THE ROLE OF METEOROLOGICAL DISPERSION AND SPACE HEATING DEMAND IN AMBIENT CONCENTRATIONS OF CARBONACEOUS AEROSOLS

John Anthony Rau
B.S., Rensselaer Polytechnic Institute, 1958
M.S., University of Michigan, 1967

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The thesis "The Role of Meteorological Dispersion and Space Heating Demand in Ambient Concentrations of Carbonaceous Aerosols" by John A. Rau has been examined and approved by the following Examination Committee:

James J. Huntzicker, Associate Professor
Thesis Research Advisor

Douglas F. Barofsky
Associate Professor

Jack Devletian
Associate Professor

M. A. K. Khalil
Assistant Professor

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ABSTRACT

This study interpreted ambient aerosol carbon data obtained during a one-year data base study conducted by the Oregon Department of Environmental Quality in Medford, Oregon. Twenty-four hour total and fine aerosol samples were taken at an urban site (Medford Justice Building), an industrial site (White City), and a background site (Dodge Road). Organic and elemental carbon analyses were done on these samples with an automated analysis system developed at the Oregon Graduate Center.

These data demonstrate the importance of carbon as a component of both total (aerosol collected with a high volume sampler) and fine (less than 2 μm) aerosol. At the Justice Building and White City sites ambient carbon concentrations of total carbon ranged from a low value of 10 $\mu g/m^3$ in the summer to high values of about 100 $\mu g/m^3$ during the winter. Fine carbon aerosol comprised about half of these values. As a fraction of aerosol mass carbon composition ranges from 15 to 50% for total aerosol and 25 to 60% for fine aerosol at these sites.

A series of models were developed to characterize fine aerosol carbon in terms of meteorological dispersion and two general carbon sources: carbon not associated with space heating and carbon resulting from space heating emissions. Meteorological dispersion was characterized by two independent methods. One was a box model in which the dispersion parameter was the inverse product of wind speed and mixing height. The other used the ambient lead concentration as a measure of

dispersion. Both methods produced similar results. Space heating demand was characterized by heating degree day values. For the urban site this modeling procedure explained 89% of the variance in the fine carbon concentration. It also showed that in winter the ratio of space heating derived carbon to non-space heating carbon ranged from 4 to 6. For data from White City and Dodge Road these models accounted for 56% of the fine carbon variance. At both these sites the ratio of space heating aerosol carbon to non-space heating aerosol carbon ranged from 1 to 1.5.

I. INTRODUCTION

A. The Relationship Between Meteorology and Ambient Air Pollution

1. Background

The major reason for most air pollution studies derives from the concept that air pollution causes detrimental health effects to at least some segment of the population and that the magnitude of these effects increases with increasing ambient concentration levels of pollution. The Clean Air Act of 1970 set allowable limits on ambient concentration of certain pollutants, such as airborne particulate matter, and required that all major urban areas must either meet the specifications of the act or devise plans by which they will bring their air quality into compliance.

Since ambient pollution levels result from the combined action of a variety of variable emission sources acted upon by an ever-changing atmospheric dispersion mechanism, it is important to understand both how much each source type contributes to local pollution composite and to understand the role atmospheric dispersion plays in forming given ambient pollution levels. Meteorology affects ambient pollution in at least three general ways. It (1) determines pollutant dispersion, it (2) determines advection direction, and, for some sources, it may (3) determine emission levels. For example, temperature determines emission levels associated with space heating and cooling. Zeldin and Meizel (1977) point out that while it is recognized that meteorological dispersion plays a role in the determination of ambient pollution concentrations, efforts to quantify this role have not received a large share of current air pollution research efforts.

To understand the nature of urban pollution, it is necessary to understand both the strength and composition variability of sources as well as the nature of the atmospheric dispersion mechanism acting in the area being studied. Certainly emission sources are the easier of these two pollution aspects to understand. Understanding the atmospheric dispersion mechanism, on the other hand, presents formidable problems. Ideally, one would like to characterize the strength of the dispersive mechanism, in terms of readily measurable meteorological parameters existing at any time between a receptor and all sources impinging on that receptor. If dispersive strength can be characterized, it can be used to predict ambient pollution levels when pollution source strengths are known. Also the ability to separate ambient pollution variability into the variability due to meteorological dispersion and that due to emission strength variability makes it possible to determine how emission variability influences ambient pollution levels. This is the basis of environmental planning. Dispersion strength information is also necessary to evaluate the effectiveness of pollution control strategies to assure that ambient pollution concentration averages are not biased by differences in dispersion strength.

The purpose of this study was to evaluate the atmospheric dispersion mechanism strength in terms of the variability that it introduced into ambient concentration levels of organic, elemental, and total carbon at three receptor sites in the Medford, Oregon, area. These particular pollutant species were used because the data were available as part of a larger study. This study could have been done with any other species of primary pollutant that was not highly subject to atmospheric removal mechanisms. The procedure used was to correlate ambient carbon levels with a meteorological factor which was chosen to be a measure of atmospheric dispersion strength for the whole area of study.

2. Box Model

The meteorological dispersion factors used in this study were derived from the "box model." In this model:

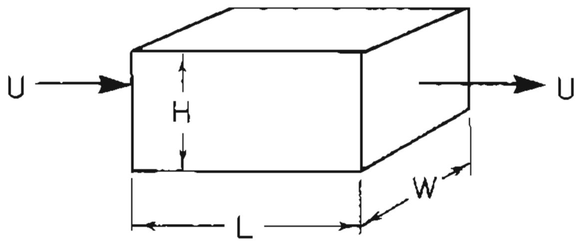


Figure 1. The Box Model

$$\frac{d\chi}{dt} = \frac{Q}{H} - \eta\chi \qquad (1)$$

χ = ambient pollution concentration in the box ($\mu g/m^3$)

Q = emission source strength (μg/sec)

U = wind speed (m/sec)

H = mixing height (m)

η = loss factor (1/sec)

The solution of Eqn. (1) is:

$$\chi = \chi_o e^{-\eta t} + \frac{Q}{H\eta}\,(1 - e^{-\eta t}) \tag{2}$$

For steady state conditions:

$$\chi = \frac{Q}{H\eta} \tag{3}$$

In general, η represents all mechanisms that remove particulate matter from the box. When wind transport is the only loss mechanism

$$\eta = \frac{U}{L} \tag{4}$$

and

$$\chi = \frac{QL}{UH} \tag{5}$$

We can define $\tau = L/U$ = the travel time of the wind through the box. When a variety of loss mechanisms act together, the combined effect can be treated like resistances in parallel or like a number of pipes flowing out of the box. This model assumes that pollutants in the box are uniformly mixed. A detailed look at a point source operating in the box model will illustrate how this model approximates the atmosphere and some of the limitations of the model. In the time Δt the source will emit an amount of pollutant Δm. A volume of air Δv with the dimensions of $W x H x U x\ \Delta t$ will have passed the point during this time.

The instantaneous mixing criterion of the box model requires that Δm is uniformly mixed into Δv during the time. But since the receptor is located at the downwind end of the box, if Δm is uniformly mixed into Δv by the time Δv gets to the receptor, the receptor will perceive that the instantaneous mixing criterion has been satisfied. Thus the difference between the box model and the real atmosphere involves only whether Δm is uniformly mixed in Δv or it isn't. For point sources located in the upwind regions of the box, the transit time from the source to the receptor may be quite long. Thus, if the atmosphere is sufficiently unstable, uniform vertical mixing will be accomplished, and if eddy turbulence is sufficient, uniform horizontal mixing will be accomplished. For point sources closer to the receptor the mixing time is less and the possibility of uniform mixing becomes less. When uniform mixing is not accomplished, any source may impact the receptor to a larger or smaller degree than under conditions of uniform mixing. For any given source the degree to which it is uniformly mixed cannot be evaluated because in the real atmosphere the requirement of uniform mixing depends on the complex interaction of a variety of atmospheric transport mechanisms such as turbulent diffusion and vertical buoyancy forces. These mechanisms, in turn, depend on wind speed, wind shear stresses, solar energy input, cloud cover, and the atmospheric vertical temperature profile. All of these mechanisms require a finite time period to act and may not be compatible with the instantaneous mixing assumption of the model. Therefore, the model may not correctly deal with transients if they take longer to accomplish than the transit time from the source to the receptor. The box model does not distinguish between point and area sources and also, as expressed in Eqn. (5), does not incorporate any other pollutant removal mechanisms except wind advection. Mechanisms which add aerosol within the box, such as secondary gas to particle conversion or mechanisms which remove aerosol from within the box such as rainout, washout, fallout, or surface deposition, are not considered by the simple box model. Even the assumption of constant wind velocity through the box is not satisfied in reality, because wind speed increases and changes direction with increasing altitude. Despite these deviations from reality the box model does a reasonably good job of dispersion modeling, and its simplicity certainly recommends its use.

The box model has been used by numerous authors to examine the effects of atmospheric dispersion. Kleinman et al. (1976) showed that wintertime aerosol emissions were higher than summertime values in New York City, even though better wintertime dispersion conditions prevented this conclusion from being drawn from ambient aerosol levels. Trindade et al. (1980) correlated TSP with 1/UH and with wind speed and mixing height individually for two years of data from Rio de Janiero. They found higher correlation coefficients for 1/UH than for either wind speed or mixing height alone. Van Dop and Kruizinga (1975) developed a relation which predicts daily SO_2 concentration in Rotterdam using a 1/UH term to predict daytime meteorological dispersion. Haagenson (1979) examined meteorologically induced CO variability in Denver using U and H in separate correlations. He noted that higher

CO levels during the winter months were associated mainly with lower mixing heights. Edwards and Wheat (1978) correlated monthly average particulate lead concentrations in Denver with monthly average values of 1/UH and were able to attribute about 69% of the monthly lead concentration variance to meteorological variability described by 1/UH. Summers (1969) modelled smokiness in Montreal by the form QL/UH where Q equaled the rate of smoke production and L is a measure of city size. Buch (1976) correlated wind speed and direction with ambient particulate matter concentrations in Denmark and showed that particulate concentration decreased with increasing wind speed. He also noted that pollution concentration was a strong function of wind direction. In some studies wind speed alone was used to characterize atmospheric dispersion. Hanna (1978) used the factor CQ/U, where Q is the area source strength and C is an empirically determined constant, to model diurnal CO concentrations in Los Angeles. Miller (1978) used the same form to model SO_2 in Tennessee. Holzworth (1969) associated high pollution episodes with periods of low wind speed and low mixing height.

3. The Correlation Coefficient

When a least squares regression line is fitted to ambient carbon data plotted against 1/UH, the equation of the regression line can be written as:

$$\hat{\chi} = A + B\ (\frac{1}{UH}) \tag{6}$$

A = Y intercept

B = slope

$\hat{\chi}_i$ = the ith ambient carbon concentration predicted by the equation. Then:

$$\text{total variance} = S_T^2 = \Sigma\ (\chi_i - \overline{\chi})^2 \tag{7}$$

χ_i = the ith measured ambient carbon concentration

$\overline{\chi}$ = average ambient carbon concentration

n = number of ambient samples

Then the variance accounted for by the regression line is given by:

$$S_m^2 = \sum_{i=1}^{n}\ (\hat{\chi} - \chi_i)^2 \tag{8}$$

Then the fraction of the variance described by a particular meteorological factor is given by the meteorological coefficient of determination (r^2met.).

$$r_{MET} = \frac{S_m^2}{S_T^2} \tag{9}$$

r = correlation coefficient

In this study when ambient aerosol concentrations are correlated with 1/UH, the magnitude of the correlation coefficients obtained depends on how well the model approximates physical reality and how well:

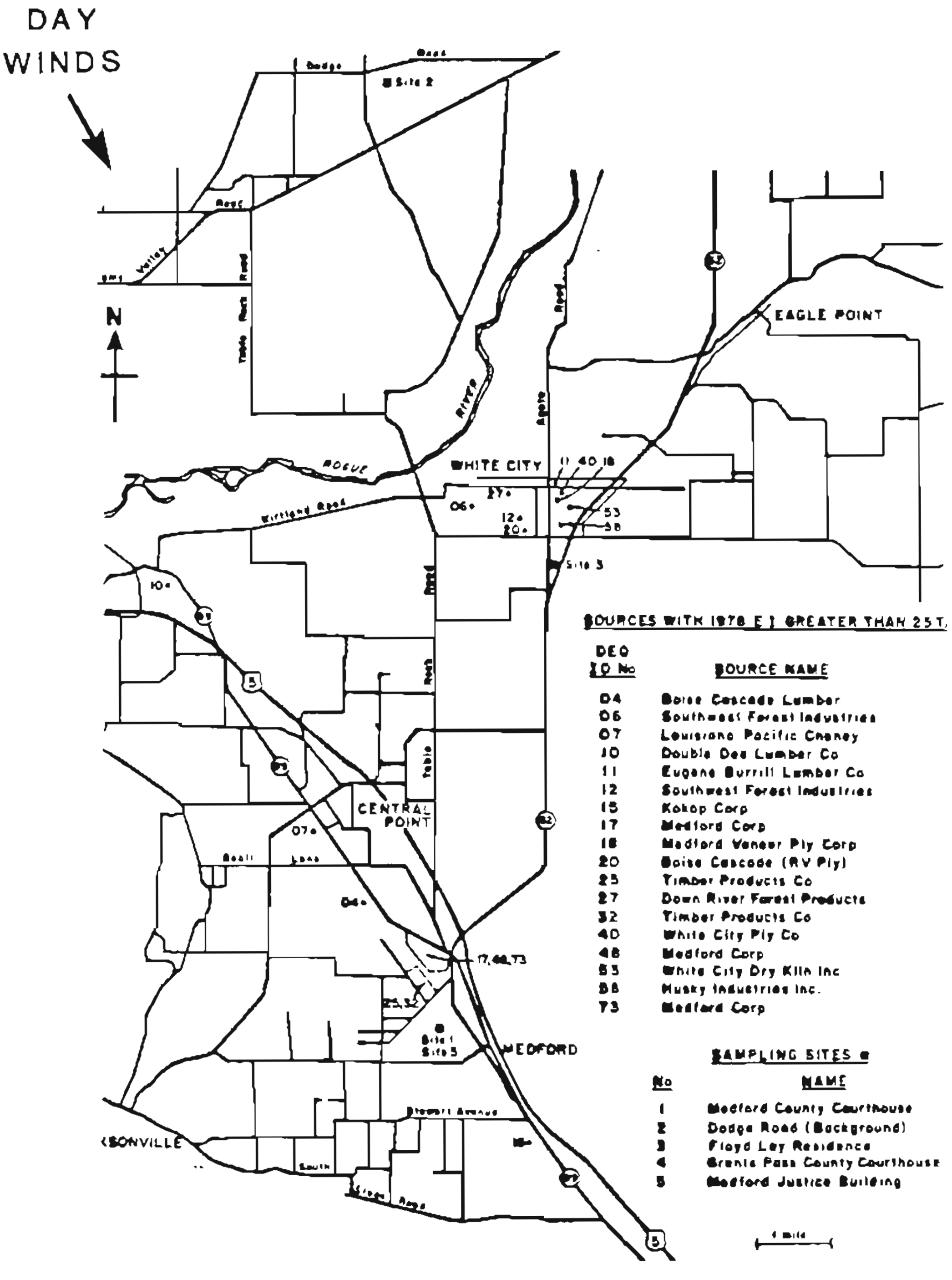
$$\begin{array}{l}\text{Average daily } 1/UH \\ \text{determined in} \\ \text{this study}\end{array} = \frac{1}{24} \int_{t_1 = 0}^{t_2 = 24} \frac{dt}{U(t)H(t)} \qquad (10)$$

The actual values of U and H are continuous functions of time and position. In the study it was assumed that they were not functions of position, i.e., a single value of both H and U was considered characteristic of the whole study area. The time dependence of wind speed was well approximated since wind speed was measured every six seconds and vector-summed to obtain hourly averages. The time dependence of H was treated by estimating hourly average values of H. Certainly some lack of correlation is introduced because Eqn. (10) is probably not exactly satisfied. In addition when ambient pollutant concentration values are correlated with 1/UH, no account is taken of the variation of emissions with time. This also results in a decrease of the correlation values obtained. Certainly in all cases where a meteorological dispersion factor is correlated with ambient pollutant concentrations, some further arguments or comparisons with other meteorological dispersion factors are necessary to determine how well the meteorological factor being considered models meteorological dispersion.

B. Description of Medford Area Particulate Matter Pollution

1. The Medford Valley TSP History and Wind Patterns

This study applied to the area around Medford, Oregon, located in the Bear Creek Valley in Jackson County. It is a roughly 15 km east-west by 20 km north-south plain about 365 m above sea level and surrounded by a 90 to 250 m high ring of mountains. An appendage to the valley, about 6.5 km wide, extends to the south-southeast toward Ashland. Since the surrounding mountains contain an abundant timber resource, a large timber-related industry has developed in the valley. The major point sources in the area and the sampling sites are shown in Figure 2. Most of the industry is located in the White City area, which is about 8 km north of the Medford city center. Several large companies are also located on the northern edge of Medford. TSP has been measured in this area since 1969. These measurements have shown that the annual geometric mean TSP average has been above the 75 $\mu g/m^3$ standard required by the 1970 Clean Air Act in all years except 1973 and 1975. Because this area has been classified as an Air Quality Maintenance Area (AQMA), it is required that plans be made to reduce the ambient TSP levels to meet the requirements of the Clean Air Act. During the early 1970's considerable industrial pollution control equipment was installed, and while this certainly had some effect on TSP levels, a long term, significant reduction of the annual geometric



SOURCES WITH 1978 E I GREATER THAN 25 T.

DEQ ID No	SOURCE NAME
04	Boise Cascade Lumber
06	Southwest Forest Industries
07	Louisiana Pacific Cheney
10	Double Dee Lumber Co
11	Eugene Burrill Lumber Co
12	Southwest Forest Industries
15	Kokop Corp
17	Medford Corp
18	Medford Veneer Ply Corp
20	Boise Cascade (RV Ply)
25	Timber Products Co
27	Down River Forest Products
32	Timber Products Co
40	White City Ply Co
48	Medford Corp
53	White City Dry Kiln Inc
58	Husky Industries Inc.
73	Medford Corp

SAMPLING SITES ●

No	NAME
1	Medford County Courthouse
2	Dodge Road (Background)
3	Floyd Ley Residence
4	Grants Pass County Courthouse
5	Medford Justice Building

Figure 2. Medford study area map.

mean TSP level did not result. In fact, in recent years this average has increased considerably. It is important to pollution control strategy planning to determine how much of this increase results from short term poorer meteorological dispersion conditions and how much has resulted from industrial expansion or perhaps from new particulate pollution sources.

Low dispersion conditions involving low wind speeds and low atmospheric mixing heights often exist in Medford. Wind speeds generally average less than 2 to 3 meters per second and blow generally from the northwest during the day and from the southeast at night. This wind pattern is mainly the result of katabatic winds. Ashland is at the higher altitude end of the valley; thus, in the evening when the valley walls cool by radiative heat loss to the night sky, cool air flows down the valley walls and flows down the valley toward Medford. This causes southeast winds in Medford at night. In the morning when the valley walls heat the adjacent air, this air flows up the valley walls and causes air to flow up the valley from Medford to Ashland. This establishes northwest winds. During the day when the inversion layer weakens, surface winds also couple to the prevailing winds, usually resulting in the addition of a westerly component to the surface wind.

Inversions are common in the region and result from three mechanisms. In clear weather ground inversions result from radiational cooling of the ground and conduction cooling of the adjacent air after the sun goes down in the evening. This forms a pool of cool air next to the ground which lies under the warm air. The effect is enhanced by cooling of the air along mountain slopes by radiative cooling. Subsidence is a second mechanism, occurring mostly during the summer, that causes upper air inversions. In this mechanism a high pressure region causes upper air masses to subside. The subsiding air is warmed by compression and thus establishes an inversion. A third inversion mechanism is air mass advection, in which a warm air mass moves over a resident cool air mass or a cool air mass slides under a resident warm air mass. In either case an upper air inversion occurs. In a nationwide air pollution potential survey by Holzworth (1972) the Medford area was singled out as having one of the highest pollution potentials in the United States. Thus, because of unfavorable meteorology and geography, the Medford area has the potential for having high levels of pollution even under conditions of moderate emissions. The results of the Medford DEQ study will have a critical effect on the economic growth of Jackson county because the future economic growth is constrained by the ability of existing pollution dispersion mechanisms to remove pollutants without allowing federal, state, or local air quality standards to be exceeded.

2. Sources of Carbon in the Atmosphere

On a mass basis the major components of urban aerosols are carbon, oxygen, sulfur, and silicon (Watson, 1979). Carbon and sulfur enter the atmosphere primarily as a result of combustion process emissions. For major sources in Medford the total carbon fraction of combustion-generated aerosols varies from 15% for hogfuel burners to 60% for residential wood burning emissions. Diesel engine emissions are almost totally carbon, but are not expected to be a large emission source in Medford. Based on emission inventories in the Medford area prepared by the Department of Environmental Quality (DEQ), it was found that area sources emit about 8000 tons/year of particulate matter, of which 317 tons/year is of automotive origin and 2510 tons comes from other combustion sources, such as space heating, forest fires, orchard heating, and field burning (DeCesar and Cooper, 1981). Point sources emit about 3700 tons/year of aerosol, most of which is composed of wood combustion-generated aerosol, wood industry dryer emissions, and particles of raw wood and bark.

II. EXPERIMENTAL PROCEDURE

A. Description of the Department of Environmental Quality Sampling Sites and Sampling Procedures

Three Medford sampling sites were used in this study, the Justice Building site, the White City site and the Dodge Road site. The Justice Building site was located on the roof of the newly constructed Justice Building on the corner of King and 8th Streets. The site is 16 m above the street level and is located between the downtown urban area and an older residential area. The White City site is located about 1.6 km south of White City, an area which contains a large industrial complex on Agate Road near the junction with Crater Lake Highway. The sampler was located on a platform 2.9 m above the ground. Roads near the site carry a considerable amount of high speed truck traffic. Also there are some agricultural areas to the south, east, and west. The Dodge Road site is a rural site located about 22.5 km north of downtown Medford. The sampler was located on a platform 2.9 m above the ground. The surrounding area is agricultural and sparsely populated.

Sampling was done by standard high volume samplers using glass fiber filter material. Fine mass samples were obtained by mounting a Sierra high volume impactor in front of the standard high volume filter. This device removed particles with aerodynamic diameters greater than 2 μm from the sampling air stream. Samplers were run continually starting at midnight for 24-hour periods. A sample was taken at each site every six days starting in April 1979 and ending at the end of March 1980.

B. Oregon Graduate Center Carbon Analysis

1. Theory and Description of the Carbon Analysis System

The carbon analysis system is described by Johnson et al. (1980) and works by oxidation of carbon to CO_2, the reduction of CO_2 to CH_4, and the measurement of CH_4 by using a flame ionization detector (FID). A schematic drawing of the system is shown in Figure 3. Analysis starts by placing the sample (punchings taken directly from a glass or quartz sampling filter) into an oven with a continuously flowing, composition controlled atmosphere. The separation of organic carbon from elemental carbon is accomplished by first heating the specimen to 350°C in an atmosphere of 2% O_2 in helium. This procedure volatilizes or oxidizes low boiling point organic carbon compounds. The vapors along with the CO_2 produced are swept into the oxidation zone (MnO_2 at 950°C) of the oven where organic vapors are oxidized to CO_2, which then joins any CO_2 produced earlier. The He-CO_2 mixture then passes through a chromatographic column packed with Spherocarb. The Spherocarb column serves no analytical purpose, but is used to detect system malfunctions. After the He-CO_2 mixture leaves the Spherocarb column, it is mixed with H_2 and enters the catalytic methanator. Here the CO_2 is reduced to CH_4 by a Ni catalyst. The CH_4 is then measured by a FID. The FID signal is measured by an electrometer and processed by a microprocessor-controlled system. Signal peaks are recorded on hard copy, typic-

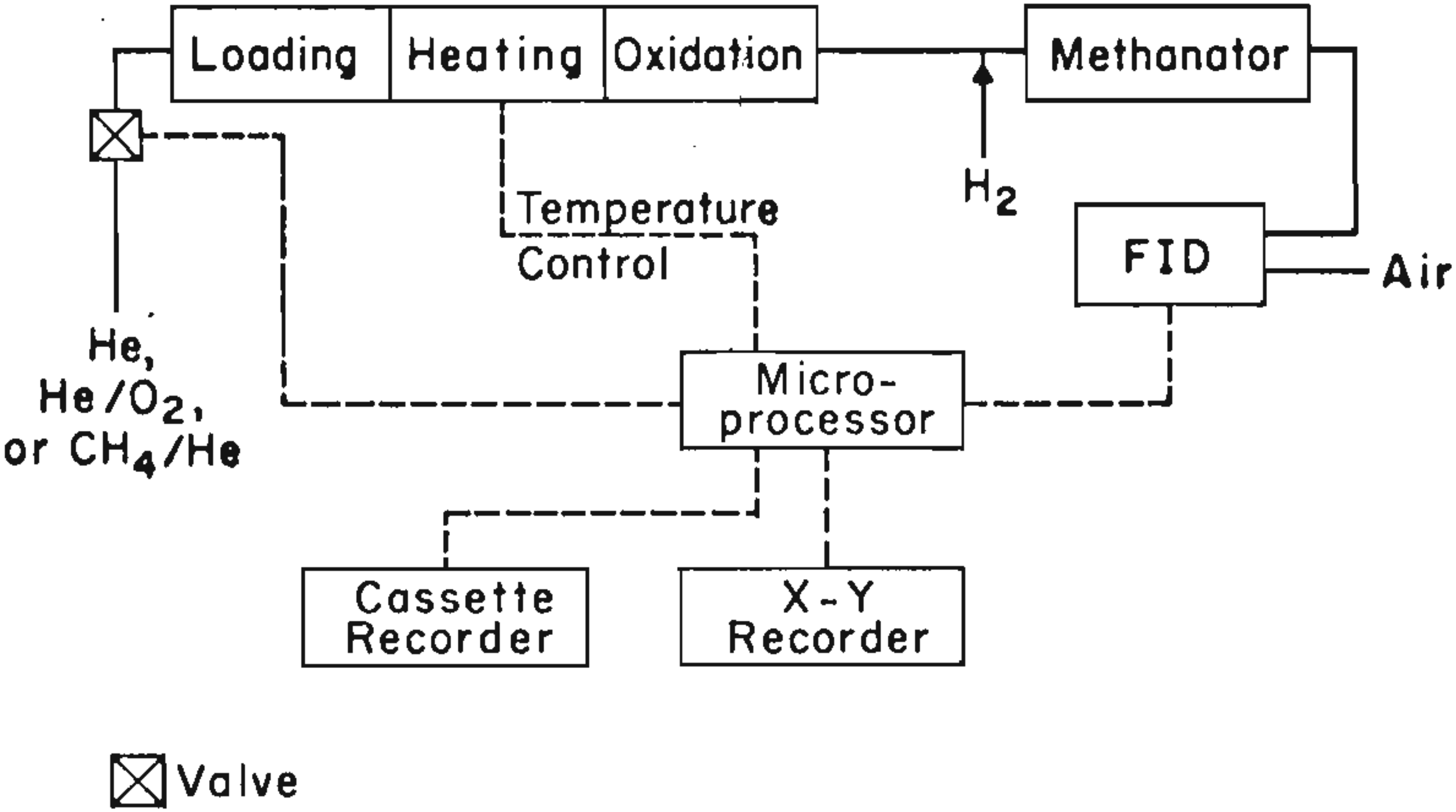


Figure 3. Block diagram of the carbon analyzer.

ally as shown by Figure 4 and also on tape for later transmission to the Prime computer system. Integration of the area under the signal peaks is also done by the microprocessor system.

Following the 350°C, 2% O_2 in helium part of the cycle, the oven atmosphere is changed to pure He and the oven temperature is raised to 600°C. This step volatilizes the higher boiling point organic compounds, and these are analyzed by the same procedure described earlier.

During the 600°C organic analysis phase of the cycle, some of the organic carbon compounds may pyrolyze to elemental carbon. As a result, some of the original organic carbon in the sample could appear as elemental carbon in the analysis results. In the OGC analysis system a correction of the pyrolysis error is made by using a laser system in which a laser beam signal is reflected from the sample while the sample is undergoing analysis. Figure 5 shows a schematic drawing of the laser system. The laser signal, which is detected by a photodetector, is proportional to the concentration of elemental carbon in the sample. During an analysis run the laser signal varies as shown by the relative reflectance curve in Figure 4. Point A, on this curve, indicates the beginning of pyrolysis in the sample. As the sample becomes blacker, because organic carbon is being converted to elemental carbon, the laser signal decreases because less of the signal is being reflected by the sample. When the oxidation of elemental carbon begins, the laser signal again increases. When the laser signal returns to its initial level, point B, it is assumed that the elemental carbon concentration in the sample has returned to its initial value at the

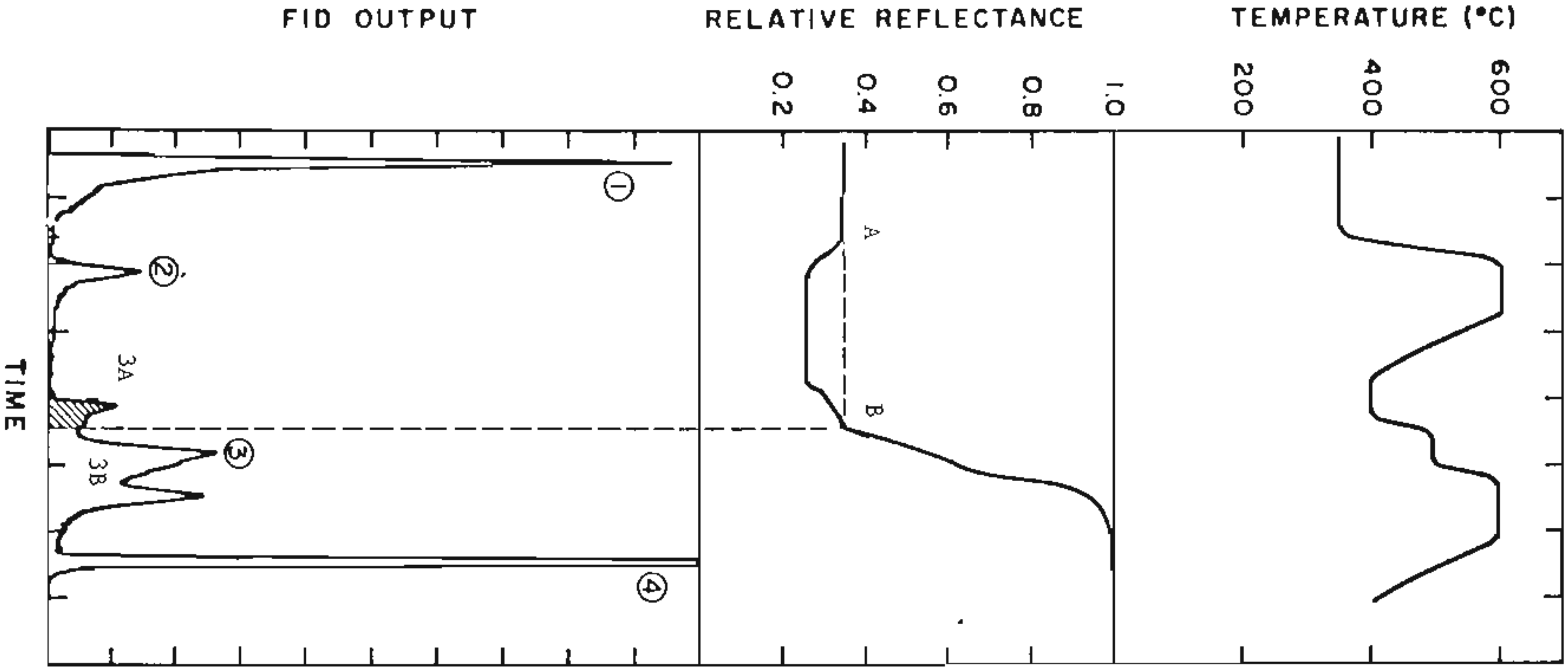


Figure 4. Graphical representation of the carbon analyzer outputs.

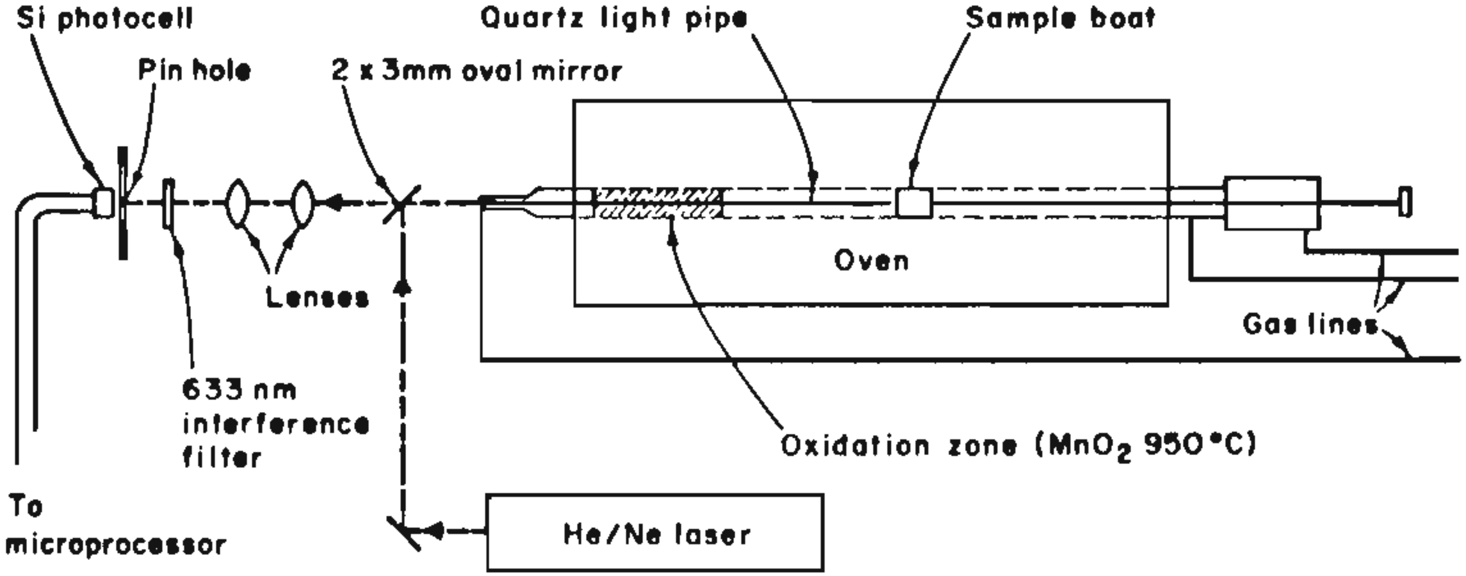


Figure 5. Schematic diagram of the laser system.

start of the analysis. Therefore, the amount of elemental carbon measured by peak (3A) in the FID output curve of Figure 4 represents the amount of organic carbon that was pyrolized in the organic analysis phase of the cycle. This amount of carbon is subtracted from the amount of elemental carbon recorded in the results and is added to the amount of organic carbon measured.

After the conclusion of the organic detection part of the analysis cycle, the oven temperature is reduced to 400°C, and the oven atmosphere is changed to 2% O_2 in helium. This begins the analysis of elemental carbon. The analysis proceeds in two more steps where the oven temperature is increased to 500 and 600°C. The temperature stepping procedure decreases the slope of the leading edge of the elemental carbon peak and aids in locating the break between peak 3A and 3B. The CO_2 produced in the elemental carbon analysis phase of the cycle is measured as before. Peak 3B in Figure 4 represents the elemental carbon content of the sample.

After the elemental analysis phase of the cycle is complete, one ml of calibration gas containing a known amount of methane is admitted to the oven. This gas is oxidized to CO_2 by the oxidation chamber and reduced to CH_4 by the methanator and is subsequently detected by the FID. Thus, the system is calibrated as part of every sample analysis. In addition, if the oxidation oven is not functioning properly so that not all the CH_4 is oxidized, then the Spherocarb column will separate the CO_2 and CH_4 and produce two output peaks in response to the calibration gas. Similar results occur if the methanator is malfunctioning.

Since carbonate carbon, which is in most cases a component of soil aerosols, is detected as organic carbon in the previously described analysis, it is necessary to correct organic carbon data for carbonate carbon. Carbonate analysis was done by a separate device where a single 1/4 cm^2 filter sample is wetted with phosphoric acid. The CO_2 generated is measured by a similar system as is used in the previously described carbon analysis. Since very few samples had even a trace of carbonate, carbonate correction of organic data was not required, and carbonate data will not be reported in this study.

2. Analysis Procedure

The OGC carbon analysis system is a fully automated system which runs under the control of a microprocessor. At the start of a day's running, the system is initialized by entering the date and setting the sample counter to zero. Communication with the microprocessor is via a conventional CRT computer terminal. To start a run, a sample (usually four 1/4 cm^2 punchings from a filter) is loaded into a sample boat through an access port. The port is then closed and the start analysis instruction is entered into the terminal. As soon as the first valve switches, which is almost immediately, the sample boat is slid into the oven. The analysis then proceeds under the control of the microprocessor. When the run is finished, a hard copy of the results is made by a plotter. The system is now ready to receive a new sample.

3. Data Handling

Output data from the carbon analysis system are recorded on tape using a Techtran cassette tape recording unit. The data tape is then entered into the Prime computer by also using a Techtran unit. Total filter area and volume of air sampled are also entered into the computer for each sample. Using these data the computer calculates μg of organic, elemental, and total carbon per m^3 of air for each sample. Carbonate data are also stored on tape by a Techtran unit and are later transmitted to the Prime computer.

C. Meteorological Data

1. Wind Data

In the ideal case one would like to have wind direction and speed data for each sampling point; however, such data are not available. Three sources of wind data were available in this study.

a. Instantaneous hourly wind speed and direction data taken by the Weather Bureau at the Medford Airport.

b. Hourly average wind speed and direction data taken by the DEQ at the Bear Creek sampling site in the southeastern corner of the airshed. Wind speed and direction were measured every six seconds and vector-summed to compute hourly averages.

c. Hourly average wind data taken by the DEQ at the Dodge Road sampling site. These data were available for only part of the sampling period.

Airport wind data were not used because these were instantaneous data and not as representative of wind behavior as average hourly data. DEQ wind speed and direction data were measured every six seconds. These data were vector-summed to compute hourly average wind speed and direction.

2. Mixing Height Data

Mixing height was determined by a procedure suggested by Holzworth (1972). In this procedure a Weather Service Pseudoadiabatic chart is used. This chart shows the vertical temperature and dewpoint profiles measured twice daily by radiosondes flown by the Weather Service. Pseudoadiatatic charts were available for radiosonde flights made at 4 AM and 4 PM at the Medford airport. The Holzworth procedure is illustrated in Figure 6. In this procedure a reference temperature point is located on the ground level altitude line on a Weather Service Pseudoadiabatic Chart. Pseudoadiabatic refers to the fact that the chart contains lines of saturated air vertical temperature profiles. The reference temperature is the local surface temperature plus a temperature correction which compensates for the urban heat island effect. In the usual applications of the Holzworth procedure, a morning mixing height is determined by using a reference temperature equal to the surface temperature at 4 AM + 5 degrees Centigrade. The 5 degree tempera-

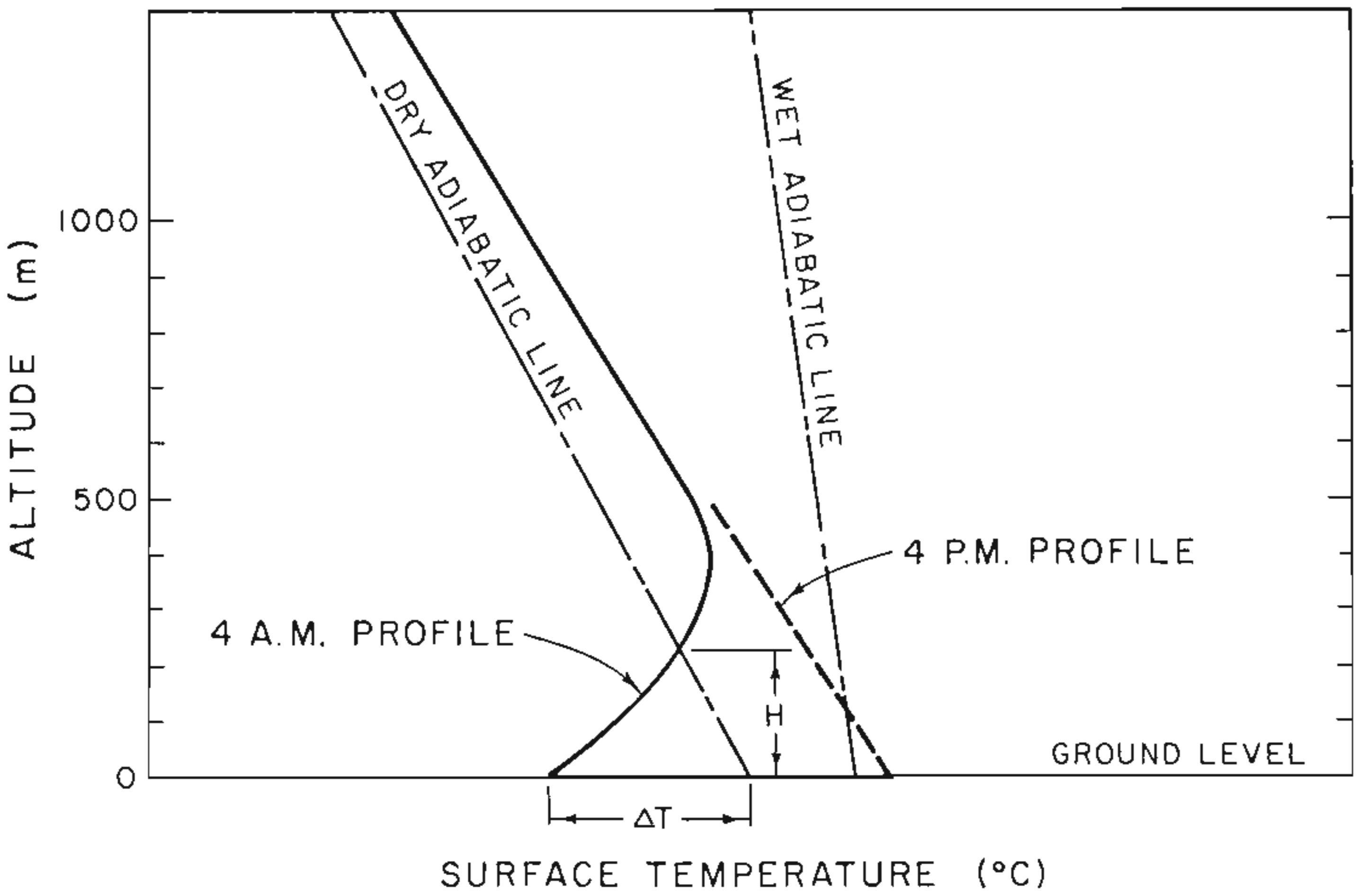


Figure 6. Typical Pseudoadiabatic Diagram

ture addition corrects for the urban heat island effect, i.e., the temperature difference between urban areas and the associated airport which is usually located outside of the urban area. The mixing height so determined represents the mixing height existing during the early morning. To determine an afternoon mixing height using the usual Holzworth procedure the reference temperature used is the highest afternoon temperature with no temperature correction. The morning vertical temperature profile is usually used for this determination even though the afternoon temperature profile is available. This is usually physically valid because the morning and afternoon temperature profiles are often identical above 1-2 km. If the afternoon temperature profile is used to determine the afternoon mixing height instead of the morning profile, somewhat lower values for the afternoon mixing heights may be obtained. In order to implement the Holzworth procedure once the reference temperature point has been located on the ground level altitude line, proceed up a dry adiabatic line until this line intersects the vertical temperature profile. The altitude of this intersection point is the mixing height as determined by the Holzworth procedure. In this study, where the Holzworth procedure yielded a mixing height of less than 100 m, a value of 100 m was arbitrarily assumed.

Mixing heights were determined at 1 AM, 4 AM, 7 AM, 10 AM, 1 PM, 4 PM, 7 PM, and 10 PM. The 1, 4, 7, and 10 AM determination used the morning temperature profile of the sampling day, the 4 PM determination used the afternoon temperature profile, and the 10 PM determination used the 4 AM temperature profile of the following day. A 5-degree temperature correction was added to the 4 AM reference temperature; likewise 3 degrees were added to the 7 AM surface temperature, and 1 degree was added to the 10 AM and 10 PM surface temperatures to obtain the respective reference temperatures.

3. Meteorological Factor Computation

Hourly mixing height and wind speed values, at 4, 7, and 10 AM and 1, 4, 7, and 10 PM, were multiplied together to form the UH product. These values were then inverted and summed for each day of the sampling period to model atmospheric dispersion for that day. The sum of these hourly average 1/UH values is a factor which is proportional to the meteorological dispersion of pollution for the given day. If Q were known, within the assumptions of the box model, these values could be substituted into Eqn. (5), and the average pollutant concentration measured by a 24-hour sample could be computed.

III. DISCUSSION OF THE DATA

A. The Role of Carbon in Aerosol Air Pollution

Carbon is known to be one of the larger components of urban aerosols. Therefore, in the study of these aerosols it is important to determine how much carbon aerosols contain and how this quantity varies over time. Information of this kind is essential to the understanding of the types of processes or emission sources that create aerosol pollution. This study examined specifically the following points in relation to Medford aerosol air pollution.

1. The amount of carbon found in the fine and total mass fractions of ambient aerosol.
2. The fractional part contributed by carbon to the fine and total mass fraction of ambient aerosol.
3. The composition of carbon in terms of organic and elemental carbon.
4. The form of a model that describes ambient carbon concentrations in terms of meteorological dispersion and the magnitude of space heating emissions as characterized by heating degree day values.

Items 1 and 2 discuss the magnitude of the carbon component of ambient aerosols in the test area. It will be shown that carbon is a major contributor to aerosol mass. Item 3 will divide the aerosol carbon component into two classes of carbonaceous material, organic carbon and elemental carbon, in order to further facilitate the understanding of the sources of carbon in aerosols. Item 4 then demonstrates that ambient carbon concentrations can be well described in terms of meteorological dispersion and two carbon emission terms, one describing space heating emissions and one describing non-space heating emissions. While data were collected at three sampling sites in the study, the major part of the data analysis concerned data gathered at the urban site (the Justice Building) because the sources of urban aerosol pollution, and especially the role of wood stove emissions in this pollution, were a major focus of this study. At this site space heating appears to be a major contributor to the ambient carbon concentrations. At the industrial site (White City) and at the background site (Dodge Road) emissions which are not correlated with degree day values play a larger role so that the role of space heating is consequently reduced.

In this study the aerosol sampled by a standard high volume sampler was called the total aerosol. The aerosol which passed a Sierra 2 μm cutpoint impactor attached ahead of a standard high volume sampler was called the fine aerosol, and the difference obtained by subtracting the fine aerosol from the total was called the coarse aerosol. The term carbon was used to mean the sum of organic and elemental carbon. Organic and elemental carbon are defined by the analysis procedure.

Figures 7-9 show the carbon in total and fine aerosol data at the three sampling sites. These graphs show that most of the time half

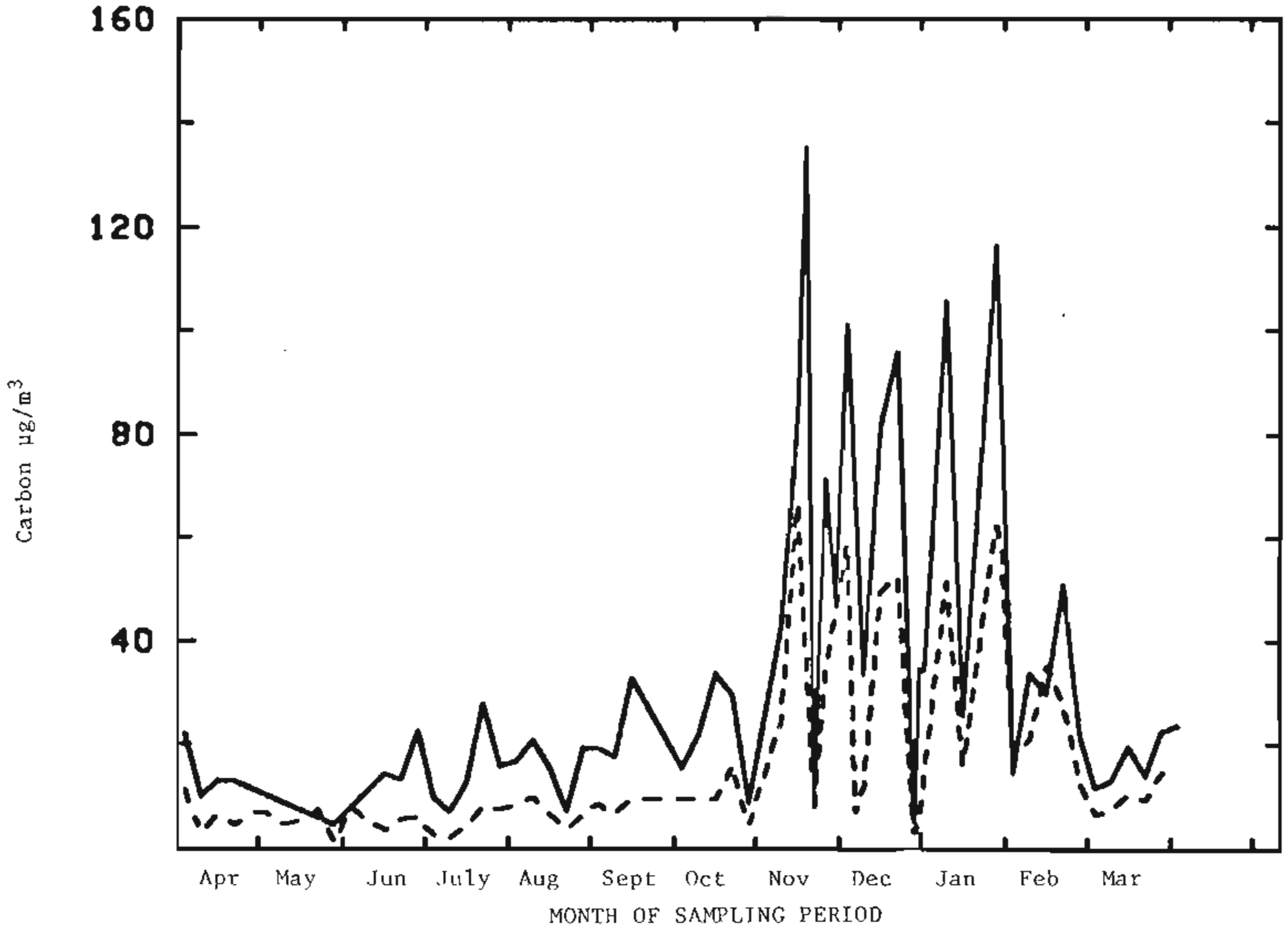


Figure 7. Time series of Medford Justice Building ambient total and fine carbon concentrations. Line = total aerosol; dash = fine aerosol. April 1979 to March 1980 data.

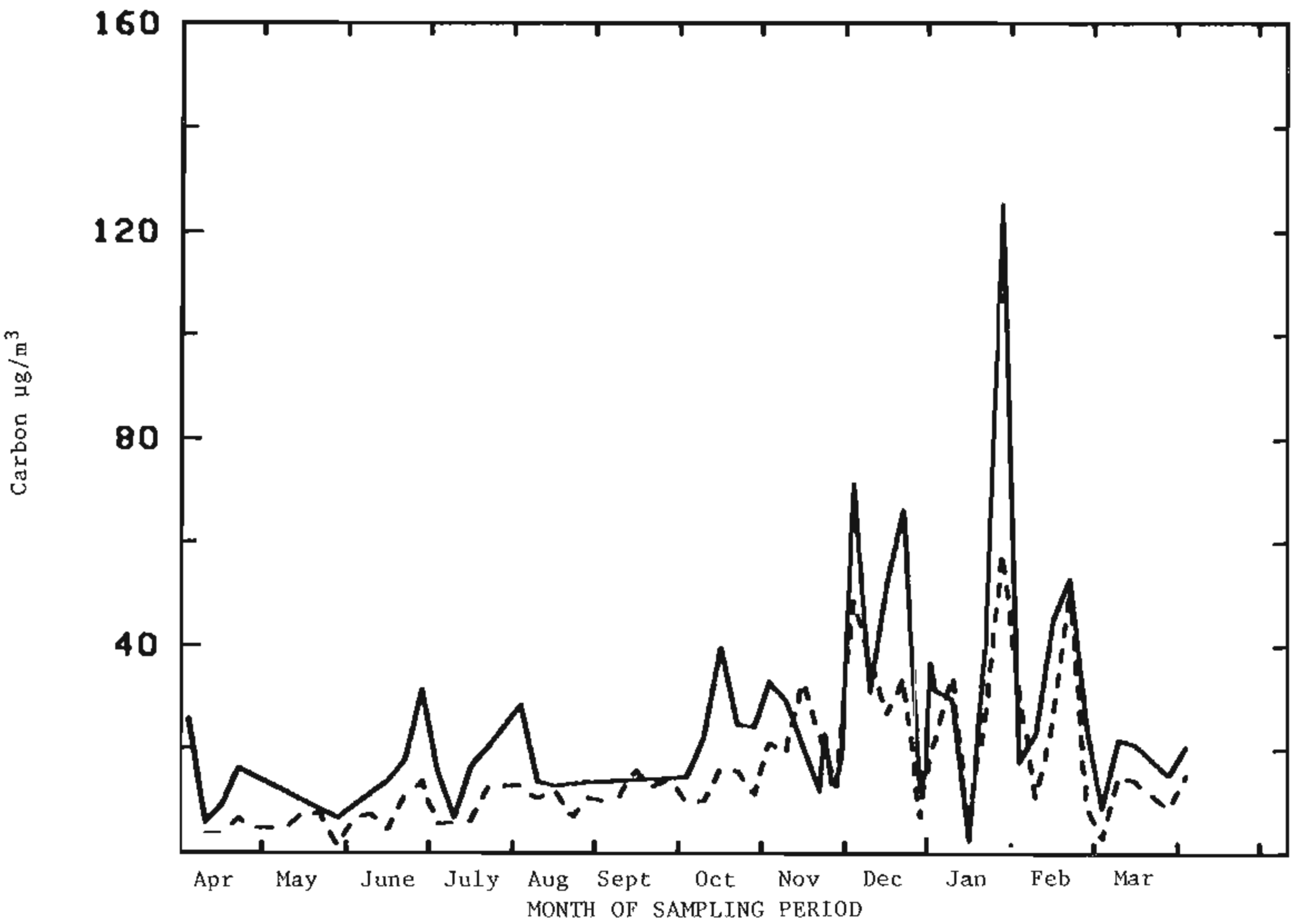


Figure 8. Time series of White City ambient total and fine carbon concentrations. Line = total aerosol; dash = fine aerosol. April 1979 to March 1980 data.

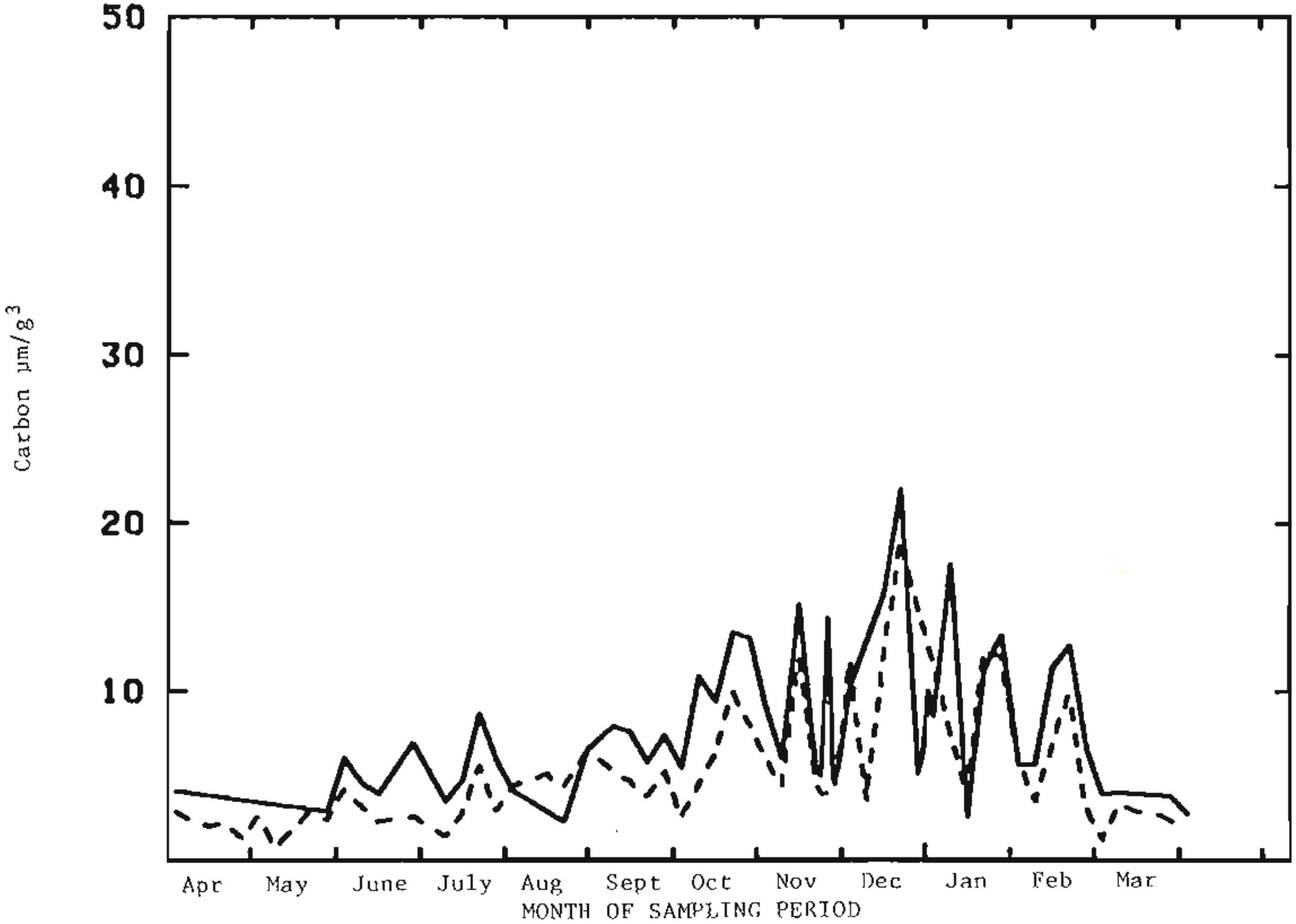


Figure 9. Time series of Dodge Road ambient total and fine carbon concentrations. Line = total aerosol; dash = fine aerosol. April 1979 to March 1980 data.

or more of the aerosol carbon is found in the fine aerosol mode and that both the total and fine aerosol fraction carbon levels are much higher many times during the winter than their average value during the summer. Figures 10-12 show the fractional part contributed by carbon to total and fine aerosol. From 20 to 60% of Medford aerosol air pollution is composed of carbon. The curve for fine aerosols at the Dodge Road site is omitted because the low levels of mass collected at this site have large attendant mass errors.

To gain further insight into the nature of aerosol carbon and the sources which emit carbonaceous aerosols, the aerosol carbon data collected at the Justice Building were subdivided into fine and coarse fractions, and then were further subdivided into organic and elemental carbon. Until very recently (Environmental Science and Technology, December 1980), elemental carbon has received very little attention as an air pollutant, not because it is unimportant but because its quantification is method-dependent. The two common quantification methods are solvent extraction and selective volatilization. In the solvent extraction method, the aerosol carbon fraction remaining after solvent extraction is assumed to be elemental carbon. In the selective volatilization method, which is the method used at OGC, gradual heating first volatilizes organic carbon and subsequently the elemental carbon (see the Experimental Procedure Section). Elemental carbon is formed in most combustion processes. Its formation is favored by low combustion temperatures and low excess oxygen. Carbon aerosols play a sig-

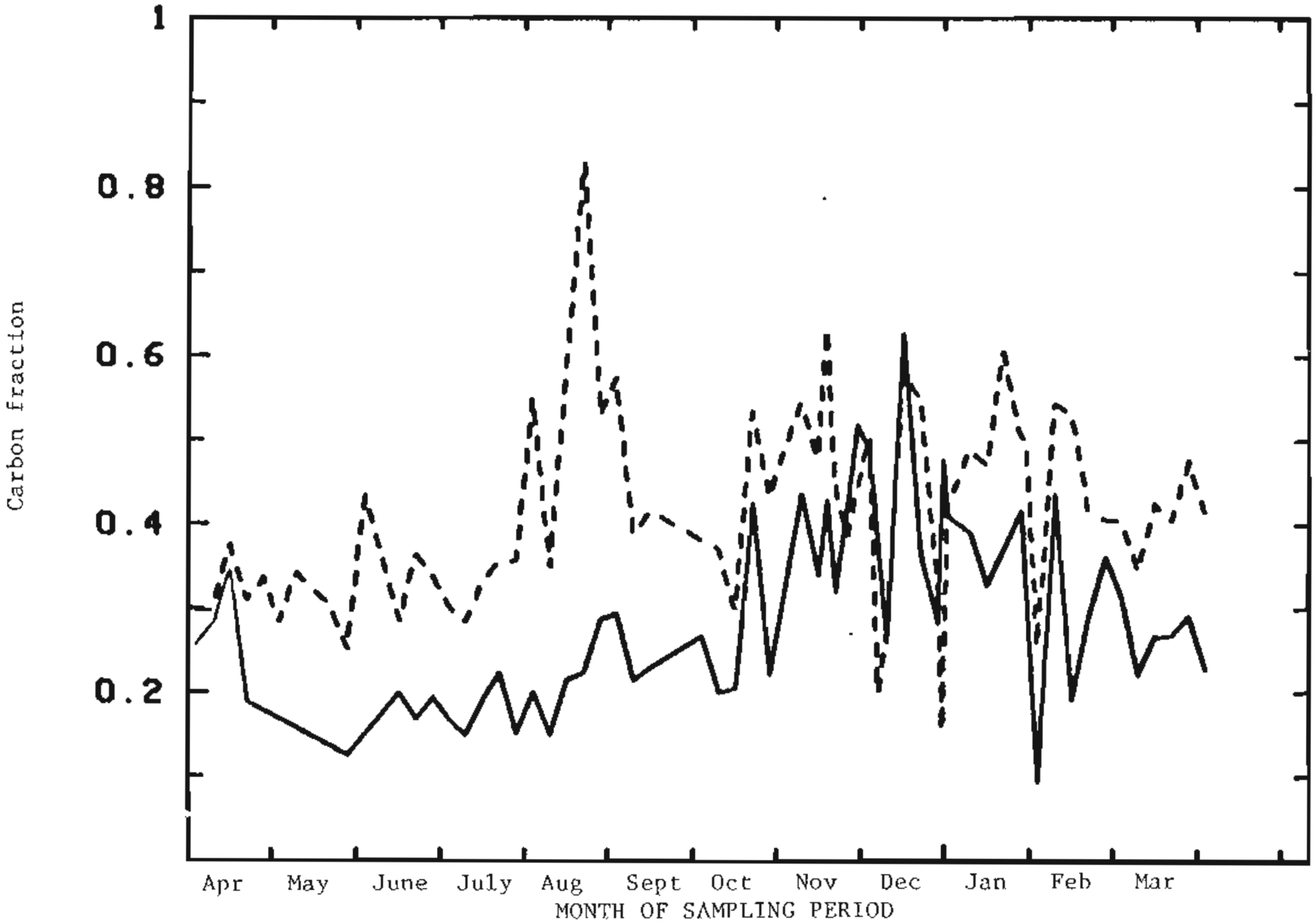


Figure 10. Time series of Medford Justice Building carbon fraction for total and fine aerosols. Line = total aerosol carbon fraction; dash = fine aerosol carbon fraction. April 1979 to March 1980 data.

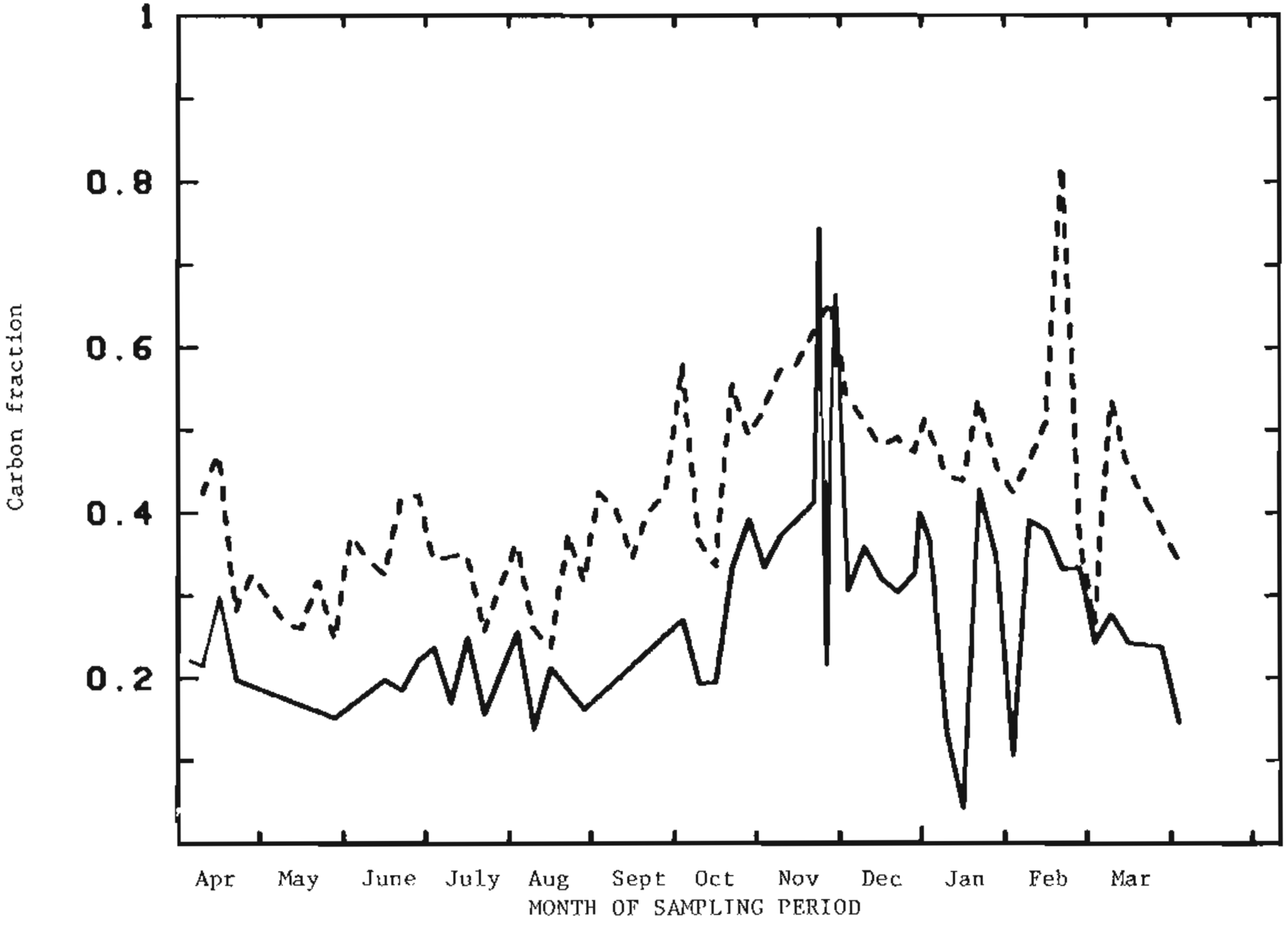


Figure 11. Time series of White City carbon fraction. Line = total aerosol carbon fraction; dash = fine aerosol carbon fraction. April 1979 to March 1980 data.

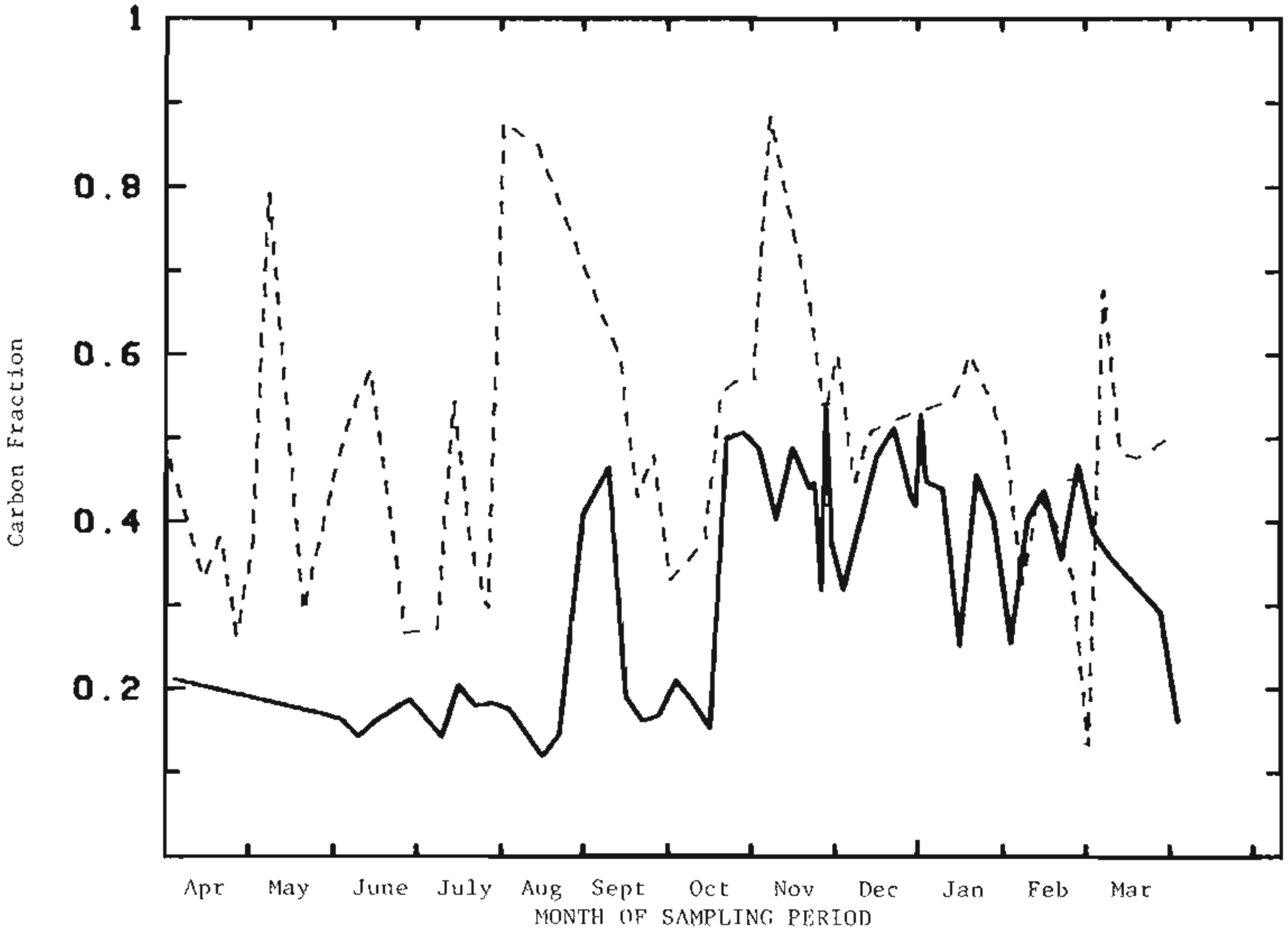


Figure 12. Time series of Dodge Road carbon fraction. Line = total aerosol carbon fraction; dash = fine aerosol carbon fraction. April 1979 to March 1980 data.

nificant role in visibility reduction and may act as catalysts in various atmospheric secondary chemical reactions (Freiberg, 1978; Cahill, 1978).

Figures 13 and 14 show a time series of Justice Building fine and coarse organic and elemental carbon concentrations, respectively. Figure 13 shows that for fine aerosols, elemental carbon concentrations are slightly less than half as great as organic carbon concentrations, i.e., the elemental carbon to organic carbon ratios of summer emission sources are very similar to the values of this ratio for winter emission sources. Figure 14 shows that for coarse aerosols, though the elemental carbon content is usually lower than the organic carbon content, the values are often quite close. Figure 15 shows the fraction of elemental carbon in fine aerosol is only slightly higher in the winter than in the summer; however, average organic carbon concentrations are significantly higher in the winter than in the summer. Because residential wood smoke contains a higher fraction of carbon than most other sources of ambient carbon aerosol, an increase in the carbon mass fraction would be expected to result from wood burning during the winter. For the Justice Building fine aerosol data, the average summer ratio of elemental carbon to organic carbon is 0.36 ± 0.04. In the winter this value increases to 0.43 ± 0.04. Watson (1979) indicates that wood stove fine mass emissions are 0.57 ± 5% carbon and distillate oil fine mass emissions are 0.36 ± 23% carbon. Therefore, oil space heating would not be expected to cause a large change in the winter elemental

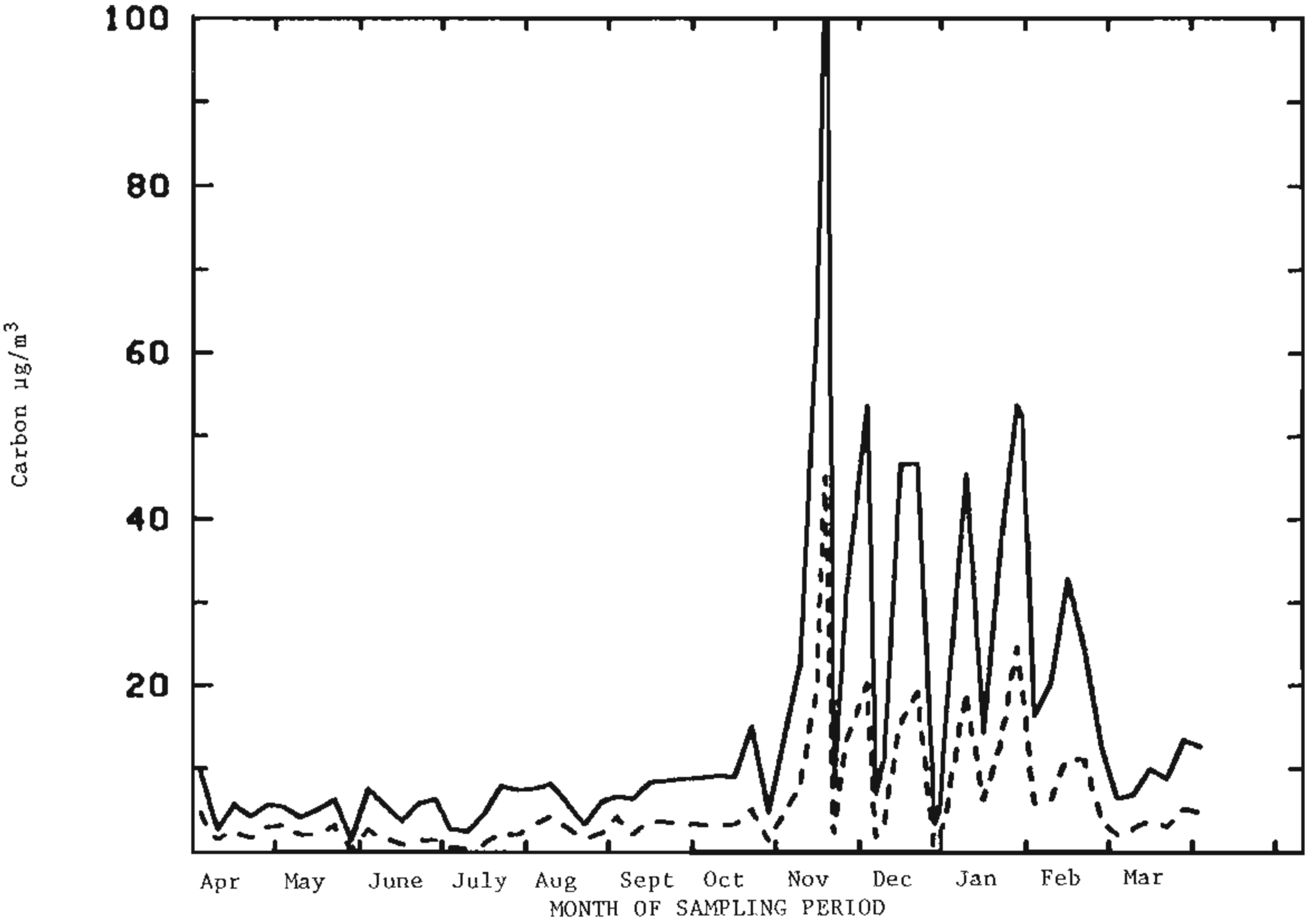


Figure 13. Time series of Justice Building fine aerosol carbon. Line = organic carbon; dash = elemental carbon.

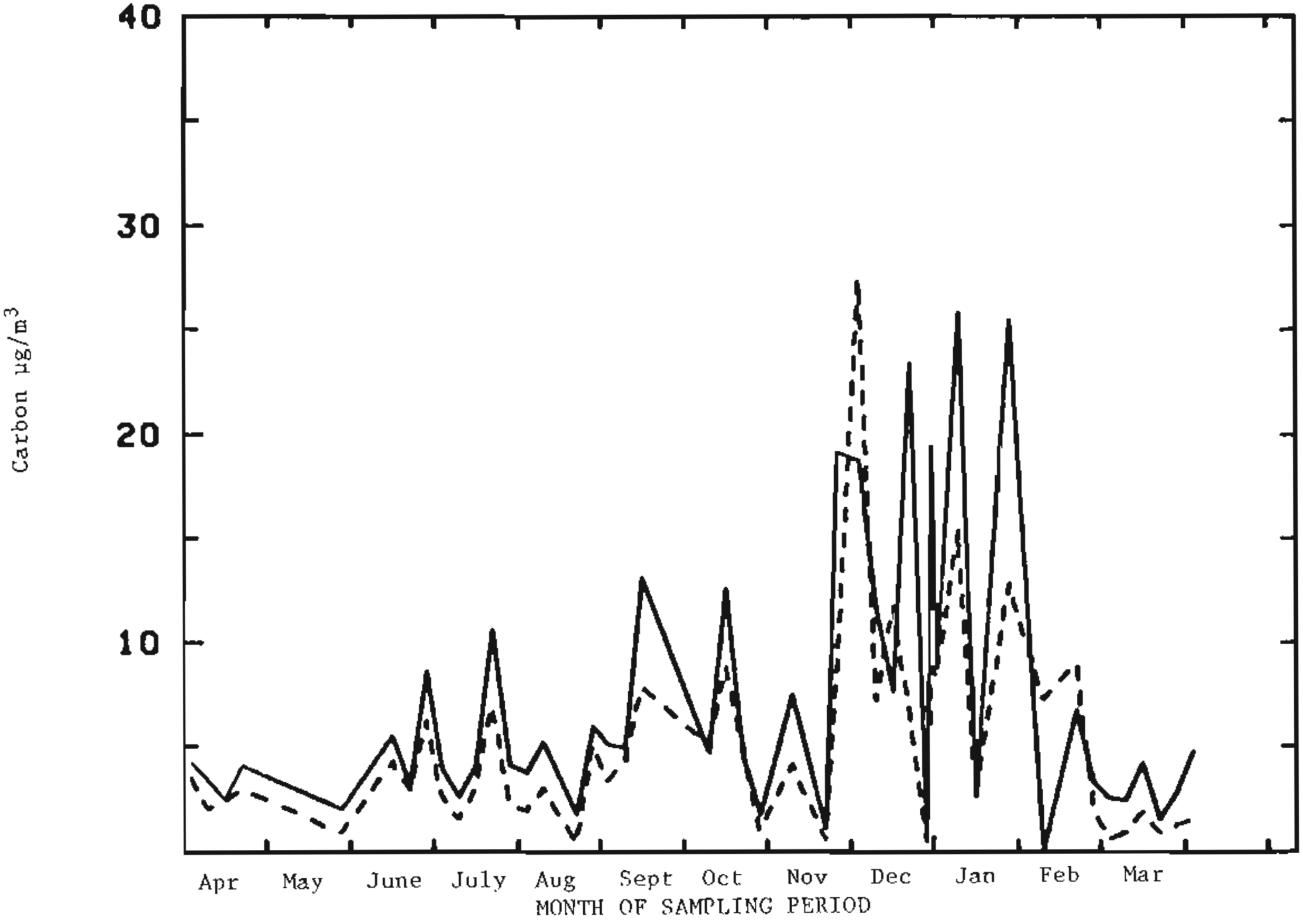


Figure 14. Time series of Justice Building coarse aerosol carbon. Line = organic carbon; dash = elemental carbon.

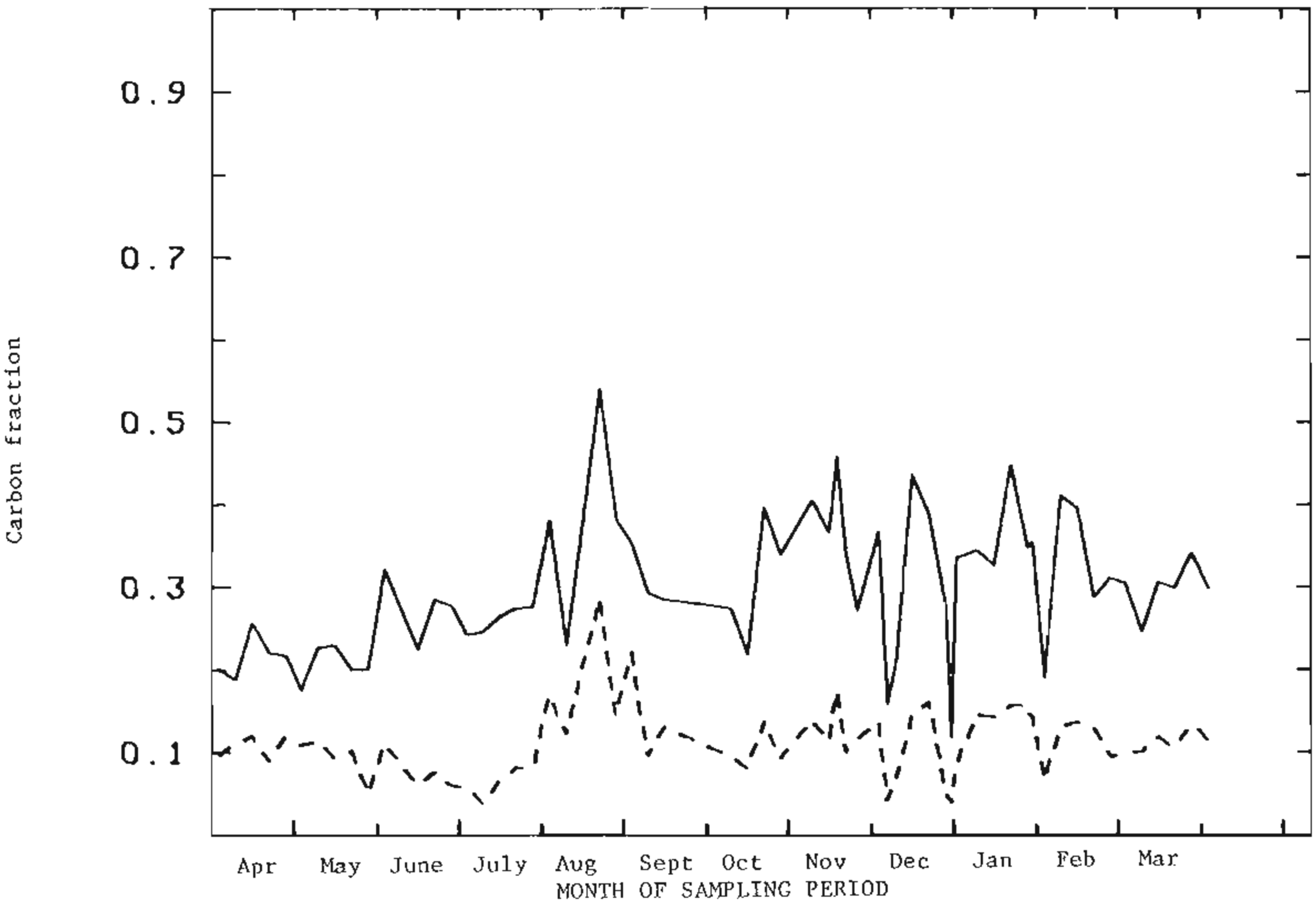


Figure 15. Time series of fine aerosol carbon composition. Line = organic carbon/aerosol mass; dash = elemental carbon/aerosol mass.

carbon to organic carbon ratio, provided that the non-space heating emission inventory is basically the same in the winter as it is in the summer. Wood space heating, however, could cause the ratio to increase.

Figure 16 shows that for coarse aerosol at the Justice Building the organic and elemental carbon fraction of coarse aerosol mass increases during the winter season. This probably does not result from increased emissions of coarse carbon during the winter, but rather from a decrease of coarse mass emissions during the winter. Data to be presented later in this study support the hypothesis that the emission of coarse aerosols is reduced in the winter. This effect may result from winter rains reducing soil resuspension.

In order to search for relationships between space heating emissions and ambient carbon aerosol levels, the summer season was defined as April through September and the winter season was defined as October through March. The average and 95% confidence limits of the mean of carbon values for these seasons are shown in Table 1. In this and the following tables, OC = organic carbon, EC = elemental carbon, and C = OC + EC.

Table 1 shows that average carbon and mass levels, at all sites, are higher in the winter than in the summer. This difference can result from some combination of increased wintertime emissions and poorer wintertime than summertime dispersion conditions. In the following sections, the increase in wintertime ambient pollution levels due to poorer wintertime dispersion conditions will be separated from the increases due to increased emissions attributable to space heating.

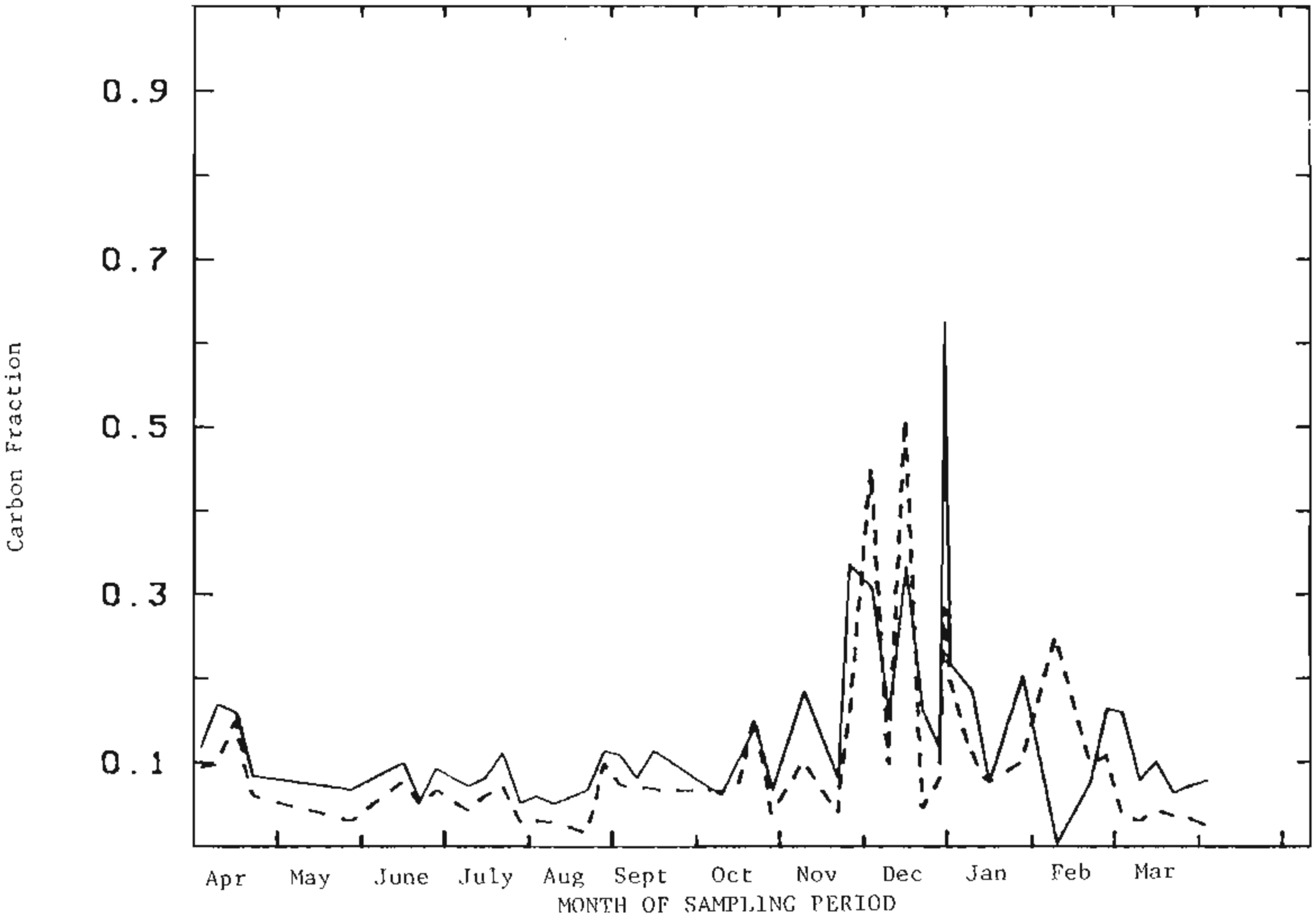


Figure 16. Time series of coarse aerosol carbon composition. Line = organic carbon/ aerosol mass; dash = elemental carbon/aerosol mass.

Table 1

Summer and Winter Seasonal Carbon and Mass Averages
($\mu g/m^3$)

(S = Summer = April-September. W = Winter = October-March)

	Ave. OC	Ave. EC	Ave. TC	Ave. Mass
Medford Justice Building Site				
S. Total	10 ± 2	6 ± 1	16 ± 3	76 ±13
W. Total	29 ± 9	14 ± 4	42 ±13	123 ±28
S. Fine	6 ± 0.8	2 ± 0.4	8 ± 1	22 ± 3
W. Fine	25 ±10	9 ± 3	35 ±13	73 ±21
White City Site				
S. Total	10 ± 2	6 ± 1	16 ± 3	78 ±16
W. Total	20 ± 5	11 ± 3	31 ± 8	105 ±25
S. Fine	6 ± 0.8	2 ± 0.4	9 ± 2	26 ± 5
W. Fine	16 ± 4	6 ± 1	22 ± 5	44 ±10
Dodge Road Site				
S. Total	4 ± 0.4	1 ± 0.2	6 ± 0.9	28 ± 4
W. Total	7 ± 1	2 ± 0.3	9 ± 2	24 ± 5
S. Fine	3 ± 0.4	1 ± 0.2	3 ± 0.4	7 ± 1
W. Fine	5 ± 1	1 ± 0.4	7 ± 2	13 ± 3

B. Meteorological Dispersion Characterization and Data Normalization

Two independent measures of dispersion strength were used and their effectiveness evaluated. First 1/UH was used because it is purely a meteorological entity. Then ambient lead concentrations were used to evaluate dispersion as well. The rationale for this procedure is discussed in Section D. The dispersion factors, 1/UH and ambient lead concentrations, were also used to normalize ambient pollution data. Normalization determined the ambient pollution values that would have existed under constant dispersion conditions. Ambient data were normalized by multiplying them by a normalization factor defined by Eqn. (11).

$$\text{Normalization Factor} = \frac{\text{Average Dispersion Factor Values}}{\text{Daily Dispersion Factor Value}} \qquad (11)$$

C. Seasonal Patterns of 1/UH

Figure 17 shows the variation of average daily 1/UH values over the course of the sampling period. This figure shows that during the fall and winter seasons the values of 1/UH are often much higher than during the spring and summer. Since under the assumptions of the box model ambient pollution concentrations, for constant emission rates, are proportional to 1/UH, winter ambient pollution levels will be higher than summer levels due to meteorological dispersion effects alone.

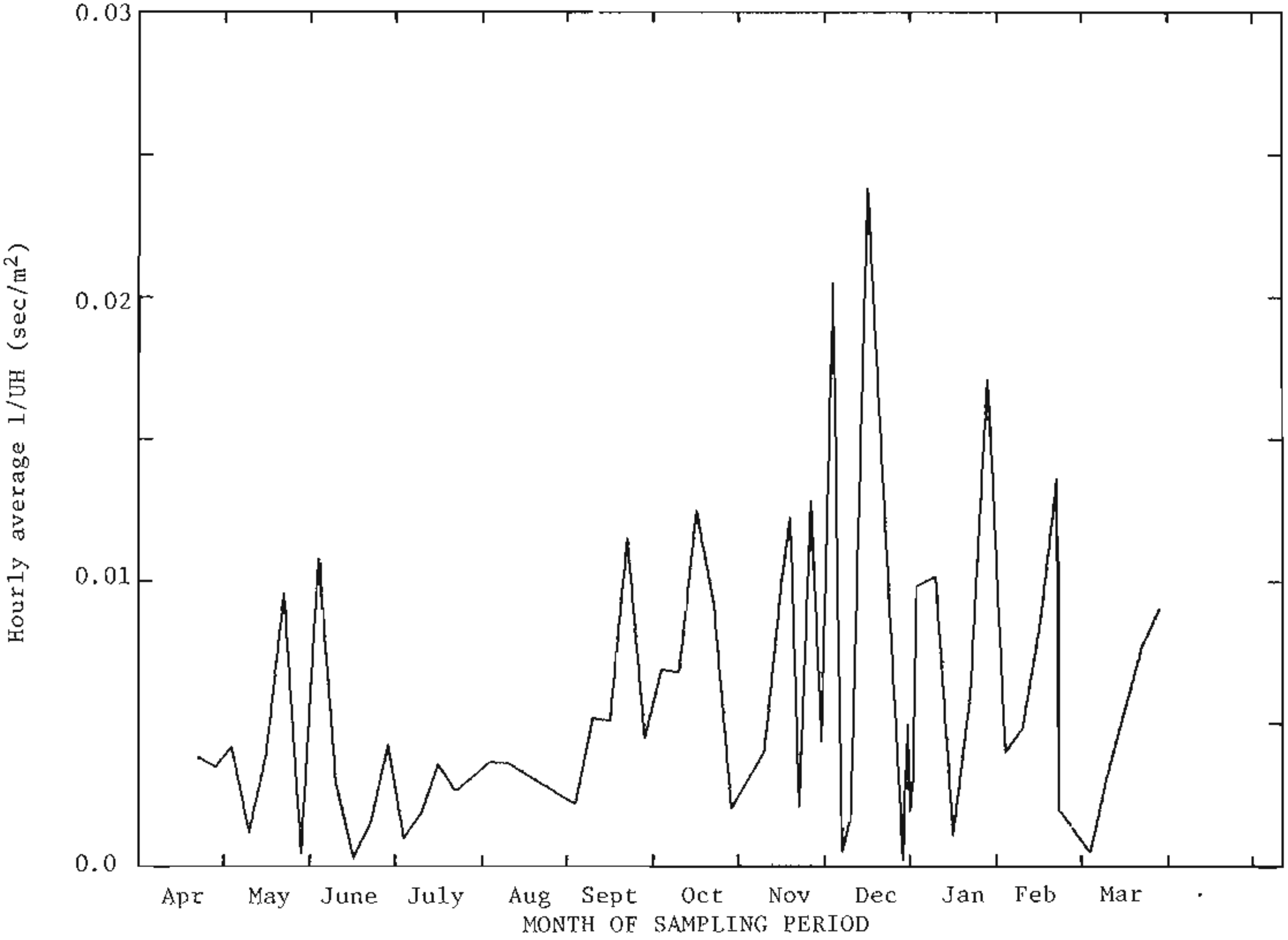


Figure 17. Time series of 1/UH.

Table 2 gives the seasonal averages of the dispersion factors generated in this study. This table indicates that due to dispersion alone, as characterized by 1/UH, average winter ambient pollution levels should be twice as high as summer values. In terms of the box model, this means that during winter, on the average, there is only half as much air volume available for pollutants to disperse into as there is during summer.

Table 2

Dispersion Factor Seasonal Averages

1979-1980	Hourly Average 1/UH (sec/m^2)
Apr., May, June	0.00382
July, Aug., Sept.	0.00428
Oct., Nov., Dec.	0.00738
Jan., Feb., Mar.	0.00723
Annual Average	0.00554

D. The Use of Ambient Lead Concentrations as a Measure of Meteorological Dispersion.

In the previous section 1/UH was used as a measure of meteorological dispersion. By comparing the complexity of aerosol mixing by turbulent diffusion in the real atmosphere with the simplicity of 1/UH and the box model, it seems reasonable to question how well 1/UH characterizes meteorological dispersion. Since no direct measure of meteorological dispersion was available in this study, an alternative procedure to evaluate the validity of 1/UH was to search for a scheme which might be a better measure of meteorological dispersion. Comparison of 1/UH to this better scheme could then be used to evaluate the effectiveness of 1/UH. Of course, the better scheme could also then be used to model meteorological dispersion.

Meteorological dispersion can be evaluated by measuring the ambient concentration of a tracer species which is emitted at a constant rate. Ambient concentrations of such a species are an exact measure of the dispersion conditions that existed between the emission source and the receptor. Since no specially emitted tracer species was available in this study, a search was made for an elemental species that appeared to be emitted at a relatively constant rate. Lead, which is emitted almost exclusively by automobiles, was found to have a reasonably constant average emission rate. This was shown by comparing summer and winter weekday and weekend average normalized emission values. In addition to being emitted at a relatively constant rate, for ambient lead concentrations to be useful in evaluating meteorological dispersion, automotive emissions containing lead must be a negligible source of carbon in the airshed. If automotive emissions were a large source of carbon, then correlations of ambient lead and carbon values would confound emission commonality with dispersion commonality. The results of Chemical Element Balance (CEB) (DeCesar and Cooper, 1981) studies and emission inventory data show that automotive emissions comprise only a few percent of Medford aerosol emissions. Therefore, they contribute on the order of 1% of the carbon to Medford aerosols.

For ambient lead concentrations to be a better measure of dispersion than 1/UH it is required that they give similar results to those obtained using 1/UH and that the associated random error be less than the random error associated with the use of 1/UH. In the following sections it will be shown that both 1/UH and ambient lead concentrations effectively model meteorological dispersion, but that a smaller amount of random dispersion is associated with the lead model.

The limitations of using ambient lead concentrations can be divided into two categories: (1) limitations specific to lead, and (2) limitations that apply to the use of a dispersion tracer species generally. In the first category the only limitation concerns whether lead is emitted at a constant rate. When lead is used as a measure of dispersion, a change in the ambient lead concentration that results from a change in lead emissions cannot be distinguished from one that results from a change in dispersion conditions. Lead aerosol is not expected to engage in atmospheric chemistry that would convert it into the gas phase; therefore, it has the required aerosol stability to make it a useful tracer species. In the second category two limitations exist. First the dispersion tracer species should have similar aerodynamic size to the pollutant whose dispersion is being evaluated. Practically, dispersion modeling is most suitable for fine aerosols (less than 2 μm) where sedimentation due to gravity can be neglected. In this study fine aerosol lead concentrations are used as a dispersion modeling species and as such are strictly applicable to fine carbon aerosol only. The second limitation in this category involves the geographic location difference between sources of the aerosol of interest and sources of the dispersion tracer species. For fine lead aerosol to be a useful dispersion tracer its emission source locations should be similar to the locations of the emitters of fine carbon. This limtation is probably a severe restriction on the use of lead as a dispersion model in many urban areas; however, in Medford the broad valley nature of the area homogenizes dispersion over the area and thus seems to weaken the effect of this limitation.

In a discussion of the limitations on the use of ambient lead concentrations as measures of dispersion it might be pointed out that a limitation on both the methods used to measure dispersion in this study is some lack of coincidence in time between the evaluation of the dispersion measure and emissions. Ideally the dispersion parameter should be integrated only over the time periods when the emission source is operating. If an emission source has a certain diurnal emission pattern, the ideal dispersion tracer species would have the same diurnal emission pattern and would thereby experience identical dispersion. This limitation may not be significant for lead because the diurnal automotive traffic level probably correlates roughly with the diurnal pattern of many other emission sources. However, it may be a source of error in the use of 1/UH because 1/UH as evaluated in this study is a 24-hour average. It does, nevertheless, not appear to be a very large source of error.

E. Dispersion Normalized Pollutant Concentrations

The seasonal ambient carbon averages given in Table 1 clearly indicate that ambient carbon concentrations are significantly greater in the winter than in the summer. However, from this table one cannot determine what part of the summer-winter difference is due to differences in meteorological dispersion and what part is due to differences in emissions. In this section the normalization factor defined by Eqn. (11) will be used to normalize ambient carbon and mass data. Since the normalization procedure corrects ambient data to what they would have been under conditions of constant meteorology, normalized data are proportional to emissions, within the errors introduced by the normalization procedure itself. Seasonal averages of normalized data are thus proportional to seasonal differences in emissions. Both $1/UH$ and ambient lead concentrations will be used to compute normalization factors. The data variations that still exist after data normalization are due to variations in emission rates, meteorological dispersion variations not described by the box model, and to experimental error.

By comparing the summer and winter average values of normalized data the changes in summer and winter average emissions are really being compared. Table 3 shows the summer and winter, weekend and weekday average and 95% confidence limit values for fine aerosol carbon and mass data from the Justice Building normalized with $\frac{1}{UH}$.

Table 3

Justice Building 1/UH Normalized
Fine Average Ambient Carbon and
Mass Concentration ($\mu g/m^3$)

		# Points	Summer	# Points	Winter
OC	WE(Weekend)	3	7 ± 6	8	15 ± 7
	WD(Weekday)	15	11 ± 3	18	22 ± 7
	All Data	18	11 ± 2	26	20 ± 6
EC	WE	3	2 ± 6	8	5 ± 2
	WD	15	5 ± 1	18	8 ± 2
	All Data	18	4 ± 1	26	7 ± 2
Carbon	WE	3	9 ± 0	8	21 ± 9
	WD	15	16 ± 4	18	30 ± 6
	All Data	18	15 ± 4	26	28 ± 8
Mass	WE	3	28 ± 9	8	48 ± 16
	WD	15	43 ± 11	18	75 ± 27
	All Data	18	41 ± 9	26	67 ± 20

Table 3 shows that the normalized average fine aerosol carbon values increase by about 100% during the winter as compared to summer values. Summer and winter carbon averages are different at least at the 95% confidence level as established by a t-test. The average ambient normalized fine carbon values obtained by combining weekend and weekday data increase by 13 $\mu g/m^3$ in the winter. It can be hypothesized that this increase is due to space heating because space heating is a known carbon aerosol emission source. Fine carbon emissions increase during colder weather. Chemical Element Balance (CEB) results (DeCesar and Cooper, 1981) show that normalized average fine ambient carbon levels attributable to vegetative burning increase by 11.4 $\mu g/m^3$ from the spring/summer season to the fall/winter season. Within the limits of experimental error and under the assumption that spring/summer sources of vegetative burning do not vary over the course of the year, this result supports the hypothesis that the summer to winter increase in the average normalized fine carbon level is due not only to space heating but to space heating with wood.

When fine aerosol carbon and mass data from the Justice Building were normalized using ambient lead concentrations, the results shown in Table 4 were obtained.

The results shown in Table 4 are quite similar to those shown in Table 3. Summer and winter carbon averages are different at least at the 95% confidence level as established by a t-test. Comparisons are best made using relative changes rather than comparing absolute magnitudes because the average value of 1/UH may not represent the exact same dispersion condition as the average value of ambient lead concentrations.

Table 4

Justice Building Lead Normalized Fine Aerosol Carbon and Mass Averages (μg/m^3)

		# Points	Summer	# Points	Winter
OC	WE(Weekend)	6	8 ± 3	7	17 ± 5
	WD(Weekday)	16	9 ± 2	14	17 ± 4
	All Data	22	9 ± 1	21	17 ± 3
EC	WE	6	3 ± 2	7	5 ± 2
	WD	16	4 ± 1	14	6 ± 1
	All Data	22	4 ± 1	21	6 ± 1
Carbon	WE	6	11 ± 4	7	22 ± 6
	WD	16	13 ± 3	14	24 ± 6
	All Data	22	12 ± 2	21	23 ± 4
Mass	WE	6	28 ± 13	7	51 ± 17
	WD	16	38 ± 9	14	53 ± 12
	All Data	22	35 ± 8	21	52 ± 9

The Clean Air Act of 1970 requires that the annual geometric average of total aerosol concentration values not exceed 75 μg/m^3. In Medford, aerosol concentrations satisfy this standard in the summer but not in the winter. While it is useful to reduce both summer and winter emissions, one strategy to attack the compliance problem is to ask what reduction in wintertime emissions alone would be required to bring the area into compliance. The geometric average of wintertime total aerosol concentrations is 102 μg/m^3. Therefore, a 26% reduction of wintertime ambient total aerosol concentrations is required to satisfy the Clean Air Act of 1970. Under the assumption that winter meteorological dispersion is similar from year to year, a 26% reduction in emissions will be required for compliance. The average winter normalized ambient concentration is 130 μg/m^3 (from Table 5). Since normalized average aerosol concentration values are proportional to emissions, a 26% reduction (34 μg/m^3) in this value is required to satisfy the Clean Air Act. The normalized average ambient fine carbon increase from wummer to winter is 13 μg/m^3 (from Table 3). Therefore, space heating emissions form 13/0.57 x 34 = 67% of the total reduction required in emissions. Thus, to satisfy the Clean Air Act other emission reductions besides the elimination of residential wood burning will be required.

In the previous paragraphs normalized average ambient values of fine aerosol carbon and mass were presented to determine the weekend, weekday, and seasonal variations in emissions of these species. However, from a regulatory viewpoint only total aerosol values are currently of interest. One of the reasons that Medford fails the requirements of the Clean Air Act is that the annual geometric average of total aerosol is greater than the allowed 75 μg/m^3. Earlier in this section it was stated that a 26% reduction of wintertime emissions would be required to bring the Medford airshed into compliance with the Clean

Table 5

Justice Building Total 1/UH Normalized Average
Ambient Aerosol Concentrations
(μg/m^3)

	Summer	Winter
Weekend		
Number of Points	4	9
OC	13 ± 3	22 ± 8
EC	6 ± 1	11 ± 3
Carbon	19 ± 4	33 ± 11
Mass	105 ± 36	95 ± 20
Weekday		
Number of Points	9	18
OC	28 ± 6	32 ± 10
EC	15 ± 3	15 ± 5
Carbon	42 ± 10	47 ± 16
Mass	215 ± 56	148 ± 53
All Data		
Number of Points	13	27
OC	23 ± 6	28 ± 8
EC	12 ± 3	14 ± 4
Carbon	35 ± 9	42 ± 11
Mass	181 ± 49	130 ± 37

Air Act. This means that a 26% reduction in normalized wintertime ambient mass average pollution concentration level will be required to accomplish this reduction.

In order to examine the relative emission levels of total aerosol carbon and mass, seasonal weekend and weekday normalized averages were evaluated for total aerosol data from the Justice Building site. These are shown with 95% confidence limits in Table 5.

Table 5 shows that total aerosol carbon increases considerably on winter weekends and slightly on winter weekdays, as compared to summer comparable days. Since Tables 3 and 4 show that fine carbon emissions increase both on winter weekends and on winter weekdays, the fact that total carbon hardly increases on winter weekdays implies that coarse carbon emissions decrease during winter. A decrease in coarse emissions during the winter may result from the suppression of wood dust and other coarse carbon resuspension by winter rains. Since coarse dust emissions may result mostly from industrial activity, the rain suppression effect may be most notable on weekdays. Because of the confounding effect caused by the winter rain associated removal of coarse aerosol, the data of Table 5 are not useful for drawing conclusions about residential heating emissions. Table 5 also shows that total aerosol mass is significantly decreased during the winter. This is probably also due to rain-suppression of coarse soil aerosols. If it were not for the rainfall in the Medford area, the wintertime aerosol pollution would be much worse that it is. Table 5 also shows that weekday emissions are considerably greater than weekend emissions, indicating that commercial and industrial activity causes a large part of emissions. Medford emission inventory data show that most large industries operate on a seven day per week schedule while many smaller industries run five or six days per week. Thus, even on weekends some part of the emissions result from industrial activity. The weekday-weekend difference would be greater except that some level of industrial activity is also carried on over the weekends.

F. Modeling Ambient Fine Carbon Concentrations.

In this section ambient fine carbon aerosol was modeled by a variety of different methods to determine ambient fine carbon as a function of meteorological dispersion and heating degree days. The rationale for this section is based on the hypothesis that meteorological dispersion plays a large role in the concentration levels of ambient aerosols and that a significant fraction of urban fine carbon aerosols may result from space heating, particularly space heating using wood stoves and fireplaces. Source testing has demonstrated that residential wood burning emissions are 98% in the fine particle mode and that they are composed of about 60% carbon by weight (DeCesar and Cooper, 1981). Furthermore, it is likely that Medford has participated in the rising trend to use wood space heating which has been noted throughout the country during the past several years, especially since there is a good local wood supply in the region. Of course, it should be borne in mind that even if a correlational relationship is established between ambient levels of fine carbon aerosol and heating degree days, this only implies that fine carbon aerosol emissions change as a function of heating degree day values. Whether this effect results from increased space heating generally or from wood stoves in particular cannot be determined with the available data.

Two general sets of models were developed for the Medford Justice Building site. As mentioned previously, this is an urban site and appears to be the site where space heating emissions had the greatest impact. Models using 1/UH as the measure of meteorological dispersion are shown in Table 6. Models using ambient lead concentrations as the measure of dispersion are shown in Table 7. The factors (1/UH) and (Pb) are defined as follows:

(1/UH) = data point 1/UH value/average 1/UH value

(Pb) = data point ambient lead conc./average lead conc.

The use of these definitions causes the coefficients in the models to have similar values. Space heating emissions are modeled by heating degree day values ($18\frac{1}{3}°$ C - average daily temperature below $18\frac{1}{3}°$ C).

For 1/UH models the largest data set available contained 44 points. For lead models the largest data set available contained 41 points. The largest set of common points contained 31 points. Several data points were eliminated from the available data set in constructing the 1/UH data sets used for modeling. These points did not conform to the assumptions implicit in the box model described by Eqn. (5), i.e., the pollutants are uniformly mixed in the wind stream moving through the box. When the wind speed is several times higher than normal or the mixing height is much higher than normal, then the uniform mixing assumption becomes invalid. Five points out of 49 were rejected because they did not fit the box model In contrast, when ambient lead concentrations are used to model meteorological dispersion, this complication does not exist because lead concentrations are the result of whatever dispersion really occurs. The size of the lead data set was determined by the number of days on which lead fine aerosol samples were taken.

Table 6

Justice Building. Fine Carbon Models Using 1/UH to Measure Meteorological Dispersion.

		σ	r
	n = 44		
(1)	$C = 21.4$	21.9	0
(2)	$C = -0.4 \pm 3.7 + 21.8 \pm 3.0\ (1/UH)$	14.8	0.738
(3)	$C = 7.4 \pm 2.1 + 0.51 \pm 0.04\ (DD)(1/UH)$	11.0	0.864
(4)	$C = 5.6 + 3.4 \pm 3.9\ (1/UH) + 0.45 \pm 0.08\ (DD)(1/UH)$	11.0	0.866
	n = 31		
(5)	$C = 17.6$	17.8	0
(6)	$C = 1 \pm 4 + 21.8 \pm 3.4\ (1/UH)$	11.7	0.755
(7)	$C = 6.5 \pm 1.9 + 0.46 \pm 0.04\ (DD)(1/UH)$	8.8	0.869
(8)	$C = 6.5 + 0\ (1/UH) + 0.46 \pm 0.04\ (DD)(1/UH)$	8.8	0.869

In all cases there is not much difference between the model for the larger set and the model for the smaller set. This can be taken as evidence that both data sets represent typical points and do not include outliers.

Table 7

Justice Building. Fine Carbon Models Using Ambient Lead Concentrations to Measure Meteorological Dispersion.

		σ	r
	n = 41		
(9)	$C = 16.6$	17.6	0
(10)	$C = -4\pm2.7 + 20.9\pm2.1\ (Pb)$	9.9	0.827
(11)	$C = 5.4\pm1.3 + 0.46\pm0.03\ (DD)(Pb)$	6.9	0.920
(12)	$C = 3.3 + 3.5\pm3.1\ (Pb) + 0.39\pm0.06\ (DD)(Pb)$	6.8	0.923
	n = 31		
(13)	$C = 17.6$	17.8	0
(14)	$C = -5\pm2.7 + 22.6\pm2.1\ (Pb)$	8.5	0.879
(15)	$C = 6.2\pm1.4 + 0.48\pm0.03\ (DD)(Pb)$	6.3	0.935
(16)	$C = 2.6 + 5.8\pm3.1\ (Pb) + 0.34\pm0.07\ (DD)(Pb)$	6.0	0.942

The value of sigma shown in the tables is the standard deviation of the differences between the carbon values predicted by the model and the actual ambient carbon values. The correlation coefficient (r) relates carbon values predicted by the model to ambient carbon values. In each group the first model which estimates the ambient carbon concentration by using the average is the simplest model that can be posed. This model has no variable predictive capability, but it does establish the maximum values of sigma.

In the next simplest model in each group ambient carbon is assumed to be a function of meteorological dispersion only. This group of models is a significant improvement over the first set of models and in fact is not greatly different in sigma and r values from the more complex models that also incorporate heating degree days as a variable. The reason for this is that the major source of ambient carbon variance is meteorological dispersion. This group of models implies that 50 to 75% of ambient carbon concentrations variance results from variations in meteorological dispersion.

The third model in each group incorporates both meteorological dispersion and heating degree day values in a single term. These models imply that ambient carbon is composed only of background carbon and carbon that is emitted only by space heating sources. If aerosol carbon correlates with heating degree day values, it is assumed to result from space heating. This means that when the degree day value is zero, all local carbon emissions go to zero in the model. Since the emission inventory includes non-space heating carbon emission sources, this model is not physically correct, even though it accounts for more of the ambient carbon data variance than any of the previous models.

In the final model in each group ambient carbon concentrations are assumed to result from the sum of a background contribution, local non-space heating emissions, and local space heating emissions. The coefficients in these models were evaluated by using stepwise multiple regression (Draper and Smith, 1966). This program does not compute standard deviation values for the constant term in the equation, but it appears that the values are similar to those given for the constant term in the third model of each group. In the model in the second group of Table 6 the variable 1/UH was too weak to be included in the regression. These models explain the data as well (1/UH models) or slightly better (lead models) than the models which consider only space heating carbon sources. Comparing models 3, 7, 11, and 15 with models 4, 8, 12, and 16 shows that the coefficient associated with degree day dependent carbon term decreases when the non-degree day dependent carbon term becomes available.

These models show that up to 88% of the fine carbon data variance at the Justice Building can be explained in terms of variations of meteorological dispersion, as characterized by ambient lead concentration and heating degree day values. Figures 18 and 20 show a comparison of ambient carbon data and the values predicted by models 4 and 16, respectively. Figures 19 and 21 show the ratio of ambient heating degree day associated carbon to non-heating degree day carbon plotted as a time series for models 4 and 16, respectively. Figures 19 and 21 are quite similar, showing that both 1/UH and lead concentrations reasonably well model a similar entity believed to be meteorological dispersion. Figure 19 indicates during the winter ambient heating degree day associated carbon is about 5 times greater than non-heating degree associated carbon, while Figure 21 indicates approximately a factor of four

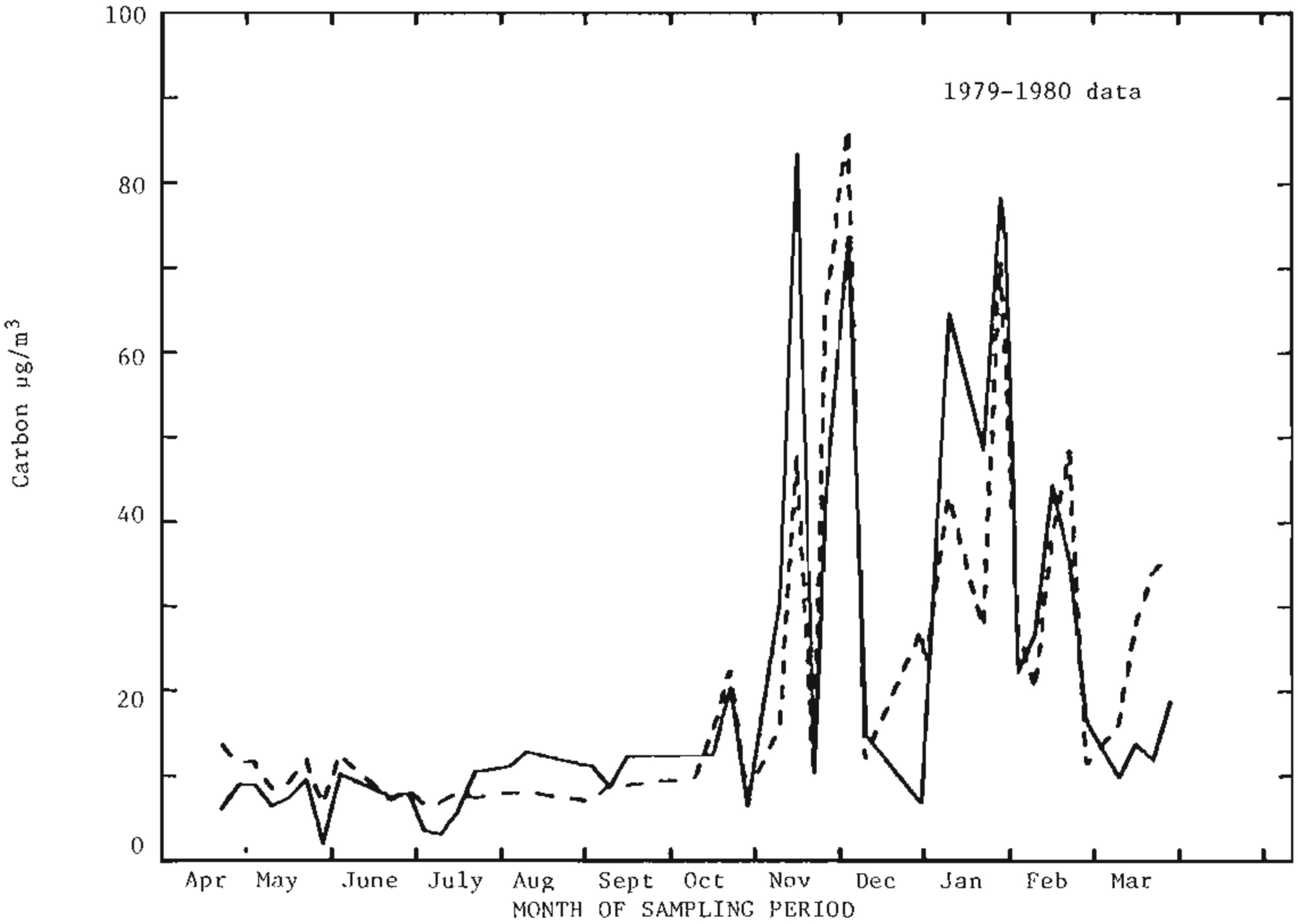


Figure 18. A comparison of Justice Building ambient fine carbon aerosol concentrations with predictions of Model 4. Line = ambient carbon aerosol; dash = predictions of Model 4.

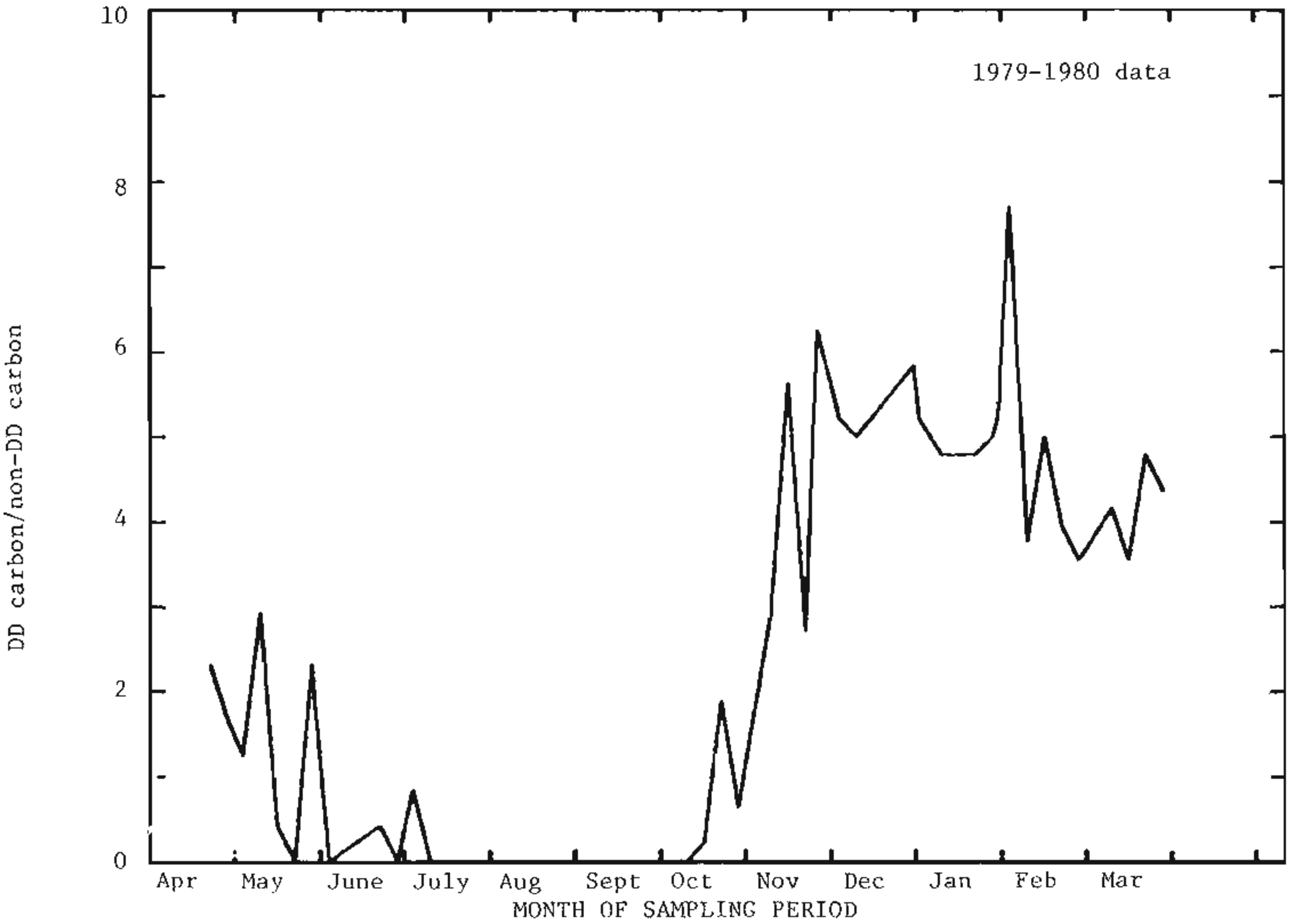


Figure 19. Time series of Justice Building degreeday carbon/non-degreeday carbon computed using Model 4.

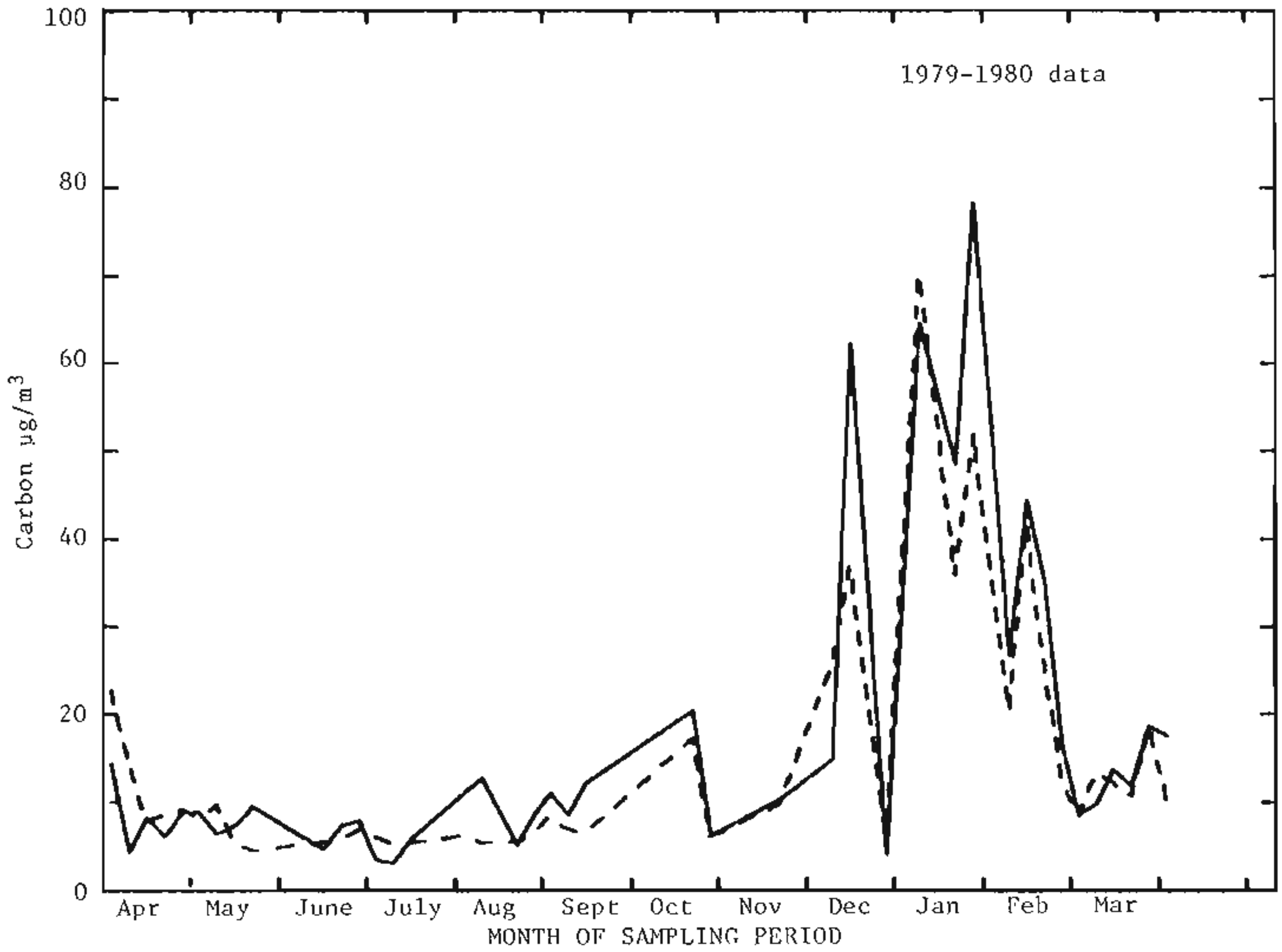


Figure 20. A comparison of Justice Building ambient fine carbon aerosol concentrations with predictions of Model 12. Line = ambient carbon; dash = predictions of Model 12.

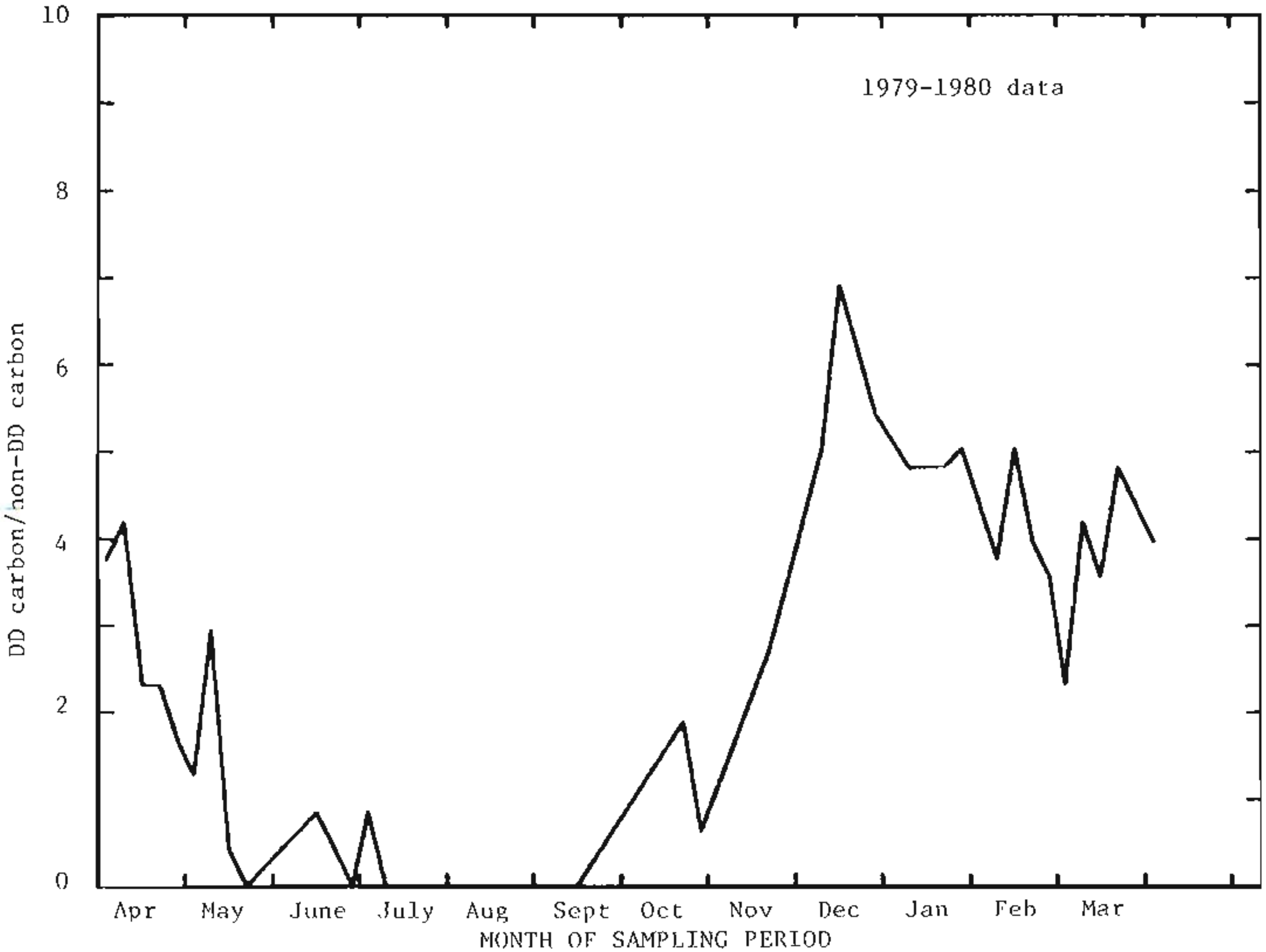


Figure 21. Time series of Justice Building degreeday carbon/non-degreeday carbon computed using Model 12.

increase. Table 2 shows that the average dispersion is decreased by about a factor of two during the winter as compared with the summer. Tables 3 and 4 show that the normalized winter fine carbon average is about twice the summer average. Therefore, on the average ambient wintertime carbon concentrations result from a doubling of emissions combined with a factor of two decrease in meteorological dispersion.

When White City and Dodge Road fine aerosol carbon data were modeled in the same manner as the Justice Building fine aerosol carbon data, using 1/UH as a measure of meteorological dispersion, the models shown in Table 8 were obtained. These models follow the same pattern as the Justice Building models shown in Tables 6 and 7, in that as the space heating variable is added to the dispersion variable, the model's predictive capability is increased. And when a multiple regression model, which incorporates both a non-space heating term and a space heating term, is used, the model achieves the best fit to the data.

The most notable feature of both the White City and the Dodge Road model 4 when compared to the Justice Building model 4 is that at the Justice Building the heating degree day associated term is much larger than the non-heating degree day associated term, while at both White City and Dodge Road the magnitude of these terms is about equal. This can be seen by comparing Figures 19, 23, and 25. Obviously space heating is a much larger component of the winter emission inventory in the vicinity of the Justice Building than it is in the vicinity of White City or Dodge Road. Because non-space heating carbon is such a large part of fine aerosols at these sites, and the variability of emission rates of this carbon is not characterized by these models, they are not as capable of modeling ambient fine carbon aerosols as the Justice Building models, hence the smaller correlation coefficients shown in Table 8 result. Nevertheless, these models do a reasonably good job of modeling ambient fine carbon aerosols. This is shown by Figures 22 and 24.

Table 8

Industrial and Background Site Fine Carbon Models
Using 1/UH to Measure Meteorological Dispertion

	White City n = 45	σ	r
(17)	$C = 15.6$	21.9	0
(18)	$C = 5.4\pm2.1 + 10.2\pm1.6\ (1/UH)$	9.1	0.699
(19)	$C = 10.4\pm1.5 + 0.36\pm0.05\ (DD)(1/UH)$	8.7	0.727
(20)	$C = 7.7 + 4.6\pm2.6\ (1/UH) + 0.13\pm0.05\ (DD)(1/UH)$	8.4	0.749
	Dodge Road n = 40		
(21)	$C = 5.1$	3.4	0
(22)	$C = 2.3\pm0.6 + 2.7\pm0.4\ (1/UH)$	2.4	0.701
(23)	$C = 3.5\pm0.4 + 0.09\pm0.01\ (DD)(1/UH)$	2.3	0.725
(24)	$C = 2.9 + 1.2\pm0.8\ (1/UH) + 0.03\pm0.01\ (DD)(1/UH)$	2.2	0.745

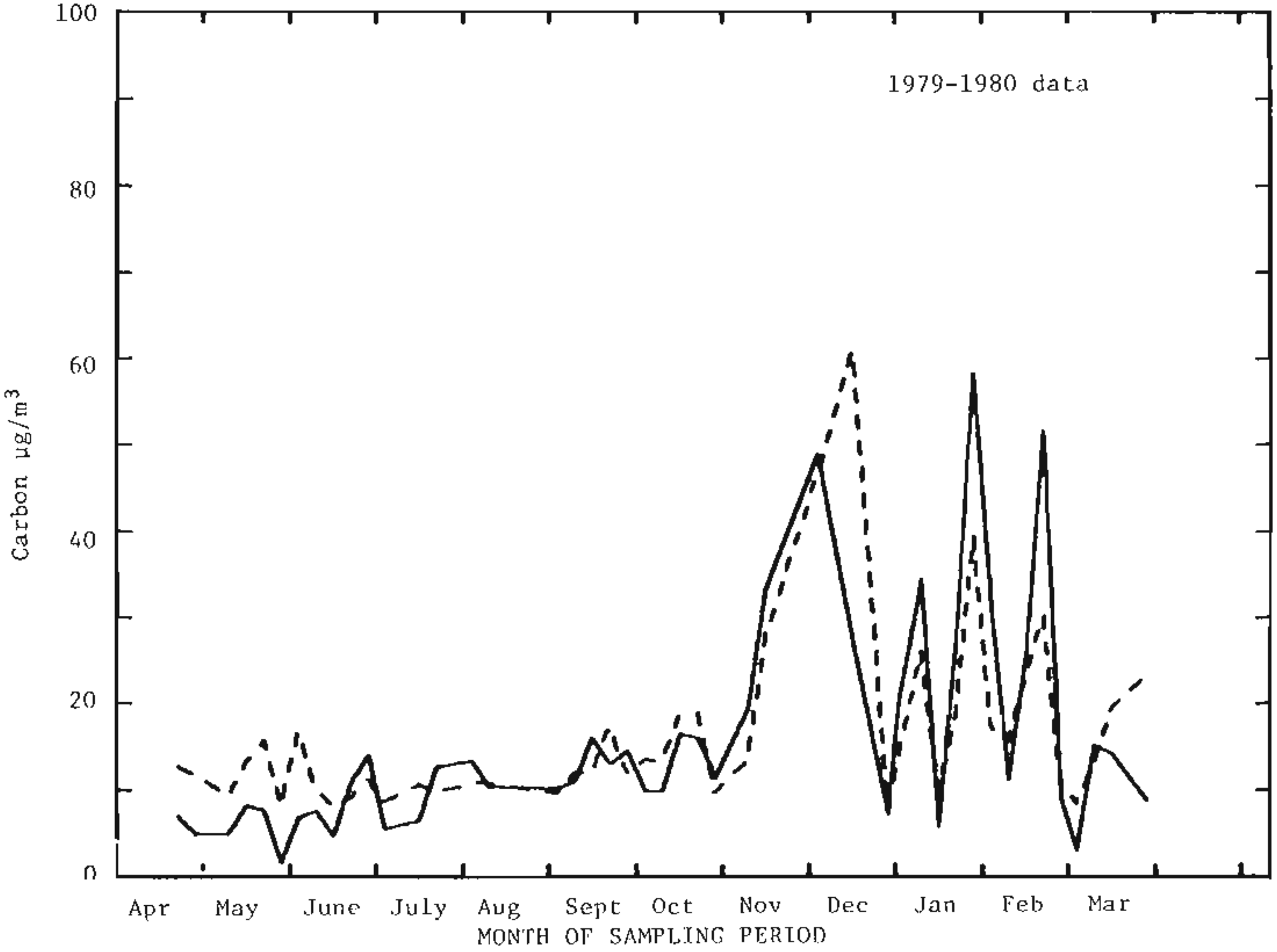


Figure 22. A comparison of White City ambient fine carbon aerosol concentrations with predictions of Model 20. Line = ambient carbon; dash = predictions of Model 20.

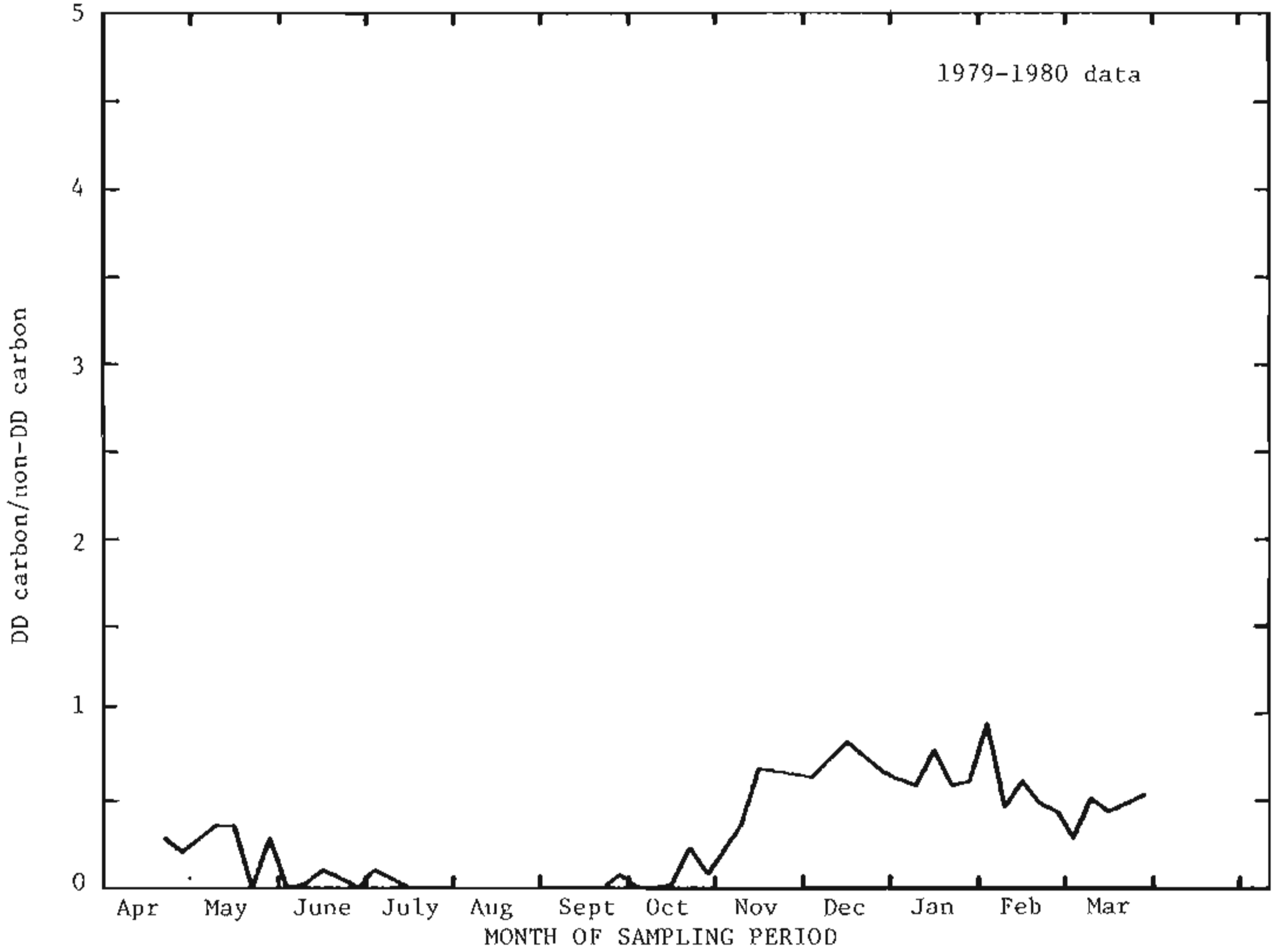


Figure 23. Time series of White City degreeday carbon/non-degreeday carbon computed using Model 20.

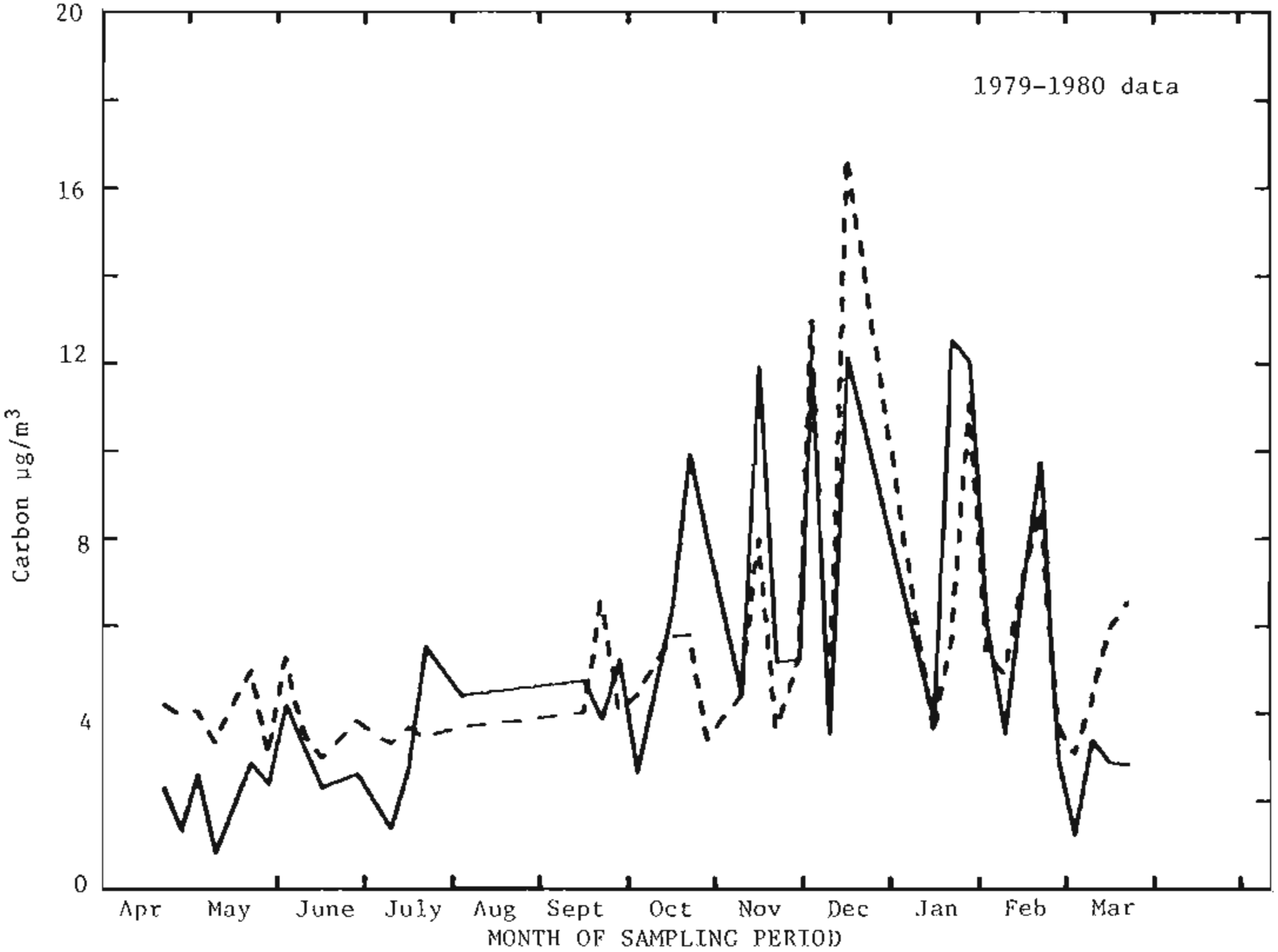


Figure 24. A comparison of Dodge Road ambient fine carbon aerosol concentrations with predictions of Model 24.

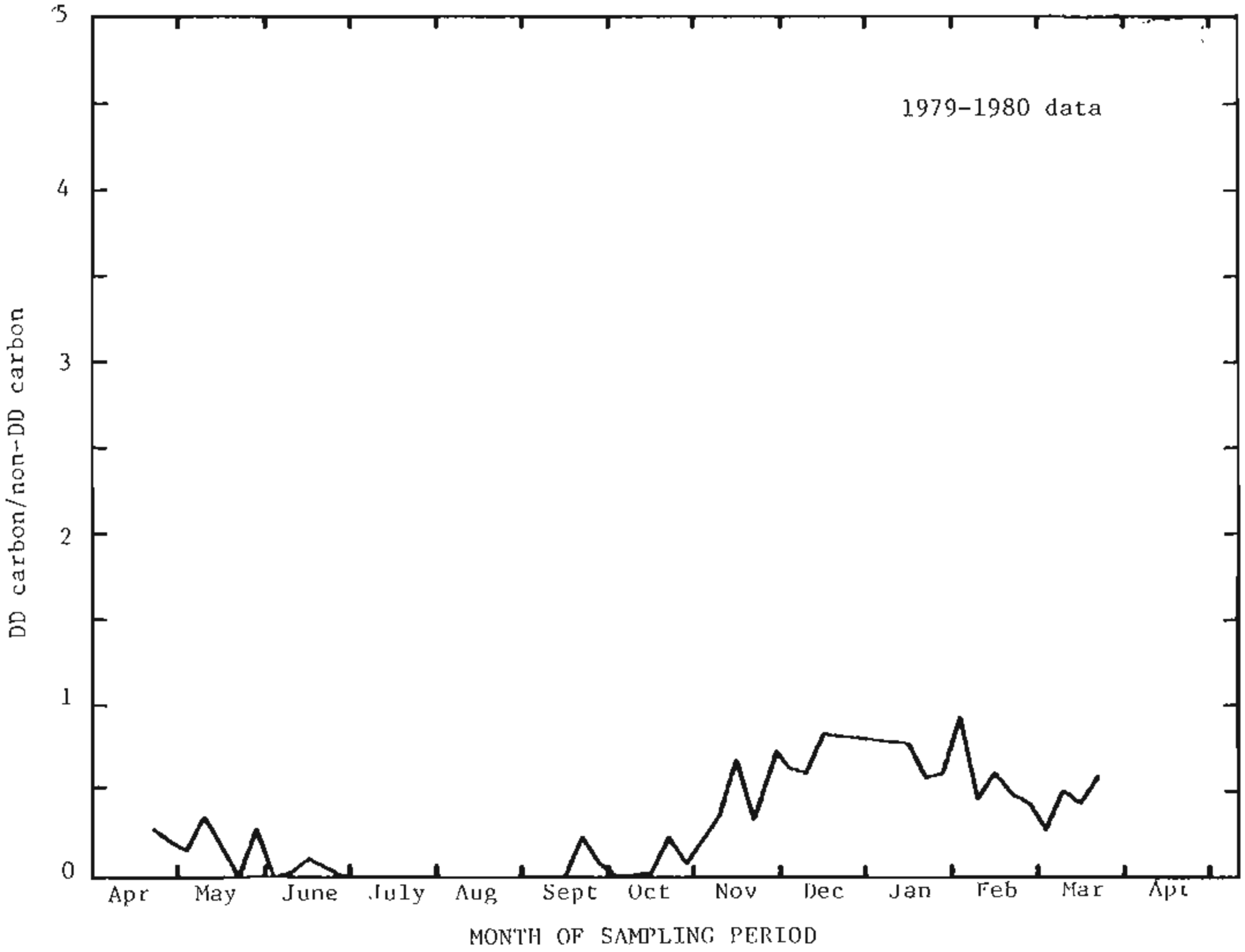


Figure 25. Time series of Dodge Road degreeday carbon/non-degreeday carbon computed using Model 24.

G. Meteorological Dispersion Analysis Applied to a Special Data Set.

During the 1979 Christmas week a special experiment was conducted at the Justice Building site to examine the effects of residential wood stove emissions in detail. The sampling time period was chosen because industrial emissions were expected to be at a minimum. High volume samplers were used to collect total aerosol samples for 12-hour periods from 4 AM to 4 PM (day) and from 4 PM to 4 AM (night). While fine aerosol sampling would have been more appropriate to study wood stove effects, the data set is useful to examine the utility of using 1/UH meteorological dispersion analysis on a data set that is small enough to examine on a point by point basis.

Sampling began at 4 AM on December 24 and ended at 4 PM on December 28. Daily total degree day values were almost constant during the whole period. Figure 26 shows that nighttime ambient carbon aerosol concentrations are higher than daytime values, except for point 27N. However, without considering the effects of meteorological dispersion these results cannot be used to make reliable statements about emissions. If these data are normalized to a common value of meteorological dispersion, the normalized data are proportional to emissions emitted during the sampling period. It is emission values that are really of interest because these must be controlled to change air pollution levels.

For this data set meteorological dispersion was characterized by 1/UH. The 1/UH values were determined by summing the 7 AM, 10 AM,

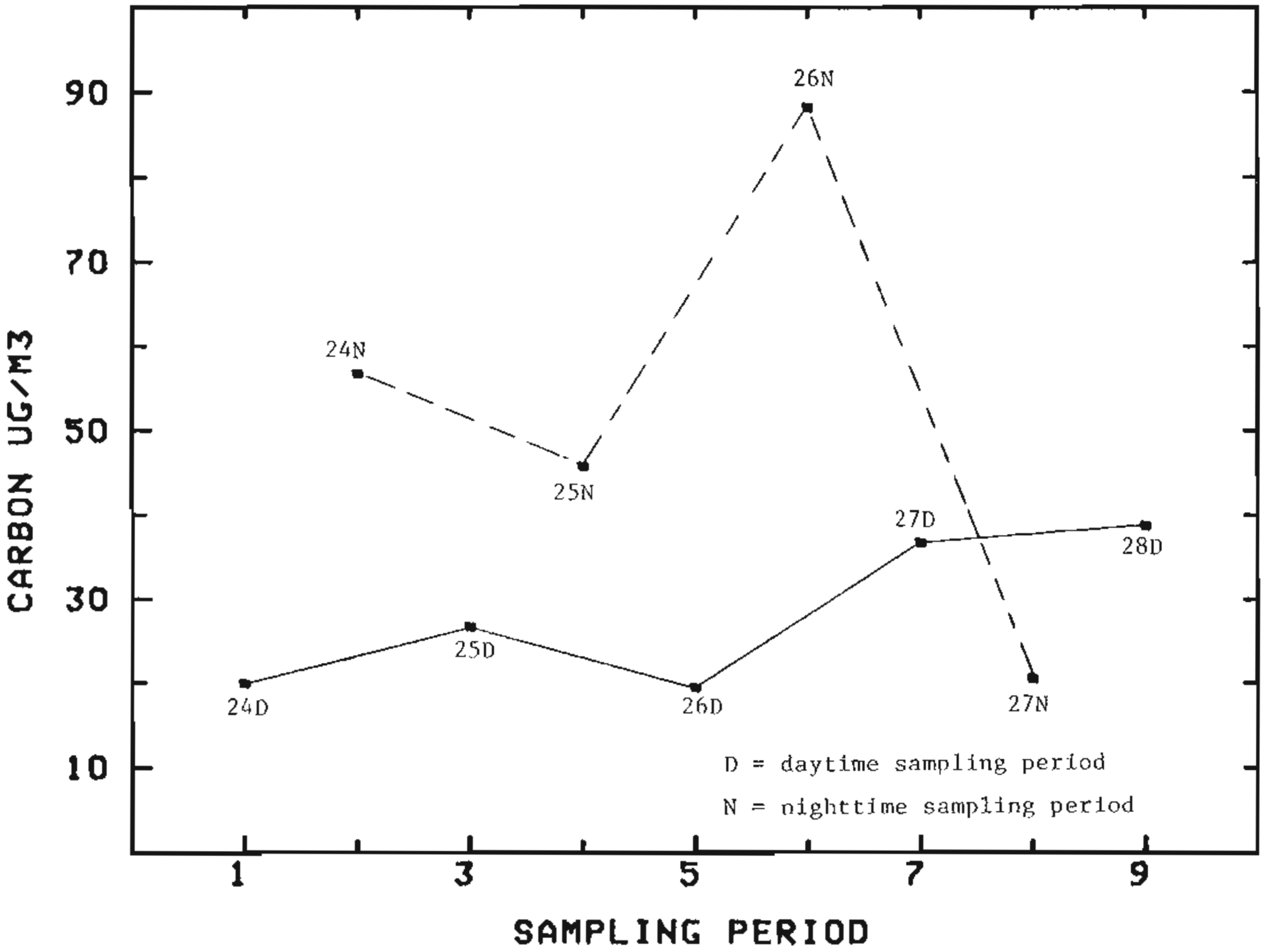


Figure 26. Time series of Justice Building ambient fine carbon aerosol for special wood burning impact experiment.

1 PM, and 4 PM 1/UH values for the day data points, and 7 PM, 10 PM, 1 AM, and 4 AM for the night data points. Figure 27 shows that daytime ambient carbon data are highly correlated with 1/UH. Figure 28 shows that points 24, 25, and 26 have aspects of a linear relation with 1/UH, but point 27 has a 1/UH value which is too high to be consistent with the other points. This inconsistency illustrates the error introduced when emissions are not constant over the sampling period. During the first 6 hours of the sampling period on the night of the 27th dispersion was high because of relatively high wind speeds, while during the second 6 hours the wind speed dropped and dispersion was weak. If most of the emissions emitted during the night sampling period come from residential wood burning, and if these emissions were emitted mostly during the early part of the evening when dispersion conditions were high, then the 1/UH value representative of the sampling period will be higher than the 1/UH value operating during the part of the sampling period when emissions were occurring. It can be assumed that in Figure 26 point 27N has a low ambient carbon value not because emissions decreased but because dispersion conditions were high.

Figures 26 and 29 can be compared to see how differences in meteorological dispersion distort statements that might be made about emissions using ambient concentration data. While day values are similar on both figures, night values are quite different. Figure 29 shows night emissions are highest on the night of Christmas day, which is to be expected, and somewhat less on the night of the 26th and probably on the 27th (adjusting the 1/UH value as explained above will move

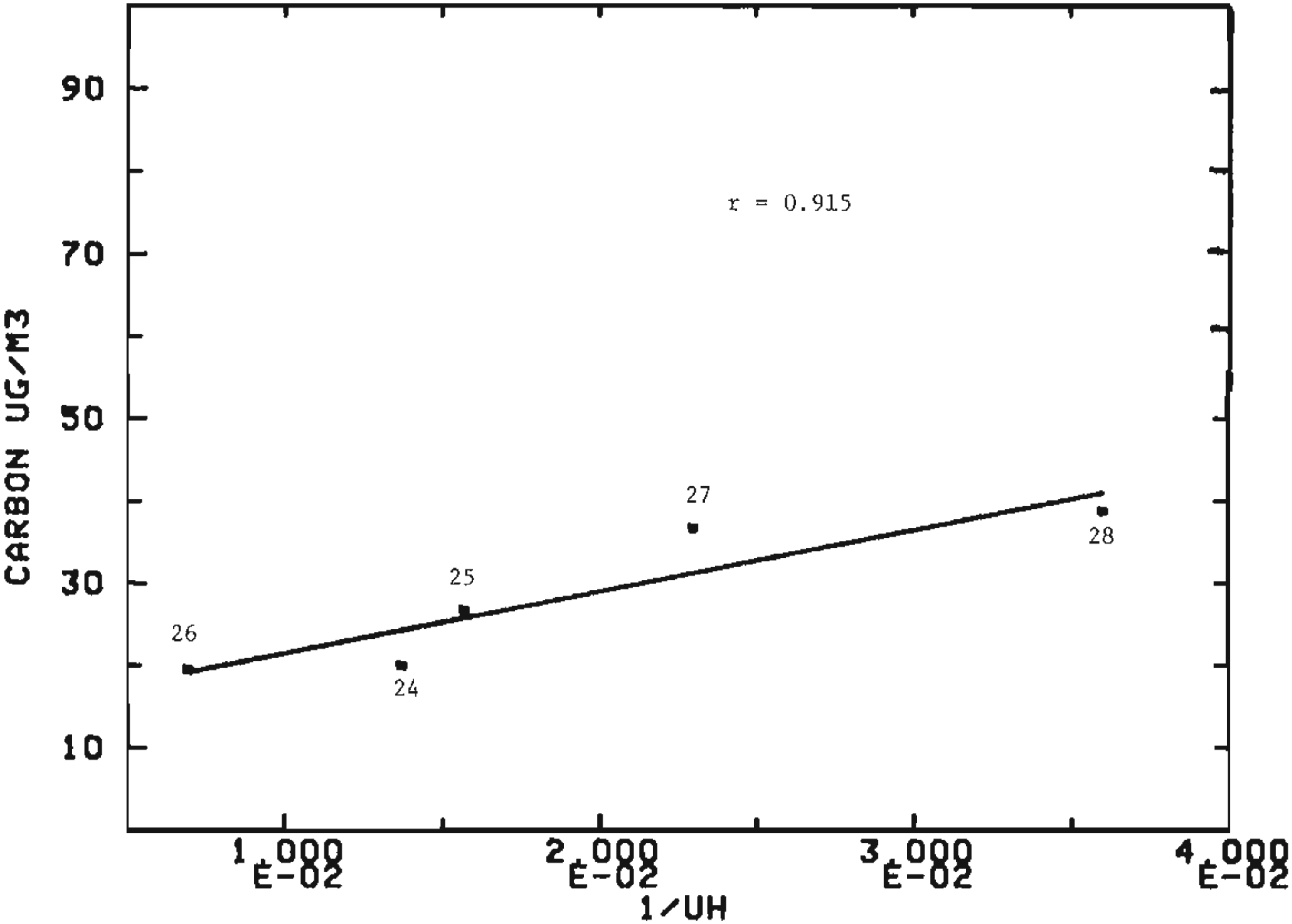


Figure 27. Daytime Justice Building ambient fine carbon aerosol vs. meteorological dispersion (1/UH) for a special wood burning impact experiment.

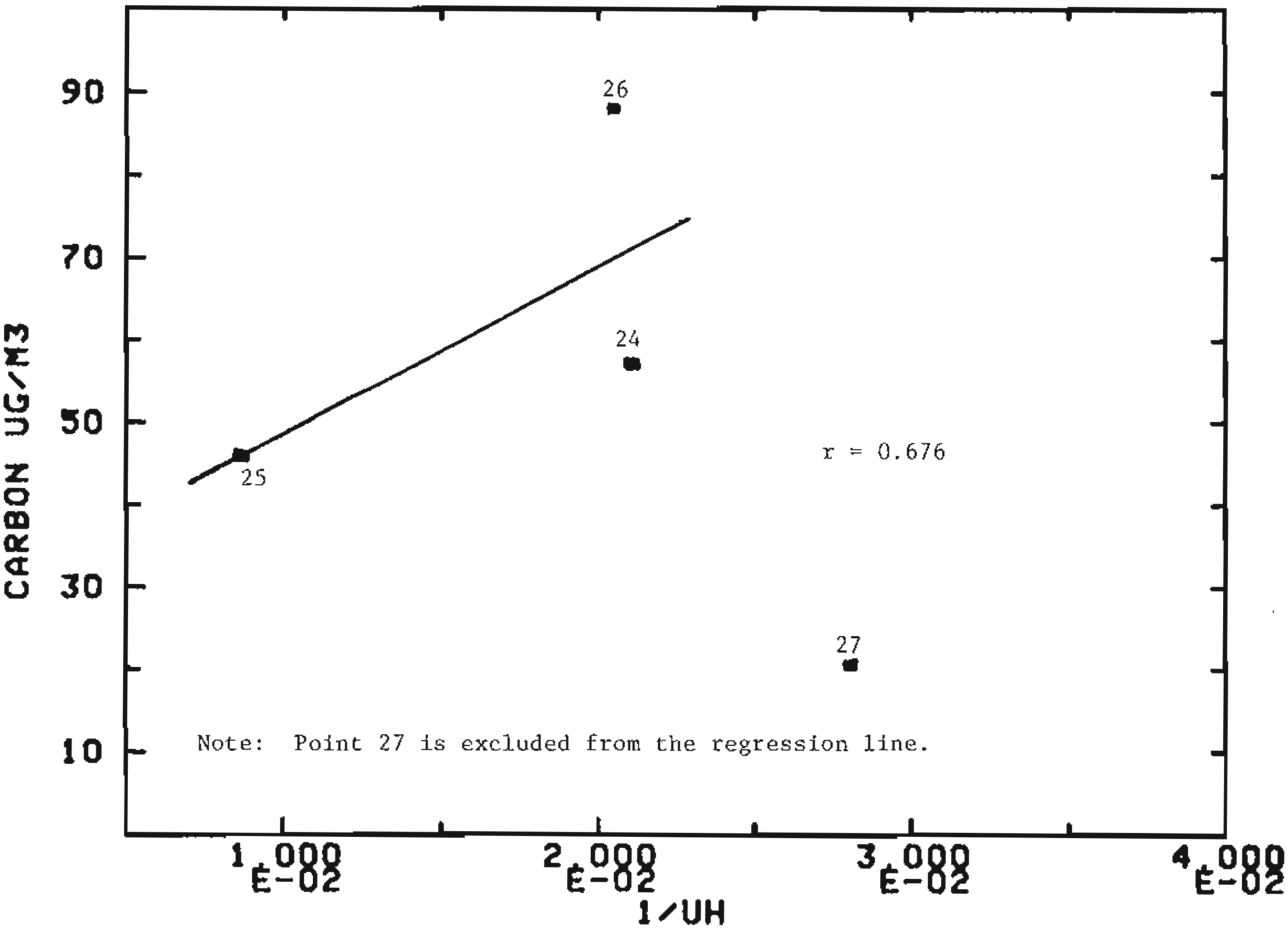


Figure 28. Nighttime Justice Building ambient fine carbon aerosol vs. meteorological dispersion (1/UH) for a special wood burning impact experiment.

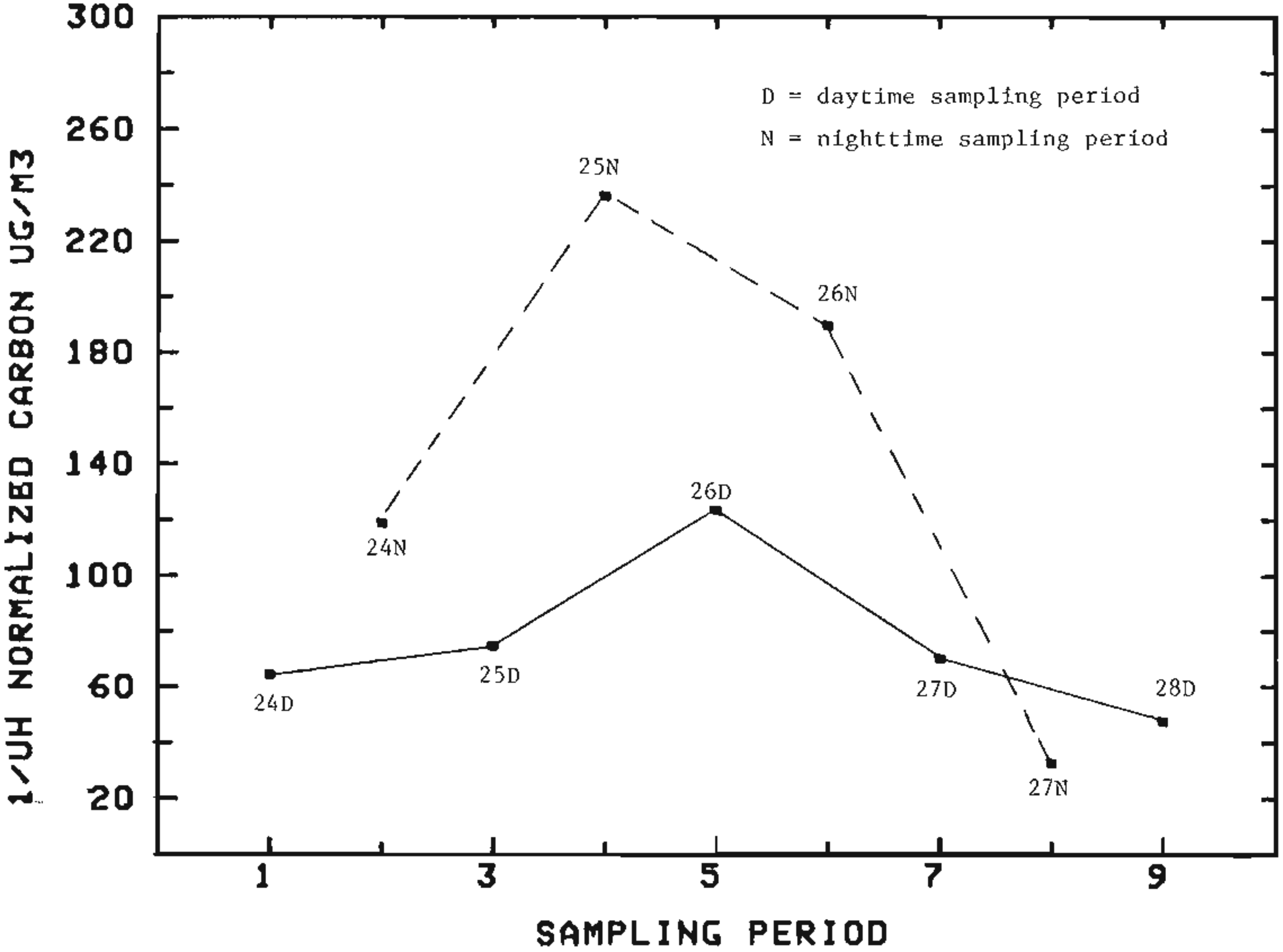


Figure 29. Time series of Justice Building normalized ambient fine carbon aerosol for a special wood burning impact experiment.

point 27N to the expected location). One could conclude from either Figure 26 or Figure 29 that nighttime emissions are about twice as great as daytime emissions, but if the conclusion had been drawn from Figure 26, then it could have been wrong if nighttime dispersion conditions had been significantly poorer than daytime dispersion.

IV. SUMMARY

This study has shown that carbon is a major component of both fine and coarse aerosols in the Medford airshed. Total aerosol, the sum of fine and coarse aerosol, was found to be composed of from 15 to 50% carbon, while fine aerosol was found to be composed of from 25 to 60% carbon. At the Justice Building site ambient total carbon concentrations range from low values of 10 $\mu g/m^3$ in the summer to high values of 100 $\mu g/m^3$ in the winter. Winter total ambient carbon aerosol concentrations averages were 2.6 times greater than summer values and winter fine ambient carbon aerosol concentration averages were 4.4 times greater than summer averages. At the White City industrial site the lowest total aerosol carbon values were also about 10 $\mu m/g^3$ and occurred during the summer. The highest values occurred during the winter and were generally below 80 $\mu g/m^3$. Data from the Dodge Road background site showed the same seasonal pattern and ranged from 3 $\mu g/m^3$ to about 15 $\mu g/m^3$.

This study separated the increases in ambient carbon concentrations due to poor meteorological dispersion conditions from those due to increased emissions, especially those due to increased emissions associated with space heating. Meteorological dispersion was characterized by 1/UH and by ambient lead concentrations. Space heating demand was characterized by heating degree values. Using these variables it was found that up to 88% of the variances in ambient fine carbon concentrations at the Justice Building could be described by a model in which the only fine carbon sources considered were background carbon, carbon not associated with space heating, and carbon that is associated with space heating. This model showed that during the wintertime at the Justice Building ambient fine carbon concentrations that correlated with heating degree day values were four times greater than ambient fine carbon concentrations that did not correlate with heating degree day values. These results support the hypothesis that a large fraction of fine carbon aerosols come from space heating with wood because survey data have shown that wood burning is a significant means of space heating in Medford, and wood burning is a known strong source of fine carbon aerosols.

Since wintertime dispersion conditions are such that average wintertime dispersion is only half as strong as summertime dispersion, a doubling of emissions during the winter actually results in a fourfold increase of ambient pollution concentrations. At the White City and Dodge Road sites modeling showed that emissions not associated with space heating were roughly equal to emissions associated with space heating.

When fine carbon data, sampled at the Justice Building, were normalized to constant meteorological dispersion conditions, it was shown that seasonal average values increased by 13 $\mu g/m^3$ from summer to winter. CEB analysis (DeCesar and Cooper, 1981) showed that average winter fine carbon sampled at the Justice Building site results from space heating with wood.

It was shown that one possible strategy to bring Medford into compliance with the aerosol standard of the 1970 Clean Air Act would require a 26% reduction in winter total aerosol emissions. If all of the winter increase in fine carbon emissions is assumed to be due to space heating, the total elimination of this source would provide 67% of the reduction in emissions needed for compliance.

By dividing carbon data into organic and elemental components it was shown that average carbon aerosol was composed of from 20 to 35% of organic carbon and from 5 to 15% of elemental carbon. Coarse aerosol was composed of 8 to 28% percent organic carbon and from 5 to 12% of elemental carbon. When elemental and organic carbon were correlated within a size fraction, correlation coefficients from $r = 0.70$ to 0.90 were obtained. Since all elemental carbon comes from combustion sources, these high correlation coefficients indicate that most of the organic carbon must come from combustion sources as well.

In addition to examining normalized carbon data, this study also determined weekday and weekend seasonal average values of fine aerosol Pb, K, Na, Zn, S, Fe, and Br. These data showed that lead emissions were reasonably constant and that based on higher weekday emission levels, K, Na, and S are mainly associated with industrial emissions.

Future aerosol studies in the Medford area should take advantage of the area's relatively consistent wind patterns. Further evaluation of meteorological dispersion using 1/UH should be done to gain an understanding of the long-term dispersion characteristics of the area. An artificial tracer study would be useful to confirm the ability of both 1/UH and ambient lead concentrations to measure meteorological dispersion. Also a search should be made for other aerosol component species that could be used as measures of dispersion when lead is phased out as a motor fuel additive.

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APPENDIX I

CARBON DATA FILES

Day: Day of Sampling Period (Day 1 = April 1, 1979)

OC: Organic Carbon ($\mu g/m^3$)

EC: Elemental Carbon ($\mu g/m^3$)

TC: Total Carbon ($\mu g/m^3$)

Mass: Aerosol Mass ($\mu g/m^3$)

(1/UH): Sum of 1/UH Values Computed Every Four Hours During the 24-hour Sampling Period

DD: Heating Degree Day Value

JUSTICE BUILDING SITE TOTAL AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
3	13.92	8.10	22.02	85	---	18
9	6.19	3.62	9.82	35	---	20
15	8.19	5.00	13.18	380	---	11
21	831	4.73	13.03	69	.0301	11
57	3.40	1.28	4.68	37	.0033	11
75	9.14	5.28	14.42	72	.0022	4
81	8.82	4.50	13.31	79	.0119	2
87	14.99	7.57	22.56	116	.0338	0
93	6.59	3.29	9.88	59	.0075	4
99	5.06	1.94	7.00	47	.0145	0
105	8.57	4.14	12.71	67	.0285	0
111	18.53	9.34	27.87	125	.0206	0
117	11.56	4.43	16.0	107	---	0
123	11.36	5.30	16.66	83	.0292	0
129	13.43	7.40	20.83	140	.0286	0
135	9.85	5.67	15.52	72	---	0
141	4.98	2.17	7.15	32	---	0
147	12.05	7.43	19.49	68	---	0
153	11.84	7.59	19.43	66	.0172	0
159	11.31	6.42	17.73	83	.0415	0
165	21.34	11.69	33.03	144	.0406	0
183	10.04	5.44	15.48	58	.055	0
189	13.77	8.29	22.06	110	.0541	0
195	21.53	12.07	33.60	164	.0996	1
201	19.60	10.14	29.74	70	.0733	9
207	6.50	2.32	8.83	40	.0160	3
219	30.18	11.99	42.17	97	.0321	14
225	55.90	26.67	82.57	243	.0797	27
228	86.19	49.19	135	316	.0977	27
231	9.01	2.89	11.89	37	.0163	13
235	49.67	21.65	71.31	170	.1027	30
239	35.82	12.71	48.54	94	.0348	29
243	72.19	29.31	101	207	.1640	25
249	22.97	10.62	33.59	130	.0133	24
255	54.19	27.20	81.38	166	.1907	24
261	69.96	25.86	95.82	265	---	25
267	3.99	1.09	5.09	18	.0016	26
269	24.23	10.45	34.69	73	.0397	28
271	22.49	11.94	34.42	84	.0244	25
279	71.14	34.57	105.71	271	.0814	23
285	16.90	8.92	25.81	79	.0089	31
297	79.14	37.32	116.46	281	.1362	24
303	10.40	4.15	14.55	157	.0315	37
309	20.21	13.69	33.90	78	.0385	18

Justice Building Site Total Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
315	13.20	16.52	29.52	157	.0673	24
321	30.80	20.12	50.92	175	.1087	19
327	15.69	5.92	21.61	60	.0160	17
333	8.95	2.69	11.64	37	.0037	17
339	9.27	3.73	13.00	59	.0253	20
345	13.99	5.64	19.63	74	.0600	17
351	10.19	3.97	14.16	53	.0615	23
357	16.08	6.48	22.57	78	.0721	21
363	17.28	6.42	23.69	104	---	19

JUSTICE BUILDING SITE FINE AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
3	9.57	4.65	14.22	48	---	18
9	2.63	1.55	4.18	14	---	20
15	5.62	2.62	8.24	22	---	11
21	4.7	1.70	5.87	19	.0301	11
27	5.64	3.10	8.74	26	.0278	8
33	5.4	3.37	8.77	31	.0330	6
39	4.07	2.08	6.15	18	.009	14
45	5.03	2.07	7.10	22	.0311	2
51	6.22	3.17	9.39	31	.0760	0
57	1.40	.36	1.76	7	.0033	11
63	7.38	2.56	9.94	23	.0861	0
75	3.60	.96	4.56	16	.0022	4
81	5.71	1.53	7.24	20	.0119	2
87	6.38	1.35	7.73	23	.0338	0
93	2.67	.63	3.31	11	.0075	4
99	2.45	.38	2.83	10	.0145	0
105	4.48	1.11	5.58	17	.0285	0
111	7.79	2.35	10.29	29	.0206	0
117	7.46	2.15	9.60	27	---	0
123	7.61	3.37	10.98	20	.0292	0
129	8.23	4.34	12.55	36	.0286	0
141	3.24	1.72	4.96	6	---	0
147	6.12	2.33	8.46	16	---	0
153	6.73	4.17	10.89	19	.0172	0
159	6.43	2.10	8.52	22	.04415	0
165	8.26	3.80	12.06	29	.0406	0
189	9.06	3.14	12.2	33	.0541	0
195	8.94	3.31	12.25	41	.0996	1
201	15.04	5.21	20.25	38	.0733	9
207	4.76	1.29	6.05	14	.0160	3
219	22.67	7.83	330.50	56	.03321	14
225	6349	19.90	83.39	174	.0797	27
228	118	45.09	163.	258	.0977	27
231	7.89	2.31	10.20	23	.0163	13
235	30.58	13.01	43.59	113	.1027	30
243	53.47	20.08	73.55	146	.1640	25
246	6.95	1.84	8.79	44	---	18
249	11.34	3.40	14.73	55	.0133	24
255	46.56	15.49	62.05	107	.0133	33
261	46.63	19.28	65.91	120	---	25
267	3.30	.61	3.91	12	.0016	26
269	4.92	1.64	6.56	42	.03397	28

Justice Building Site Fine Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
271	14.40	3.66	18.06	43	.0354	25
279	45.35	19.23	64.58	132	.0814	23
285	14.31	6.28	20.68	44	.0089	31
291	35.84	12.54	48.38	80	.0478	23
297	53.67	24.47	78.14	155	.1362	24
299	52.21	20.76	72.98	147	.1081	25
303	16.26	5.71	21.97	86	.0315	37
309	20.12	6.42	26.55	49	.0385	18
315	32.78	11.38	44.17	83	.0673	24
321	24.09	11.10	35.19	84	.1087	19
327	12.43	3.78	16.21	40	.0160	17
333	6.40	2.09	8.49	21	.0037	11
339	6.86	2.81	9.67	28	.0253	20
345	9.79	3.78	13.57	32	.0600	17
351	8.66	3.08	11.74	29	.0615	23
357	13.31	5.19	18.51	39	.0721	21
363	12.53	4.85	17.38	42	---	19

WHITE CITY SITE TOTAL AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
3	14.95	10.68	25.62	115	---	18
9	3.54	2.22	5.76	27	---	20
15	4.70	4.21	8.91	30	---	11
21	10.48	5.68	16.16	82	.0301	11
57	5.41	1.23	6.63	44	.0033	11
75	8.83	4.91	13.75	70	.0022	4
81	11.99	5.81	17.80	97	.0119	2
87	22.36	9.14	31.51	143	.0338	0
93	10.27	5.43	15.70	67	.0075	4
99	4.55	2.01	6.55	39	.0145	0
105	11.34	5.24	16.58	67	.0385	0
111	13.28	6.85	20.13	130	.0206	0
123	17.67	10.63	28.31	111	.0292	0
129	8.37	5.20	13.57	99	.0286	0
135	7.83	5.00	12.83	61	---	0
147	8.44	5.10	13.54	84	---	0
183	8.55	6.00	14.55	54	.0551	0
189	14.34	7.80	22.14	115	.0541	0
195	22.49	17.33	39.82	206	.0996	1
201	15.86	8.77	24.64	74	.0733	9
207	15.20	9.00	24.20	62	.0160	3
213	20.54	12.46	33.00	99	---	17
219	20.45	8.87	29.32	79	.0321	14
231	8.26	3.70	11.96	29	.0163	13
233	16.64	6.39	23.02	31	---	30
235	8.44	4.98	13.42	62	.1027	30
237	10.43	3.40	13.83	27	---	27
239	13.62	4.91	18.53	28	.0348	29
243	49.18	22.16	71.34	234	.1640	25
249	16.80	14.26	31.07	87	.0133	24
255	30.91	20.94	51.86	162	.1907	33
261	34.09	32.09	66.18	218	---	25
267	7.61	4.43	12.04	37	.0016	26
269	9.35	6.21	15.56	39	.0397	28
271	24.34	12.28	36.62	96	.0244	25
273	19.22	12.08	31.30	86	---	18
279	21.86	7.83	29.69	224	.0814	23
285	1.86	.59	2.44	60	.0089	31
291	27.79	13.69	41.48	97	.0478	23
297	91.03	34.16	125	362	.1362	24
303	11.71	5.79	17.50	166	.0315	37
309	13.40	9.36	22.96	39	.0385	18

White City Site Total Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
315	25.78	19.25	45.03	119	.0673	24
321	27.91	24.84	52.75	160	.1087	19
327	17.20	6.83	24.03	72	.0160	17
333	4.72	3.73	8.44	35	.0037	11
339	14.50	7.24	21.73	79	.0253	20
345	12.66	8.26	20.92	87	.0600	17
357	9.42	5.26	14.68	62	.0721	21
363	14.81	5.50	20.31	140	---	19

WHITE CITY SITE FINE AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
9	2.91	.88	3.80	9	---	20
15	2.88	.88	3.77	8	---	20
21	4.97	1.80	6.77	24	.0301	11
27	3.48	1.40	4.88	15	.0278	8
39	3.36	1.47	4.83	18	.0090	14
45	6.24	1.79	8.03	31	.0311	14
51	5.45	2.14	7.59	24	.0760	0
57	1.38	.11	1.48	6	.0033	11
63	5.05	1.42	6.67	18	.0861	0
69	5.26	2.36	7.61	22	.0228	1
75	3.84	.73	4.57	14	.0022	4
81	9.19	1.64	10.82	26	.0119	2
87	11.45	2.46	13.91	33	.0338	0
93	4.83	.65	5.48	16	.0075	4
105	5.28	1.05	6.33	18	.0285	0
111	8.91	3.62	12.52	49	.0206	0
123	9.06	4.33	13.38	37	.0292	0
129	6.69	3.65	10.34	40	.0286	0
135	7.72	4.58	12.30	52	---	0
142	4.70	2.39	7.10	19	---	0
147	7.67	3.08	10.76	34	---	0
153	7.12	3.04	10.17	24	.0172	0
159	7.15	3.79	10.95	27	.0415	0
165	10.41	5.50	15.91	46	.0406	0
171	8.10	4.70	12.81	32	.0920	0
177	9.43	4.93	14.36	34	.0358	3
183	7.10	2.75	9.85	17	.0551	0
189	7.47	2.43	9.89	27	.0541	0
195	10.55	5.87	16.42	49	.0996	1
201	11.44	4.61	16.06	29	.0733	9
207	8.51	2.81	11.32	23	.0160	3
213	15.36	6.06	21.42	41	---	17
219	14.11	5.35	19.46	34	.0321	14
225	23.11	9.99	33.10	57	.0797	27
237	10.29	2.82	13.11	20	---	27
243	34.93	13.88	48.82	90	.1640	25
255	19.09	7.88	26.97	56	.1907	33
261	22.44	11.44	33.88	69	---	25
267	5.45	1.65	7.10	15	.0016	26
271	14.47	5.49	19.96	39	.0244	25
273	14.96	7.05	22.02	44	---	18

White City Site Fine Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
279	27.91	6.35	34.26	77	.0814	23
285	4.06	1.63	5.69	13	.0089	31
291	22.48	4.88	27.36	51	.0478	23
297	42.82	15.38	58.19	127	.1362	24
303	25.28	5.73	31.01	73	.0315	37
309	8.98	2.05	11.03	24	.0385	18
315	19.61	5.87	25.48	50	.0673	24
321	41.47	10.10	51.57	63	.1087	19
327	6.85	2.16	9.00	24	.0160	17
333	2.21	7	2.91	11	.0037	11
339	10.65	4.33	14.98	28	.0253	20
345	10.27	3.86	14.13	31	.0600	17
357	5.03	3.71	8.74	23	.0721	21
363	11.73	3.42	15.15	44	---	19

DODGE ROAD SITE TOTAL AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
3	2.84	1.15	4.00	19	---	18
57	2.58	.29	2.87	17	.0033	11
63	4.8	1.21	6.01	37	.0861	0
69	3.51	1.04	4.55	32	.0228	1
75	2.84	1.01	3.84	24	.0022	4
87	5.71	1.18	6.89	37	.0338	0
99	2.92	.46	3.38	24	.0145	0
105	3.67	.99	4.66	23	.0285	0
111	7.01	1.60	8.60	48	.0206	0
117	4.88	.92	5.81	32	---	0
123	3.30	.70	4.00	23	.0292	0
135	2.46	.33	2.8	24	---	0
141	1.85	.31	2.16	15	---	0
150	5.14	1.37	6.51	16	---	0
159	6.35	1.54	7.89	17	.0415	0
165	6.03	1.56	7.59	40	.0406	0
171	4.61	1.15	5.76	36	.0920	9
177	5.45	1.90	7.35	44	.0358	3
183	4.46	.98	5.45	26	.0551	0
189	7.95	2.91	10.86	59	.0541	0
195	6.85	2.56	9.40	62	.0996	1
201	10.32	3.16	13.48	27	.0733	9
207	10.14	3.01	13.16	26	.0160	3
213	6.86	2.37	9.23	19	---	17
219	4.99	1.04	6.02	15	.0321	14
225	10.42	4.71	15.13	31	.0797	27
231	3.76	1.51	5.27	12	.0163	13
233	3.74	1.16	4.90	11	---	30
235	9.98	4.31	14.29	45	.1027	30
237	3.81	1.00	4.81	9	---	27
239	4.33	.93	5.26	14	.0348	29
243	6.67	3.52	10.20	32	.1640	25
255	9.74	5.99	15.73	33	.1907	33
261	15.15	6.80	21.95	43	---	25
267	3.86	1.28	5.15	12	.0016	26
269	4.64	1.64	6.28	15	.0397	28
271	7.65	2.38	10.03	19	.0244	25
273	6.05	2.44	8.49	19	---	18
279	14.46	3.04	17.50	40	.0814	23
285	1.93	.58	2.51	10	.0089	31
291	9.14	2.24	11.38	25	.0478	23
297	9.70	3.63	13.32	33	.0478	24

Dodge Road Site Total Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
303	4.34	1.28	5.61	22	.0315	37
309	3.82	1.81	5.63	14	.0385	
315	8.67	2.66	11.33	26	.0673	24
321	9.35	3.38	12.74	36	.1087	13
327	4.71	1.82	6.53	14	.0160	17
333	2.62	1.25	3.87	10	.0037	11
339	3.17	.74	3.91	11	.0253	26
357	3.13	.66	3.79	13	.0721	21
363	1.86	.88	2.74	17	---	19

DODGE ROAD SITE FINE AEROSOL DATA

Day	OC	EC	TC	Mass	(1/UH)	DD
3	2.11	.70	2.81	6	---	18
15	1.48	.48	1.96	6	---	11
21	1.62	.64	2.26	6	.0301	11
27	.85	.44	1.29	5	.0278	8
33	2.18	.40	2.58	7	.0330	6
39	.74	.06	.79	1	.0090	14
51	2.47	.39	2.86	10	.0760	0
57	1.46	.90	2.36	2	.0033	11
63	3.48	.69	4.17	9	.0861	0
69	2.53	.68	3.20	6	.0228	1
75	1.65	.66	2.31	4	.0022	4
87	2.18	.43	2.61	10	.0338	0
99	1.21	.12	1.33	5	.0145	0
105	2.45	.24	2.69	5	.0285	0
111	4.78	.72	5.50	15	.0206	0
117	2.46	.43	2.89	10	---	0
123	3.04	1.35	4.38	5	.0292	0
135	3.84	1.24	5.08	6	---	0
141	2.87	1.52	4.39	2	---	0
150	4.44	1.99	6.43	5	---	0
165	4.03	.70	4.73	8	.0406	0
171	3.33	.48	3.82	9	.0920	9
177	4.29	.92	5.21	11	.0358	3
183	2.33	.26	2.59	8	.0551	0
195	5.26	1.05	6.32	17	.0996	1
201	8.20	1.70	9.91	18	.0733	18
207	6.60	1.32	7.92	14	.0160	3
213	5.00	1.21	6.21	11	---	17
219	3.47	.93	4.40	5	.0321	14
225	8.99	2.91	11.90	11	.0797	27
231	3.68	1.46	5.15	5	.0163	13
233	3.71	.23	3.94	6	---	30
237	3.32	.72	4.04	4	---	27
239	4.11	1.09	5.21	10	.0348	29
243	8.74	3.13	11.87	20	.1640	25
249	2.95	.58	3.53	8	.0133	24
255	9.30	2.80	12.09	24	.1907	33
261	14.38	4.64	19.02	37	---	25
285	3.15	.64	3.79	7	.0089	31
291	9.40	3.12	12.51	21	.0478	23
297	9.13	2.90	12.03	22	.1362	24

Dodge Road Site Fine Aerosol Data (continued)

Day	OC	EC	TC	Mass	(1/UH)	DD
303	5.14	.84	5.98	12	.0315	37
309	2.86	.63	3.49	11	.0385	18
315	5.57	1.26	6.83	16	.0673	24
321	7.68	2.09	9.77	25	.1087	19
327	2.43	.49	2.92	9	.0160	17
333	1.00	.22	1.22	10	.0037	11
339	2.90	.46	3.36	5	.0253	20
345	2.69	.16	2.85	6	.0600	17
351	2.82	.02	2.82	6	.0615	23
363	1.95	.04	1.99	4	---	19

APPENDIX II

SEASONAL AVERAGES OF VARIOUS ELEMENTAL SPECIES IN JUSTICE BUILDING FINE AEROSOLS

In this section the normalization factor defined by Eqn. (11) will be used to normalize various elemental species data to obtain seasonal normalized data averages. Differences in these seasonal averages are proportional to differences in average seasonal emissions, within the accuracy limitations of the normalization procedure.

The data presented in Table A.2.1 were normalized using 1/UH normalization.

Table A.2.1 shows how the average normalized concentrations of various elemental species change by comparing summer and winter weekends and weekdays. Lead values, which can be assumed to be a measure of automotive activity, do not show a strong day or seasonal dependence. Sodium and to a lesser extent potassium, sulfur, and zinc, emission levels are larger on weekdays than on weekends. This may indicate that these species are associated with commercial or industrial activity. Since potassium, which is a component of residential wood smoke, does not show the same pattern as carbon, it appears that the existing potassium emitters are large compared to the potassium emission associated with residential wood stoves. Iron average normalized concentrations decrease greatly during the winter. This may result because the wetting action of winter rains decreases the resuspension of road dust and soil. In most urban aerosols soils are a major source of aerosol iron. Bromine average normalized concentrations show high weekend to weekday similarity, but unlike lead show a marked seasonal difference. Because bromine, as shown by the Pb/Br ratio, appears to be of automotive emissions origin, one would expect that the bromine average pattern would be similar to the lead average pattern. Differences probably involve the stability of aerosol bromine.

Normalized average seasonal elemental averages have also been evaluated using ambient lead concentrations in Eqn. (11) to compute the normalization factors. Seasonal weekend and weekday averages of lead normalized data are shown in Table A.2.2.

Certain species ratios, such as Br/Pb and C/Pb are useful in the identification of certain emission sources. Br/Pb ratios of about 0.33 are characteristic of automotive emissions (Watson, 1979). The Br/Pb ratio in gasoline is 0.39, but in automotive aerosol emissions the Br/Pb ratio is less because some of the bromine is emitted in the gaseous phase (Moyers et al., 1972). Source data in this study indicate that C/Pb ratios in the range of 5 to 10 are characteristic of automotive emissions, values of 90 are characteristic of road dust, and values of 300 or higher are characteristic of hog fuel emissions. Table A.2.3 presents these ratios and 95% confidence limits for the Justice Building fine fraction aerosol. The C/Pb ratios indicate that automotive emissions have the greatest impact on summer weekends and that at other times, especially on winter weekends, other carbon emitters play a greater role in the emission inventory.

TABLE A.2.1

Justice Building Meteorologically Normalized Fine Average Ambient Elemental Pollution Concentration
($\mu g/m^3$)

		SUMMER		WINTER	
Pb	WE (weekend)	0.22	0.07	0.24	0.16
	WD (weekday)	0.28	0.09	0.30	0.14
	All data	0.26	0.07	0.27	0.11
K	WE	0.53	0.12	0.51	0.27
	WD	0.96	0.15	0.69	0.26
	All data	0.86	0.15	0.70	0.35
S	WE	0.52	0.26	0.68	0.45
	WD	0.92	0.25	0.89	0.06
	All data	0.83	0.22	0.81	0.41
Zn	WE	0.022	0.01	0.021	0.01
	WD	0.037	0.02	0.047	0.02
	All data	0.034	0.02	0.037	0.01
Na	WE	0.45	0.07	0.49	0.26
	WD	1.6	0.39	1.1	0.59
	All data	1.4	0.38	0.84	0.40
Fe	WE	0.31	0.18	0.065	0.05
	WD	0.36	0.14	0.17	0.10
	All data	0.24	0.12	0.13	0.07
Br	WE	0.12	0.11	0.07	0.06
	WD	0.10	0.07	0.06	0.01
	All data	0.10	0.06	0.07	0.03

TABLE A.2.2

Justice Building Lead Normalized Fine Average Ambient Elemental Pollution Concentrations

($\mu g/m^3$)

		SUMMER		WINTER	
K	WE (weekend)	0.56	0.17	0.55	0.15
	WD (weekday)	0.83	0.18	0.59	0.16
	All data	0.76	0.18	0.57	0.11
S	WE	0.58	0.20	0.62	0.15
	WD	0.72	0.14	0.58	0.14
	All data	0.69	0.15	0.59	0.11
Zn	WE	0.022	0.01	0.048	0.04
	WD	0.027	0.01	0.036	0.01
	All data	0.026	0.01	0.040	0.01
Na	WE	0.59	0.30	0.54	0.17
	WD	1.4	0.45	0.77	0.25
	All data	0.22	0.45	0.68	0.17
Fe	WE	0.24	0.04	0.08	0.04
	WD	0.26	0.08	0.11	0.03
	All data	0.26	0.06	0.050	0.01
Br	WE	0.068	0.04	0.056	0.01
	WD	0.056	0.03	0.046	0.01
	All data	0.059	0.02	0.050	0.01

TABLE A.2.3

Useful Species Ratios from the Justice Building Fine Fraction Aerosol

		SUMMER		WINTER	
Br/Pb	WE (weekend)	0.25	0.10	0.29	0.05
	WD (weekday)	0.29	0.13	0.26	0.04
C/Pb	WE	0.41	0.12	1.69	0.86
	WD	0.79	0.18	1.25	0.30

VITA

John A. Rau was born in Albany, N. Y., on May 28, 1935. He graduated from Vincentian Institute High School in 1953 and obtained a B.S. in Mechanical Engineering from Rensselaer Polytechnic Institute in 1958. He worked for the Boeing Company from 1958 to 1963 and then for Pratt and Whitney Aircraft from 1963 to 1966. He obtained an M.S. in Mechanical Engineering from the University of Michigan in 1967, after which he worked for Eastman Kodak until 1970. He then worked for the Reed College Physics Department until 1974. In 1977 he entered the Oregon Graduate Center.