   Al
   Si   *     .216+-  .034     .831      .211+-  .011   .975+- .161   Si
   S          .412+-  .029    1.585      .075+-  .031   .181+- .077   S
   Cl         .051+-  .004     .196      .146+-  .009  2.865+- .287   Cl
   K    *     .232+-  .017     .892      .230+-  .025   .990+- .128   K
   Ca   *     .041+-  .003     .158      .046+-  .014  1.128+- .342   Ca
   Ti   *     .010+-  .003     .038      .008+-  .003   .764+- .341   Ti
   V          .005+-  .002     .019      .000+-  .000   .040+- .055   V
   Cr            <    .005               .000+-  .000   .000+- .000   Cr
   Mn         .007+-  .001     .027      .001+-  .000   .137+- .044   Mn
   Fe   *     .054+-  .004     .208      .054+-  .012  1.001+- .228   Fe
   Cu         .015+-  .001     .058      .000+-  .000   .023+- .025   Cu
   Zn         .014+-  .001     .054      .011+-  .001   .752+- .086   Zn
   Br   *     .029+-  .002     .112      .033+-  .011  1.138+- .393   Br
   Pb   *     .089+-  .006     .342      .087+-  .013   .975+- .159   Pb
   OC                                  12.598+- 3.557                 OC
   EC                                   3.473+- 2.166                 EC
----------------------------------------------------------------------
   MASS     26.0  +- 2.9            * FITTING SPECIES
```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Br
Pb
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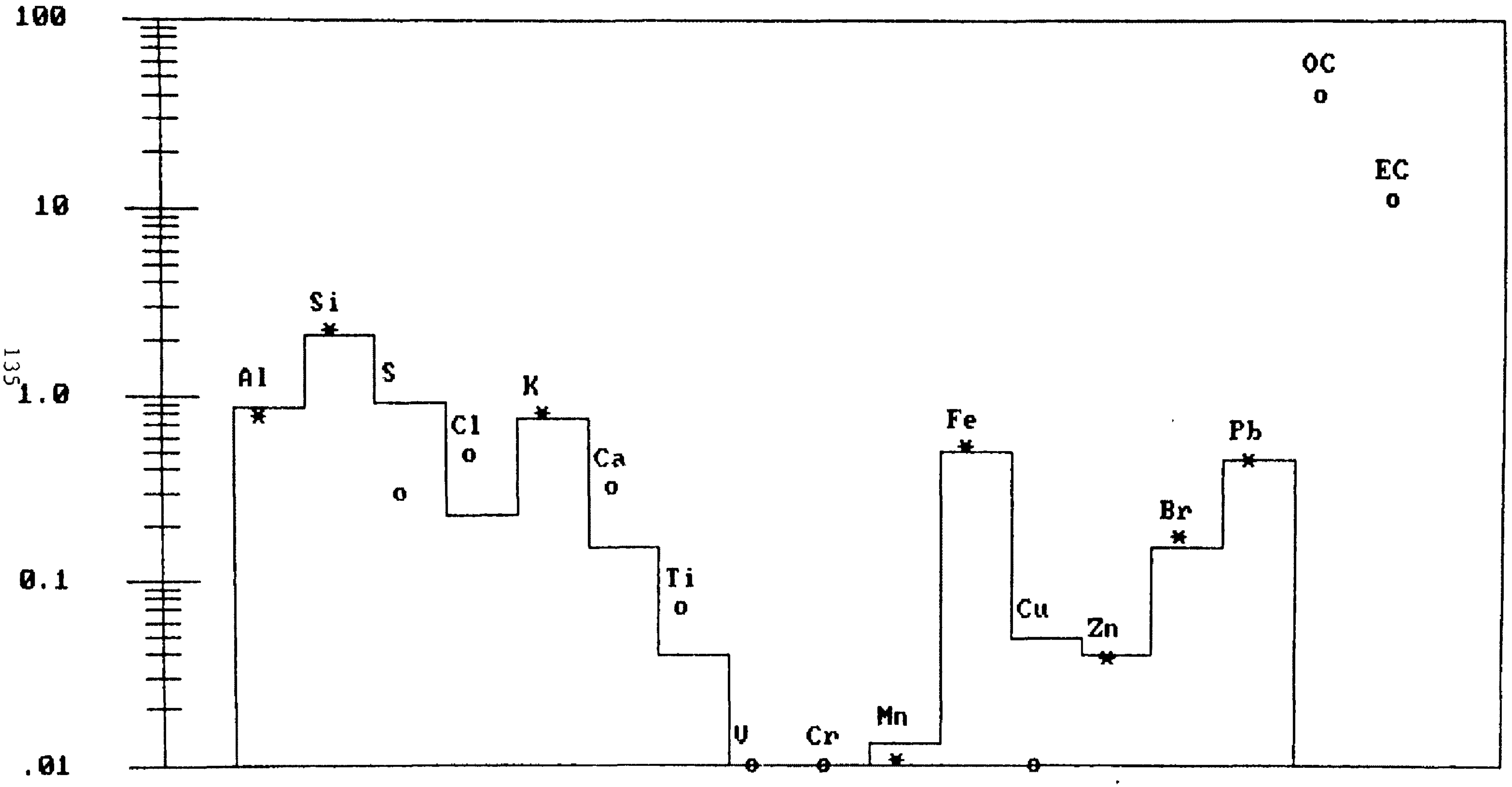
EC

```
SAMPLE ID: MMF07                                PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821126  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .122  DEGREES OF FREEDOM:   3

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  56.505+- 7.547   70.631+-12.303
          5017  ROADS  F   1.906+-  .274    2.382+-  .434
          5018  TRANS  F   1.837+-  .274    2.297+-  .428
          -------------------------------------------------
               TOTAL:     60.248+- 7.557   75.310+-12.654

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al   *     .157+-  .048     .196       .169+-  .008  1.075+- .331  Al
   Si   *     .490+-  .077     .613       .464+-  .025   .948+- .158  Si
   S          .801+-  .057    1.001       .182+-  .092   .228+- .115  S
   Cl         .169+-  .012     .211       .332+-  .023  1.962+- .190  Cl
   K    *     .504+-  .035     .630       .503+-  .054   .999+- .129  K
   Ca         .067+-  .004     .084       .107+-  .036  1.597+- .541  Ca
   Ti         .010+-  .004     .013       .018+-  .007  1.827+-1.025  Ti
   V            <     .008                .000+-  .001   .000+- .000  V
   Cr         .003+-  .002     .004       .000+-  .001   .165+- .220  Cr
   Mn         .006+-  .002     .007       .002+-  .001   .349+- .154  Mn
   Fe   *     .115+-  .008     .144       .127+-  .034  1.108+- .304  Fe
   Cu         .009+-  .001     .011       .001+-  .001   .085+- .090  Cu
   Zn         .035+-  .003     .044       .024+-  .002   .672+- .078  Zn
   Br   *     .090+-  .007     .113       .098+-  .033  1.086+- .378  Br
   Pb   *     .263+-  .018     .329       .256+-  .038   .973+- .160  Pb
   OC                                   27.823+- 7.799                OC
   EC                                    7.739+- 4.750                EC
----------------------------------------------------------------------
   MASS     80.0   +- 8.9            * FITTING SPECIES
```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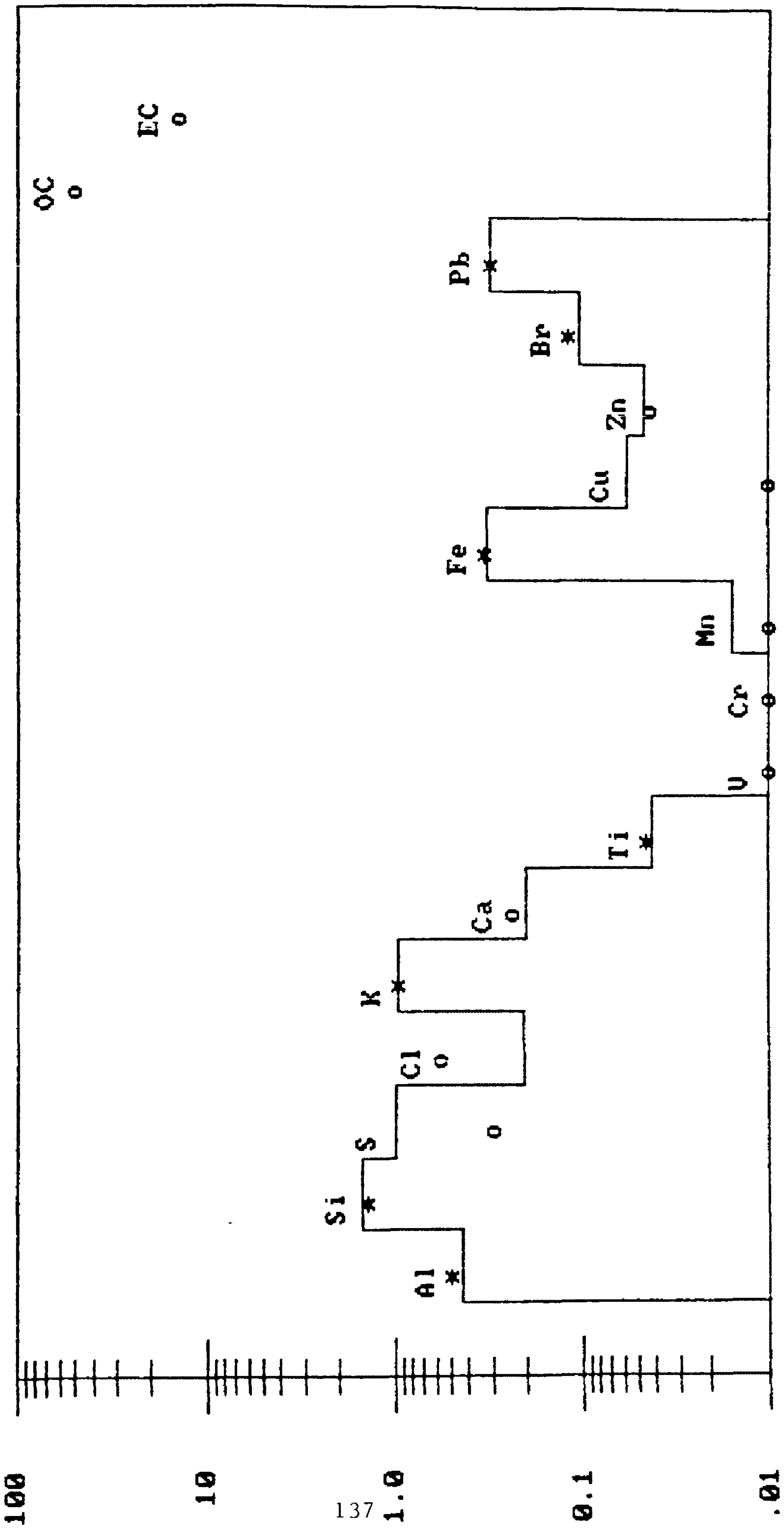
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF09                                PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821202  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .391  DEGREES OF FREEDOM:  5

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  28.429+- 3.817   94.764+-16.557
          5017  ROADS  F   1.435+-  .186    4.783+-  .818
          5018  TRANS  F    .946+-  .142    3.153+-  .589
          -----------------------------------------------
               TOTAL:     30.810+- 3.824  102.700+-17.156

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al   *      .144+-  .044     .480      .124+-  .005   .858+- .262  Al
   Si   *      .355+-  .056    1.183      .347+-  .019   .978+- .163  Si
   S           .384+-  .027    1.280      .095+-  .047   .246+- .124  S
   Cl          .079+-  .006     .263      .168+-  .011  2.120+- .210  Cl
   K    *      .261+-  .018     .870      .258+-  .027   .987+- .126  K
   Ca   *      .046+-  .003     .153      .066+-  .022  1.435+- .479  Ca
   Ti   *      .015+-  .003     .050      .012+-  .004   .824+- .308  Ti
   V           .006+-  .002     .020      .000+-  .000   .055+- .053  V
   Cr             <    .002               .000+-  .000   .187+- .237  Cr
   Mn          .006+-  .001     .020      .002+-  .000   .263+- .070  Mn
   Fe   *      .080+-  .006     .267      .088+-  .018  1.098+- .237  Fe
   Cu          .016+-  .001     .053      .001+-  .001   .036+- .032  Cu
   Zn          .020+-  .001     .067      .012+-  .001   .604+- .068  Zn
   Br   *      .041+-  .003     .137      .050+-  .017  1.227+- .426  Br
   Pb   *      .140+-  .010     .467      .132+-  .020   .946+- .155  Pb
   OC                                   14.055+- 3.924                OC
   EC                                    3.906+- 2.390                EC
----------------------------------------------------------------------
   MASS     30.0  +- 3.4            * FITTING SPECIES
```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Ti
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Cr
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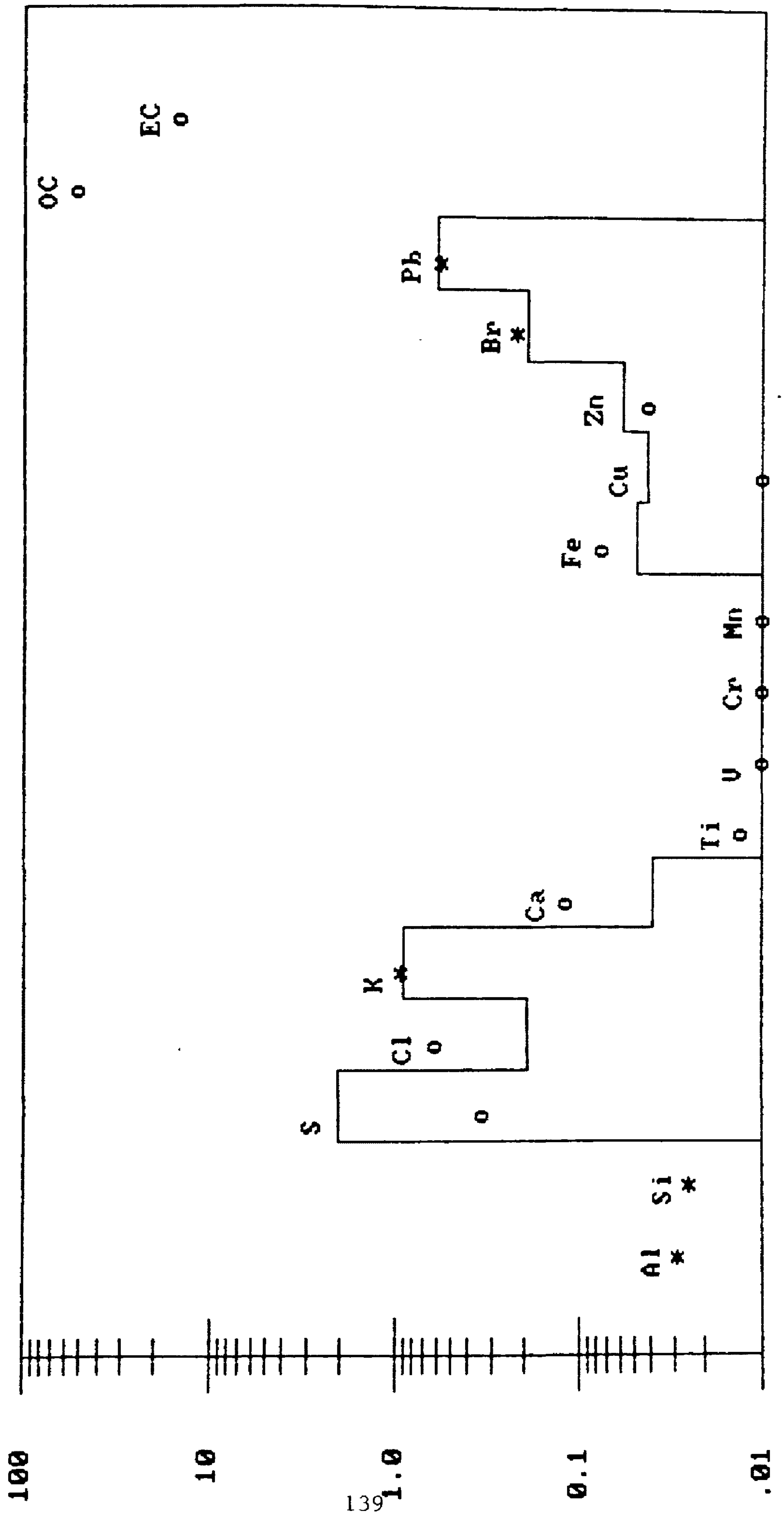
Cu
Zn
Br
Pb

```
SAMPLE ID: MMF10                              PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821205  START TIME:   .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:    .392  DEGREES OF FREEDOM:  5

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  52.606+- 6.954   90.700+-15.696
          5017  ROADS  F   1.241+-  .182    2.139+-  .394
          5018  TRANS  F   1.317+-  .197    2.272+-  .423
          -----------------------------------------------
               TOTAL:     55.164+- 6.959   95.111+-16.024

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al   *      .151+-  .046    .260      .113+-  .007   .751+- .231  Al
   Si   *      .307+-  .048    .529      .303+-  .016   .987+- .165  Si
   S           .478+-  .034    .824      .152+-  .066   .319+- .140  S
   Cl          .161+-  .011    .278      .299+-  .019  1.859+- .177  Cl
   K    *      .474+-  .034    .817      .464+-  .051   .979+- .127  K
   Ca   *      .057+-  .004    .098      .082+-  .025  1.433+- .446  Ca
   Ti   *      .013+-  .003    .022      .012+-  .005   .945+- .451  Ti
   V                <  .007              .000+-  .001   .000+- .000  V
   Cr          .003+-  .002    .005      .000+-  .001   .108+- .190  Cr
   Mn          .003+-  .001    .005      .001+-  .001   .455+- .237  Mn
   Fe   *      .076+-  .006    .131      .085+-  .024  1.121+- .327  Fe
   Cu          .013+-  .001    .022      .000+-  .001   .038+- .050  Cu
   Zn          .031+-  .003    .053      .021+-  .002   .686+- .077  Zn
   Br   *      .064+-  .004    .110      .070+-  .024  1.095+- .380  Br
   Pb   *      .192+-  .014    .331      .183+-  .027   .955+- .158  Pb
   OC                                  25.680+- 7.261                OC
   EC                                   7.095+- 4.421                EC
----------------------------------------------------------------------
   MASS    58.0  +- 6.5           * FITTING SPECIES
```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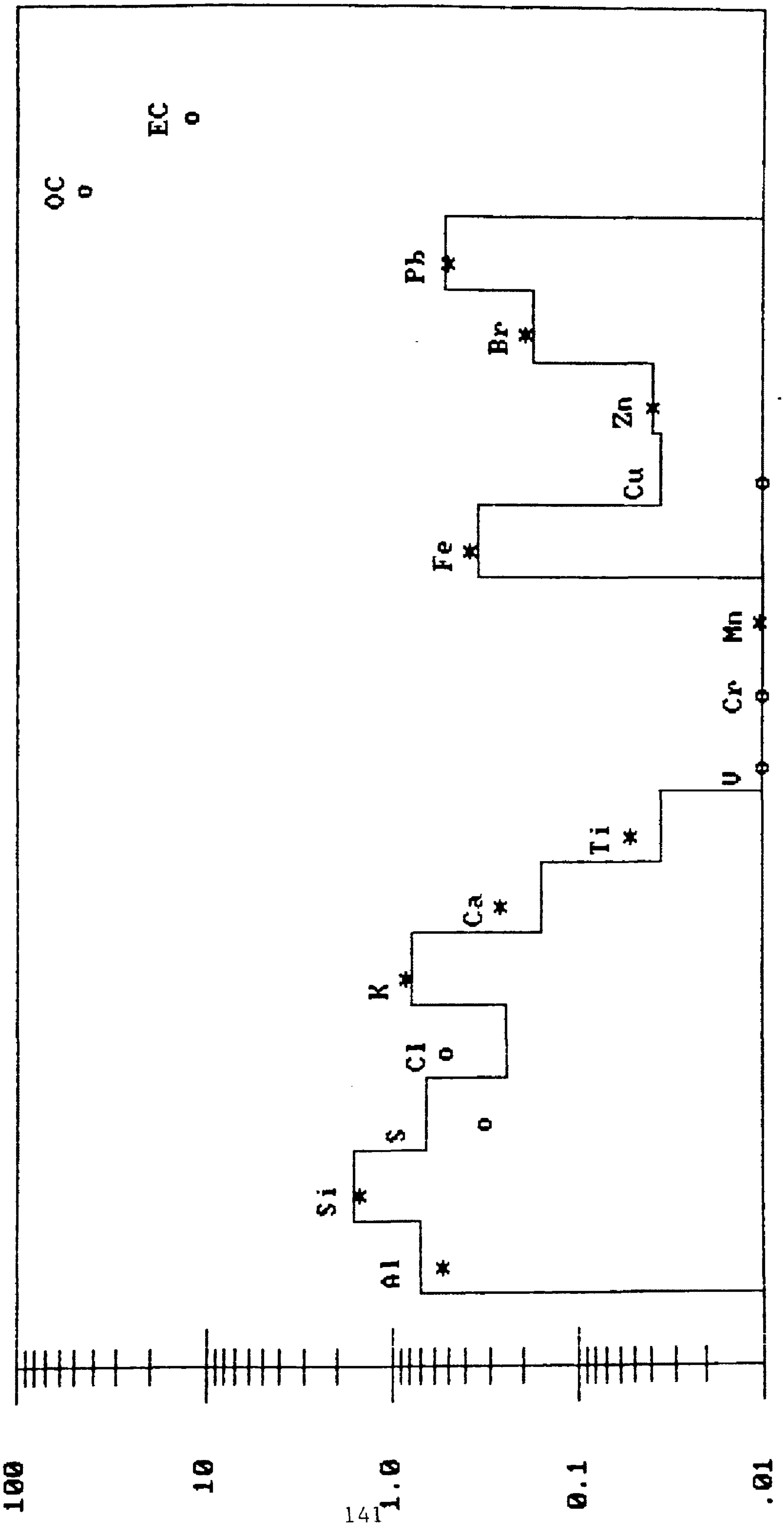
V
Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE  ID: MMF11                           PARTICLE SIZE: FINE
FIELD FLAG:    MASS FLAG:    ANALYSIS FLAGS:
SITE: 20   ose Lawn Park
SAMPLE DATE: 821208  START TIME:    .0  DURATION:  24.0 HOURS
REDUCED CHI SQUARE:     .369  DEGREES OF FREEDOM:  3

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  32.251+- 4.186   87.165+-14.944
          5017  ROADS  F     .633+-  .100    1.711+-  .330
          5018  TRANS  F     .724+-  .108    1.956+-  .364
          -----------------------------------------------
               TOTAL:     33.608+- 4.189   90.832+-15.220

--SPECIES----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al         .091+-  .027    .246      .059+-  .004    .649+- .200  Al
   Si   *     .156+-  .024    .422      .155+-  .008    .993+- .164  Si
   S          .756+-  .054   2.043      .090+-  .036    .119+- .049  S
   Cl         .082+-  .006    .222      .181+-  .011   2.212+- .209  Cl
   K    *     .276+-  .020    .746      .283+-  .031   1.026+- .134  K
   Ca   *     .057+-  .004    .154      .046+-  .013    .805+- .241  Ca
   Ti             <   .009              .006+-  .003    .000+- .000  Ti
   V              <   .006              .000+-  .000    .000+- .000  V
   Cr             <   .005              .000+-  .000    .000+- .000  Cr
   Mn         .002+-  .001    .005      .001+-  .000    .348+- .240  Mn
   Fe   *     .038+-  .003    .103      .044+-  .013   1.169+- .358  Fe
   Cu         .008+-  .001    .022      .000+-  .000    .032+- .047  Cu
   Zn         .020+-  .001    .054      .013+-  .001    .645+- .066  Zn
   Br   *     .033+-  .003    .089      .038+-  .013   1.167+- .407  Br
   Pb   *     .104+-  .007    .281      .101+-  .015    .968+- .159  Pb
   OC                                 15.695+- 4.451                 OC
   EC                                  4.326+- 2.710                 EC
-------------------------------------------------------------------
   MASS     37.0  +- 4.1           * FITTING SPECIES
```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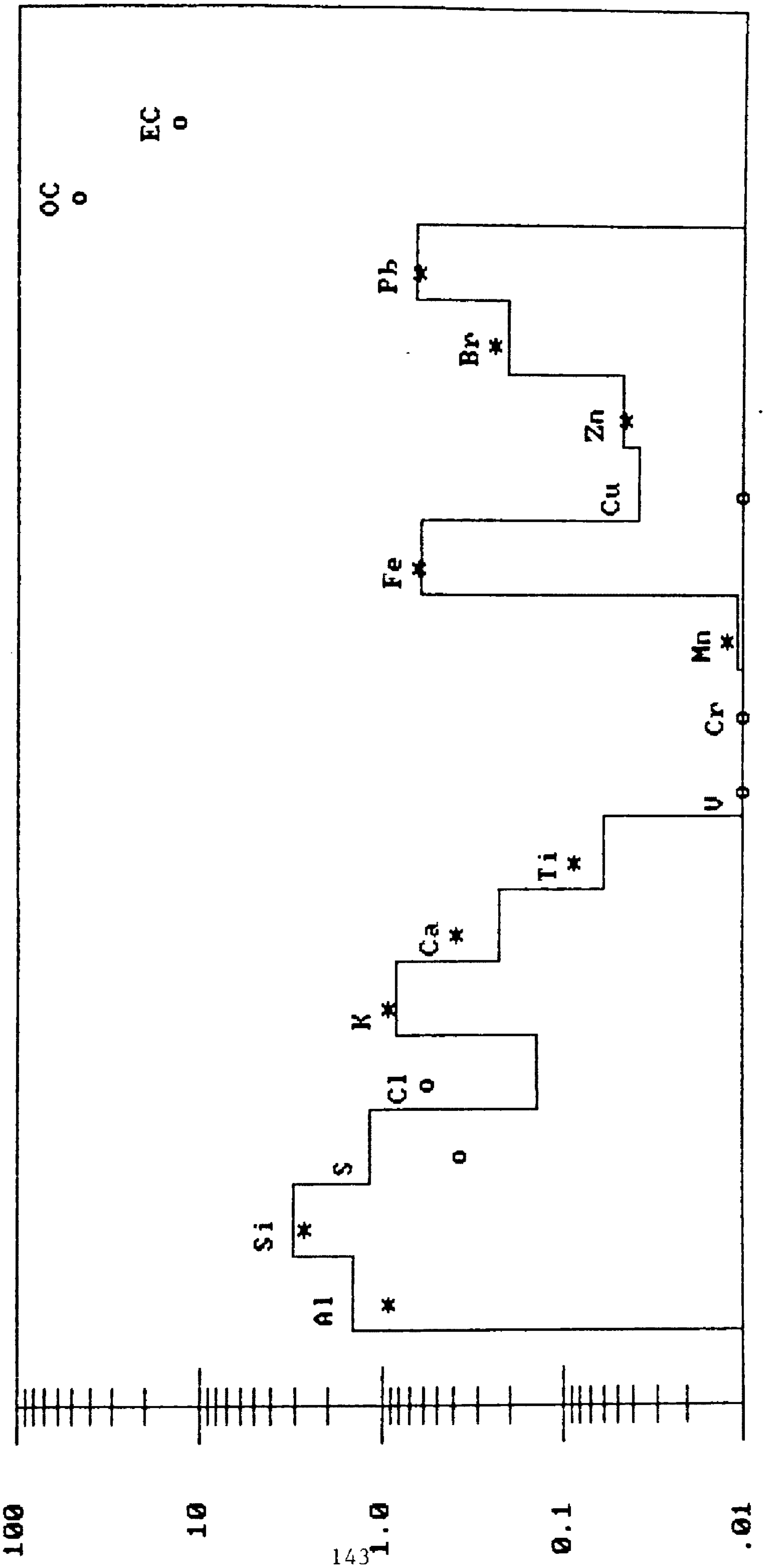
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF12                                  PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 821211  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .043  DEGREES OF FREEDOM:   2

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  78.372+-10.369  105.908+-18.359
          5018  TRANS  F   1.644+-  .245    2.222+-  .414
          ---------------------------------------------
               TOTAL:     80.016+-10,372  108.130+-18.524

--SPECIES----MEAS. UG/M3-----%--------CALC. UG/M3---CALC./MEAS.------
   Al         .108+-  .032    .146    .019+-  .008   .174+- .092  Al
   Si         .144+-  .023    .195    .010+-  .004   .067+- .028  Si
   S         1.640+-  .115   2.216    .207+-  .083   .126+- .051  S
   Cl         .136+-  .010    .184    .438+-  .027  3.217+- .307  Cl
   K    *     .674+-  .048    .911    .674+-  .075  1.000+- .132  K
   Ca         .053+-  .004    .072    .072+-  .026  1.354+- .499  Ca
   Ti         .004+-  .003    .005    .005+-  .006  1.315+-1.824  Ti
   V              <   .007            .000+-  .001   .000+- .000  V
   Cr             <   .002            .000+-  .001   .000+- .392  Cr
   Mn         .006+-  .001    .008    .000+-  .001   .000+- .131  Mn
   Fe   *     .028+-  .002    .038    .030+-  .029  1.086+-1.056  Fe
   Cu         .005+-  .001    .007    .000+-  .001   .000+- .157  Cu
   Zn         .050+-  .004    .068    .030+-  .002   .610+- .065  Zn
   Br   *     .080+-  .006    .108    .087+-  .030  1.093+- .380  Br
   Pb   *     .232+-  .017    .314    .227+-  .034   .978+- .163  Pb
   OC                               37.936+-10.816                OC
   EC                               10.461+- 6.585                EC
---------------------------------------------------------------------
   MASS    74.0  +-  8.3          * FITTING SPECIES
```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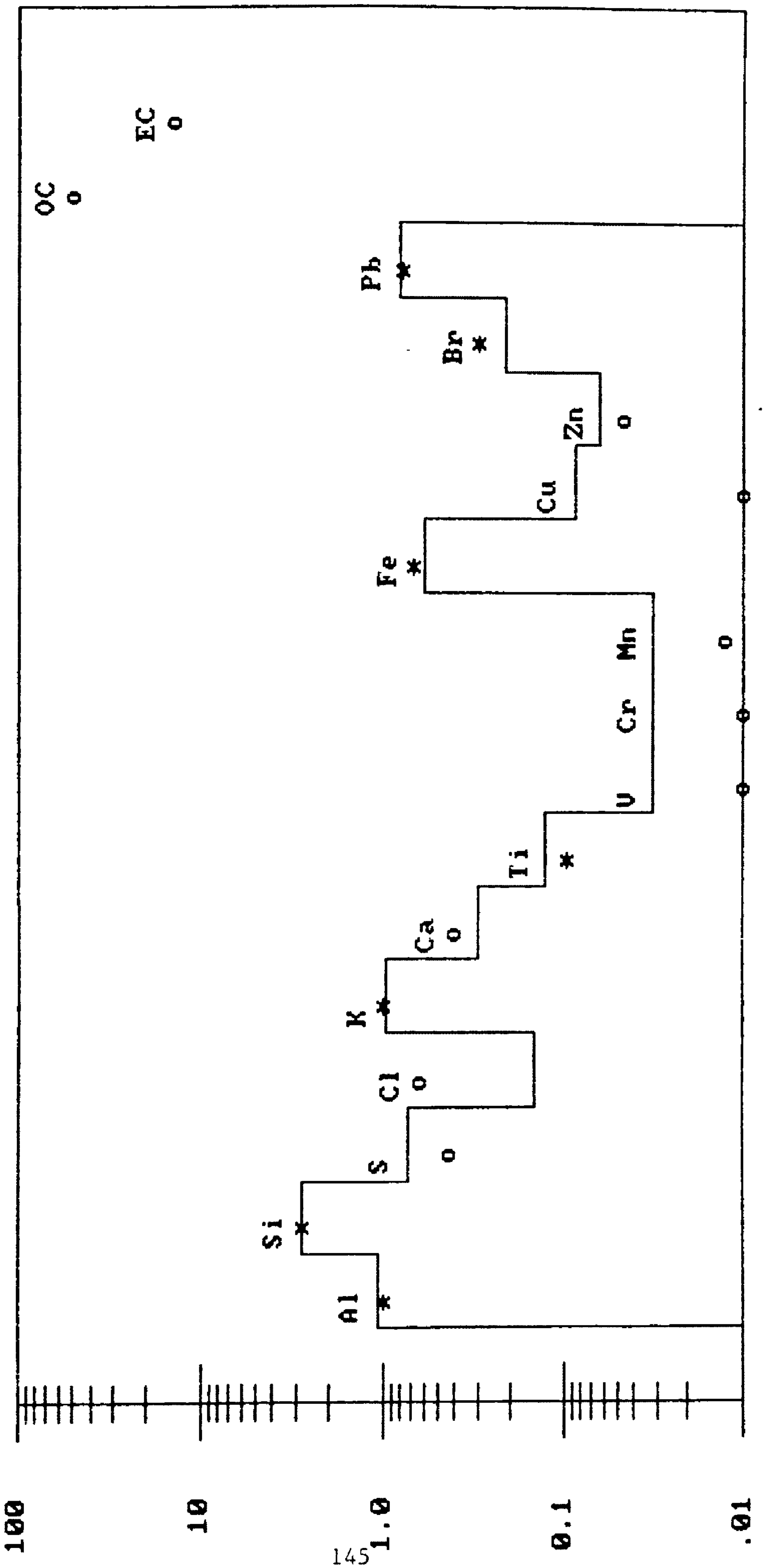
Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

```
SAMPLE ID: MMF13                                PARTICLE SIZE: FINE
FIELD FLAG:     MASS FLAG:     ANALYSIS FLAGS:
SITE: 20    ose Lawn Park
SAMPLE DATE: 821214  START TIME:    .0  DURATION:   24.0 HOURS
REDUCED CHI SQUARE:     .119  DEGREES OF FREEDOM:  2

          --SOURCE----SIZE----UG/M3------------PERCENT---
          5015  WSMOKE F  67.131+- 8.883   69.207+-11.992
          5017  ROADS  F     .291+-  .059     .300+-  .070
          5018  TRANS  F   2.098+-  .312    2.163+-  .403
          -----------------------------------------------
               TOTAL:     69.520+- 8.888   71.670+-12.176

--SPECIES-----MEAS. UG/M3-----%---------CALC. UG/M3---CALC./MEAS.------
   Al          .091+-  .027     .094       .041+-  .007   .446+- .156  Al
   Si   *      .082+-  .013     .085       .082+-  .006   .994+- .170  Si
   S          1.120+-  .082    1.155       .206+-  .105   .184+- .094  S
   Cl          .177+-  .013     .182       .391+-  .026  2.210+- .217  Cl
   K    *      .580+-  .041     .598       .580+-  .064  1.000+- .132  K
   Ca          .049+-  .003     .051       .077+-  .033  1.567+- .679  Ca
   Ti          .004+-  .003     .004       .009+-  .008  2.151+-2.533  Ti
   V           .003+-  .002     .003       .000+-  .001   .022+- .224  V
   Cr                  .005                .000+-  .001   .000+- .000  Cr
   Mn          .007+-  .001     .007       .000+-  .001   .046+- .096  Mn
   Fe   *      .039+-  .003     .040       .053+-  .038  1.361+- .969  Fe
   Cu          .005+-  .001     .005       .000+-  .001   .023+- .136  Cu
   Zn          .047+-  .003     .048       .027+-  .002   .572+- .063  Zn
   Br   *      .102+-  .007     .105       .112+-  .038  1.094+- .380  Br
   Pb   *      .299+-  .021     .308       .290+-  .043   .970+- .161  Pb
   OC                                    32.821+- 9.266                OC
   EC                                     9.145+- 5.643                EC
-----------------------------------------------------------------------
   MASS     97.0   +-10.9            * FITTING SPECIES
```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Ca
Ti
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Cr
Mn
Fe
Cu
Zn
Br
Pb
OC
EC

APPENDIX C

ELEMENTAL AND ORGANIC RAW DATA

Including Tables:

1C - 1980 XRF Analysis Results

2C - 1982-83 XRF Analysis Results

3C - Monthly Averages Of 1982-83 XRF Results

4C - Winter 1982-83 Organic Composition

TABLE 1C

1980 XRF Analysis Results: Fine and Coarse Concentrations (ng/m^3)

	02/07/80						02/18/80						02/19/80						
	F			C			F			C			F			C			
Al	197	±	25				241	±	30	0	±	0	329	±	40	1520	±	172	Al
Si	251		30				188		23	501		57	558		64	6141		688	Si
S	966		117				1552		183	0		0	689		86	0		0	S
Cl	74		14				48		12	0		0	85		15	199		26	Cl
K	209		25				215		26	0		0	296		35	432		50	K
Ca	47		7				47		7	66		8	81		11	346		40	Ca
Ti	9		2				7		2	9		2	10		2	61		8	Ti
V	0		0				0		0	0		0	0		0	0		0	V
Cr	0		0				0		0	2		1	0		0	3		1	Cr
Mn	5		1				3		0	2		0	5		1	8		1	Mn
Fe	41		6				33		5	70		9	68		9	497		57	Fe
Ni	0		0				1		1	0		0	0		0	0		0	Ni
Cu	100		12				26		4	0		0	27		4	0		0	Cu
Zn	24		3				30		4	0		0	29		4	0		0	Zn
Br	71		8				80		9	0		0	71		8	0		0	Br
Pb	226		27				317		37	0		0	225		27	0		0	Pb

	02/20/80						02/21/80						02/23/80						
	F			C			F			C			F			C			
Al	712	±	82	0	±	0	248	±	31				1433	±	163				Al
Si	687		78	1029		116	326		38				2748		309				Si
S	8501		972	0		0	566		72				599		76				S
Cl	153		26	157		21	89		16				145		21				Cl
K	927		106	0		0	190		23				388		45				K
Ca	171		21	237		28	59		8				149		18				Ca
Ti	14		3	14		2	3		1				31		4				Ti
V	0		0	0		0	0		0				1		0				V
Cr	1		1	0		0	0		0				2		1				Cr
Mn	8		1	10		1	3		0				7		1				Mn
Fe	96		13	130		16	52		8				263		31				Fe
Ni	2		1	0		0	2		1				0		0				Ni
Cu	145		17	0		0	29		4				41		6				Cu
Zn	88		11	0		0	25		3				26		4				Zn
Br	191		22	0		0	84		10				78		9				Br
Pb	849		96	0		0	244		29				254		30				Pb

TABLE 1C con't

	02/26/80				02/27/80				02/28/80				
	F		C		F		C		F		C		
Al	552	± 64	3114	± 350	940	± 108	4391	± 493	553	± 65	4066	± 457	Al
Si	1414	160	11221	1257	2185	246	16189	1813	1234	139	14416	1614	Si
S	555	71	0	0	1351	161	0	0	693	86	0	0	S
Cl	150	22	139	19	118	19	164	22	133	20	177	23	Cl
K	354	42	936	106	433	50	1289	146	311	37	1133	128	K
Ca	96	12	633	72	154	19	729	83	86	11	599	68	Ca
Ti	15	3	131	15	29	4	191	22	13	2	169	20	Ti
V	0	0	2	0	1	0	4	0	0	0	3	0	V
Cr	0	0	8	1	0	0	8	2	2	1	9	2	Cr
Mn	5	1	19	2	7	1	28	3	6	1	23	3	Mn
Fe	146	18	1034	117	220	26	1534	173	115	15	1314	148	Fe
Ni	1	1	0	0	0	0	2	1	1	1	0	0	Ni
Cu	42	6	0	0	42	6	0	0	30	4	0	0	Cu
Zn	19	3	22	3	32	4	0	0	24	3	0	0	Zn
Br	67	8	0	0	111	13	0	0	72	9	0	0	Br
Pb	183	22	0	0	345	40	0	0	227	27	0	0	Pb

	02/29/80				03/06/80				03/07/80				
	F		C		F		C		F		C		
Al	830	± 95			228	± 29	0	± 0	255	± 32	244	± 30	Al
Si	2120	239			247	29	522	60	253	30	841	95	Si
S	497	65			1671	197	0	0	1753	206	0	0	S
Cl	43	11			24	10	35	7	45	12	83	13	Cl
K	224	27			330	39	0	0	300	36	0	0	K
Ca	89	11			85	11	176	21	75	10	89	11	Ca
Ti	23	3			6	1	8	2	6	2	10	2	Ti
V	0	0			0	0	0	0	0	0	0	0	V
Cr	1	1			0	0	1	1	0	0	0	0	Cr
Mn	4	1			4	1	5	1	5	1	0	0	Mn
Fe	176	21			32	5	65	9	35	6	81	10	Fe
Ni	2	1			1	1	0	0	2	1	0	0	Ni
Cu	45	6			63	8	0	0	89	11	0	0	Cu
Zn	15	2			43	6	0	0	38	5	0	0	Zn
Br	73	9			48	6	0	0	68	8	0	0	Br
Pb	254	30			195	23	0	0	276	32	0	0	Pb

TABLE 2C

1982-83 XRF Analysis Results:
Fine and Coarse Concentrations (ng/m^3)

	11/17/82				11/20/82				11/23/82				
	F		C		F		C		F		C		
Al	128	± 39	609	± 609	86	± 26	660	± 660	69	± 21	630	± 630	Al
Si	215	32	1,509	1,270	226	34	2,080	620	216	32	1,770	1,420	Si
S	566	28	86	17	103	5	44	9	412	21	43	9	S
Cl	107	5	63	13	34	2	68	14	51	3	107	21	Cl
K	604	30	303	30	104	5	281	28	232	12	267	27	K
Ca	174	9	542	54	26	1	175	18	41	2	172	17	Ca
Ti	10	3	31	3	15	3	46	4	10	3	39	3	Ti
V	4	2	3	2	3	2	0	7	5	2	4	2	V
Cr	3	2	2	2	3	2	4	2	0	5	3	2	Cr
Mn	15	1	18	2	2	1	7	1	7	1	10	1	Mn
Fe	62	3	338	17	47	2	345	17	54	3	332	17	Fe
Ni	1	1	0	2	1	1	1	1	1	1	1	1	Ni
Cu	14	1	4	1	10	1	3	1	15	1	3	1	Cu
Zn	32	2	10	1	8	1	10	1	14	1	11	1	Zn
Br	59	3	8	0	27	1	5	0	29	1	3	0	Br
Pb	177	9	36	2	96	5	28	1	89	4	21	1	Pb

	11/26/82				11/29/82				12/2/82				
	F		C		F		C		F		C		
Al	157	± 47	1,680	± 1,680	26	± 21	621	± 621	144	± 43	1,140	± 1,140	Al
Si	490	73	4,290	3,430	154	23	1,810	1,450	355	53	3,070	2,450	Si
S	801	40	130	26	316	16	91	18	384	19	79	16	S
Cl	169	8	165	33	90	4	100	20	79	4	35	7	Cl
K	504	25	671	67	383	19	272	27	261	13	451	45	K
Ca	67	3	445	44	150	7	628	63	46	2	293	29	Ca
Ti	10	4	91	5	17	4	28	3	15	3	66	4	Ti
V	0	8	12	3	6	2	0	6	6	2	0	7	V
Cr	3	2	9	2	7	2	2	2	2	2	6	2	Cr
Mn	6	2	20	2	8	2	19	2	6	1	12	2	Mn
Fe	115	6	861	43	49	2	334	17	80	4	583	29	Fe
Ni	0	2	2	1	3	1	1	1	1	1	1	1	Ni
Cu	9	1	4	1	13	1	3	1	16	1	7	1	Cu
Zn	35	2	25	1	22	1	11	1	20	1	15	1	Zn
Br	90	5	13	1	38	2	6	0	41	2	7	0	Br
Pb	263	13	74	4	132	7	35	2	140	7	45	2	Pb

TABLE 2C con't

	12/5/82						12/8/82						12/11/82						
	F			C			F			C			F			C			
Al	151	±	45	758	±	758	91	±	27	281	±	281	108	±	32	59	±	59	Al
Si	307		46	2,220		1,780	156		23	1,010		810	144		22	518		414	Si
S	478		24	72		14	756		38	45		9	1,640		80	131		38	S
Cl	161		8	6		2	82		4	16		3	136		7	0		5	Cl
K	474		24	337		34	276		14	156		16	674		34	139		14	K
Ca	57		3	269		27	57		3	236		24	53		3	152		15	Ca
Ti	13		3	42		4	0		9	28		3	4		3	22		3	Ti
V	0		7	9		3	0		6	3		2	0		7	0		7	V
Cr	3		2	7		2	0		5	7		2	2		2	5		2	Cr
Mn	3		1	13		2	2		1	9		1	6		1	11		1	Mn
Fe	76		4	427		21	38		2	231		12	28		2	132		7	Fe
Ni	0		2	0		2	0		2	5		1	0		2	1		1	Ni
Cu	13		1	5		1	8		1	11		1	5		1	2		1	Cu
Zn	31		2	9		1	20		1	9		1	50		3	7		1	Zn
Br	64		3	7		0	33		2	4		0	80		4	9		0	Br
Pb	192		10	40		2	104		5	27		1	232		12	44		2	Pb

	12/14/82						12/17/82						12/20/82						
	F			C			F			C			F			C			
Al	91	±	27	0	±	52	0	±	52	58	±	58	0	±	59	225	±	225	Al
Si	82		12	166		133	0		18	416		332	53		8	675		540	Si
S	1,120		60	176		35	95		5	14		3	636		32	46		9	S
Cl	177		9	0		5	14		2	2		2	67		3	14		3	Cl
K	580		29	100		10	127		6	63		6	255		13	110		11	K
Ca	49		2	108		11	41		2	165		16	26		1	114		11	Ca
Ti	4		3	14		3	20		4	17		3	9		4	21		2	Ti
V	3		2	4		2	8		2	0		7	5		2	5		2	V
Cr	0		5	3		2	4		2	2		2	3		2	4		2	Cr
Mn	7		1	6		1	6		1	4		1	2		1	4		1	Mn
Fe	39		2	80		4	15		2	91		5	32		2	150		7	Fe
Ni	0		2	2		1	1		1	0		2	1		1	2		1	Ni
Cu	5		1	3		1	3		1	0		2	26		1	11		1	Cu
Zn	47		2	8		1	6		1	2		1	17		1	4		1	Zn
Br	102		5	16		1	17		1	3		0	49		2	5		0	Br
Pb	299		15	68		3	63		3	13		1	156		8	25		1	Pb

TABLE 2C con't

	12/23/82				12/26/82				12/29/82				
	F		C		F		C		F		C		
Al	30	± 15	133	± 133	0	± 59	289	± 289	57	± 18	427	± 427	Al
Si	56	8	423	338	73	11	735	588	122	18	1,100	880	Si
S	191	10	24	5	355	18	42	8	538	27	40	8	S
Cl	56	3	13	3	75	4	33	7	84	4	17	3	Cl
K	143	7	70	7	363	18	137	14	251	13	171	17	K
Ca	21	1	69	7	69	3	287	29	28	1	108	11	Ca
Ti	9	3	8	3	5	4	18	3	4	3	21	3	Ti
V	0	6	0	6	0	7	0	6	0	6	0	6	V
Cr	0	5	0	5	0	6	2	2	0	5	3	2	Cr
Mn	2	1	0	4	7	2	10	1	2	1	7	1	Mn
Fe	20	1	91	5	24	2	149	7	32	2	220	11	Fe
Ni	0	2	2	1	0	2	1	1	0	2	2	1	Ni
Cu	26	1	11	1	25	1	11	1	26	1	11	1	Cu
Zn	9	1	2	1	25	1	5	1	19	1	5	1	Zn
Br	34	2	4	0	34	2	4	0	36	2	4	0	Br
Pb	107	5	18	1	98	5	19	1	117	6	24		Pb

	1/01/83				1/04/83				1/07/83				
	F		C		F		C		F		C		
Al	177	± 53	1,150	± 1,150	76	± 23	0	± 51	59	± 21	163	± 163	Al
Si	345	52	3,030	2,430	90	13	125	100	34	8	363	291	Si
S	759	38	66	13	1,080	50	104	21	841	42	92	18	S
Cl	190	10	55	11	138	7	13	3	167	8	73	15	Cl
K	549	27	490	49	584	29	70	7	409	20	113	11	K
Ca	49	2	307	31	66	3	175	18	34	2	145	15	Ca
Ti	7	3	69	4	7	3	11	3	9	3	16	3	Ti
V	0	7	0	7	0	7	0	6	6	2	2	2	V
Cr	0	5	5	2	3	2	3	2	2	2	3	2	Cr
Mn	2	1	17	2	7	1	5	1	5	1	7	1	Mn
Fe	81	4	606	30	41	2	64	3	44	2	165	8	Fe
Ni	0	2	1	1	0	2	2	1	0	2	2	1	Ni
Cu	33	2	12	1	28	1	8	1	24	1	8	1	Cu
Zn	32	2	10	1	41	2	3	1	36	2	8	1	Zn
Br	95	5	10	1	102	5	9	0	142	7	15	1	Br
Pb	267	13	56	3	303	15	39	2	409	20	69	3	Pb

TABLE 2C con't

	1/10/83				1/13/83				1/16/83				
	F		C		F		C		F		C		
Al	81	± 24	206	± 206	1,900	± 300	6,930	± 6,930	736	± 221	5,190	± 5,190	Al
Si	41	6	620	496	2,480	370	17,300	*********	1,600	240	12,700	*********	Si
S	175	9	26	5	355	18	164	33	688	34	143	29	S
Cl	45	2	5	1	115	6	60	12	200	10	28	6	Cl
K	177	9	102	10	657	33	2,710	270	659	33	2,010	200	K
Ca	15	1	61	6	199	10	1,150	110	126	6	819	82	Ca
Ti	0	9	18	3	46	4	347	17	31	4	250	13	Ti
V	0	6	0	6	6	3	19	4	5	2	14	3	V
Cr	2	2	0	5	6	2	30	3	5	2	21	2	Cr
Mn	2	1	4	1	16	2	71	4	10	2	48	2	Mn
Fe	26	2	145	7	532	27	3,710	190	363	18	2,610	130	Fe
Ni	1	1	0	2	1	1	5	1	1	1	3	1	Ni
Cu	11	1	4	1	23	1	13	1	50	3	21	1	Cu
Zn	11	1	2	1	22	1	36	2	42	2	32	2	Zn
Br	23	1	3	0	65	3	11	1	90	5	14	1	Br
Pb	88	4	17	1	208	10	87	4	269	13	84	4	Pb

	1/19/83				1/22/83				1/25/83				
	F		C		F		C		F		C		
Al	459	± 138	3,370	± 3,370	122	± 37	1,470	± 1,470	302	± 90	2,020	± 2,020	Al
Si	812	122	8,730	6,980	408	61	3,900	3,120	704	106	5,450	4,360	Si
S	1,760	90	190	38	642	32	73	15	873	44	130	26	S
Cl	146	7	9	2	132	7	12	2	261	13	109	22	Cl
K	780	39	1,350	130	418	21	595	59	666	33	820	82	K
Ca	87	4	544	54	56	3	328	33	89	4	492	49	Ca
Ti	17	3	173	9	11	4	71	4	24	4	106	5	Ti
V	0	7	8	3	7	3	0	8	5	3	6	3	V
Cr	4	2	12	2	3	2	7	2	0	6	11	2	Cr
Mn	9	1	35	2	7	2	18	2	12	2	24	2	Mn
Fe	176	9	1,730	90	100	5	740	37	177	9	1,030	50	Fe
Ni	0	2	4	1	0	2	2	1	1	1	1	1	Ni
Cu	28	1	10	1	20	1	7	1	21	1	6	1	Cu
Zn	36	2	22	1	23	1	10	1	35	2	15	1	Zn
Br	90	4	14	1	97	5	11	1	129	6	13	1	Br
Pb	250	12	70	3	268	13	55	3	229	17	65	3	Pb

TABLE 2C con't

	1/28/83				1/31/83				2/03/83				
	F		C		F		C		F		C		
Al	291	± 87	2,480	± 2,480	607	± 182	4,300	± 4,300	407	± 122	2,280	± 2,280	Al
Si	749	112	6,420	5,140	1,480	220	10,700	8,600	812	122	5,630	4,500	Si
S	159	8	106	21	651	33	152	30	378	19	77	15	S
Cl	38	2	68	14	159	8	38	8	50	3	23	5	Cl
K	198	10	929	93	524	26	1,700	170	251	13	870	87	K
Ca	54	3	440	44	105	5	682	68	62	3	364	36	Ca
Ti	21	4	118	6	27	4	209	10	16	3	101	5	Ti
V	6	3	6	3	3	3	9	3	0	6	3	2	V
Cr	6	2	10	2	4	2	19	2	4	2	8	2	Cr
Mn	4	2	22	2	9	2	42	2	3	1	20	2	Mn
Fe	159	8	1,180	60	346	17	2,200	110	173	9	1,110	60	Fe
Ni	0	2	1	1	0	2	5	1	0	2	1	1	Ni
Cu	20	1	9	1	34	2	11	1	22	1	9	1	Cu
Zn	6	1	18	1	27	1	31	2	12	1	15	1	Zn
Br	48	2	8	0	106	5	15	1	34	2	5	0	Br
Pb	154	8	49	2	316	16	85	4	107	5	33	2	Pb

	2/06/83				2/09/83				2/12/83				
	F		C		F		C		F		C		
Al	202	± 61	2,230	± 2,230	0	± 67	25	± 25	0	± 55	77	± 77	Al
Si	679	102	5,580	4,460	0	25	306	245	41	7	409	327	Si
S	459	23	80	16	1,050	50	37	7	430	22	40	8	S
Cl	94	5	26	5	99	5	0	5	49	2	27	5	Cl
K	438	22	914	91	470	23	54	5	295	15	105	10	K
Ca	91	5	556	56	20	1	46	5	106	5	382	38	Ca
Ti	19	4	120	6	0	11	22	3	8	3	15	3	Ti
V	3	3	11	3	0	8	4	2	3	2	2	2	V
Cr	4	2	10	2	0	6	5	2	2	2	3	2	Cr
Mn	7	2	27	2	3	2	3	1	10	1	19	2	Mn
Fe	147	7	1,130	60	25	2	88	4	27	2	112	6	Fe
Ni	1	1	2	1	0	2	0	2	0	2	1	1	Ni
Cu	26	1	11	1	22	1	6	1	11	1	3	1	Cu
Zn	21	1	17	1	30	2	1	1	14	1	4	1	Zn
Br	47	2	8	0	98	5	5	0	33	2	4	0	Br
Pb	143	7	46	2	303	15	26	1	111	6	18	1	Pb

TABLE 2C con't

	2/15/83				2/18/83				2/21/83				
	F		C		F		C		F		C		
Al	520	± 156	4,340	± 4,340	0	± 56	25	± 25	442	± 133	3,040	± 3,040	Al
Si	1,330	200	11,300	9,000	0	20	169	135	1,000	150	7,990	6,390	Si
S	323	16	106	21	318	16	63	13	411	21	107	21	S
Cl	64	3	19	4	25	2	5	1	154	8	7	2	Cl
K	425	21	1,740	170	187	9	47	5	497	25	1,210	120	K
Ca	112	6	815	82	25	1	79	8	98	5	610	61	Ca
Ti	31	4	212	11	15	4	5	3	22	3	152	8	Ti
V	5	3	17	3	7	3	0	6	0	7	10	3	V
Cr	5	2	12	2	3	2	0	5	2	2	16	2	Cr
Mn	11	2	41	2	4	2	5	1	5	1	32	2	Mn
Fe	289	14	2,220	110	13	2	50	3	214	11	1,500	70	Fe
Ni	1	1	3	1	1	1	0	2	0	2	3	1	Ni
Cu	14	1	5	1	11	1	3	1	22	1	10	1	Cu
Zn	25	1	25	1	10	1	1	1	25	1	23	1	Zn
Br	72	4	12	1	44	2	4	0	108	5	13	1	Br
Pb	233	12	76	4	134	7	18	1	326	16	71	4	Pb

	2/24/83				2/27/83				
	F		C		F		C		
Al	690	± 207	4,520	± 4,520	170	± 51	1,510	± 1,510	Al
Si	1,470	220	11,300	9,100	443	66	3,950	3,160	Si
S	545	27	140	28	117	6	57	11	S
Cl	66	3	19	4	23	2	12	2	Cl
K	398	20	1,770	180	153	8	591	59	K
Ca	106	5	790	79	47	2	310	31	Ca
Ti	28	3	227	11	20	3	68	4	Ti
V	0	7	9	3	5	2	9	2	V
Cr	3	2	17	2	5	2	4	2	Cr
Mn	5	1	44	2	5	1	14	2	Mn
Fe	290	15	2,230	110	94	5	724	36	Fe
Ni	0	2	3	1	1	1	0	2	Ni
Cu	18	1	9	1	14	1	4	1	Cu
Zn	22	1	35	2	10	26	14	1	Zn
Br	95	5	16	1	34	2	6	0	Br
Pb	309	15	90	5	129	6	40	2	Pb

TABLE 3C - SUMMARY
Monthly Averages Of
1982-83 XRF Analysis Results:
Fine and Coarse Concentrations (ng/m^3)

	November				December			
	Fine		Coarse		Fine		Coarse	
Element	Avg.	S.D.	Avg.	S.D.	Avg.	S.D.	Avg.	S.D.
Al	93	51	840	470	67	58	337	359
Si	260	132	2,292	1,135	135	114	1,033	915
S	440	263	79	36	619	462	73	61
Cl	90	53	101	41	93	50	14	13
K	365	202	359	175	340	182	173	124
Ca	92	66	392	210	45	16	180	84
Ti	12	3	47	26	8	6	26	17
V	4	2	4	5	2	3	2	3
Cr	3	2	4	3	1	2	4	2
Mn	8	5	15	6	4	2	8	4
Fe	65	28	442	234	39	22	215	165
Ni	1	1	1	1	0	0	2	1
Cu	12	3	3	1	15	10	7	4
Zn	22	11	13	7	24	15	7	4
Br	49	26	7	4	49	26	6	4
Pb	151	72	39	21	151	72	32	17

	January				February			
	Fine		Coarse		Fine		Coarse	
Element	Avg.	S.D.	Avg.	S.D.	Avg.	S.D.	Avg.	S.D.
Al	355	311	2,480	2,257	270	255	2,005	1,760
Si	795	772	6,303	5,568	642	564	5,182	4,420
S	726	448	113	48	448	255	79	34
Cl	145	65	43	33	69	41	15	10
K	511	194	990	865	346	127	811	676
Ca	80	51	468	323	74	36	439	278
Ti	18	13	126	108	18	10	102	83
V	3	3	6	6	3	3	7	5
Cr	3	2	11	9	3	2	8	5
Mn	8	4	27	21	6	3	23	15
Fe	186	162	1,289	1,164	141	109	1,018	857
Ni	0	1	2	2	0	1	1	1
Cu	27	10	10	5	18	5	7	3
Zn	28	12	17	12	19	7	15	12
Br	90	34	11	4	63	31	8	4
Pb	252	85	61	21	199	93	46	27

TABLE 4C

Winter 1982-83 Organic Composition
Total Suspended Particulate
($\mu g/m^3$)

Sample Date	Volatile Carbon	Non-Volatile Carbon
11-17	56.6 ± 5.7	4.8 ± 0.5
11-20	33.2 3.3	2.7 0.3
11-23	41.8 4.2	5.2 0.5
11-26	59.1 5.9	5.5 0.6
11-29	38.6 3.9	3.1 0.3
MEAN/STD DEV	45.9/11.4	4.3/1.3
12-02	50.7 5.1	2.0 0.2
12-05	63.6 6.4	4.8 0.5
12-08	40.9 4.1	2.9 0.3
12-11	89.1 8.9	9.6 1.0
12-14	89.5 9.0	9.0 0.9
12-17	36.8 3.7	3.2 0.3
12-20	37.6 3.8	2.9 0.3
12-23	46.5 4.7	3.6 0.4
12-26	50.5 5.1	5.5 0.6
12-29	52.5 5.3	4.5 0.5
MEAN/STD DEV	55.8/19.4	4.8/2.6
01-01	102.2 10.2	9.6 1.0
01-04	58.9 5.9	8.6 0.9
01-10	49.8 5.0	4.6 0.5
01-19	83.2 8.3	7.4 0.7
01-22	54.7 5.5	5.5 0.6
01-25	53.8 5.4	3.6 0.4
01-28	56.7 5.7	4.4 0.4
01-31	58.3 5.8	3.9 0.4
MEAN/STD DEV	64.7/18.2	5.6/2.3
02-03	43.3 4.3	3.2 0.3
02-06	52.8 5.3	3.8 0.4
02-09	44.6 4.5	4.2 0.4
02-12	39.7 4.0	4.1 0.4
02-15	70.3 7.0	5.5 0.6
02-18	27.4 2.7	1.7 0.2
02-21	58.5 5.9	4.3 0.4
02-24	69.9 7.0	5.2 0.5
02-27	43.6 4.4	2.8 0.3
MEAN/STD DEV	50.0/14.2	3.9/1.2

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