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REAL-TIME MEASUREMENT OF AEROSOL BLACK CARBON DURING THE CARBONACEOUS SPECIES METHODS COMPARISON STUDY*

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Abstract

During the Carbonaceous Species Methods Comparison Study, we used the aethalometer, an instrument developed at LBL that measures the concentration of aerosol black carbon ([BC]) in real time. Measurements were taken from August 12-21, 1986, with a 1-min time base and grouped to show 1-hr and multihour concentrations. We found concentrations generally ranging from 1 to $10\mu\text{g [BC]}/\text{m}^3$, usually increasing in the morning hours. We also observed short-duration (2-15 min) peaks in the black carbon concentration that could be directly attributed to the activity of vehicles in a delivery area less than 50m from the study site. We conclude that mobile sources were the major contributor to the short- and medium-term variability of aerosol black carbon measured at this site.

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Introduction

One of the components of the carbonaceous fraction of atmospheric aerosols is combustion-generated carbon having a microcrystalline graphitic structure. Due to its large optical absorption cross section, this material is termed "black carbon" (Novakov, 1984). It is produced only during incomplete high-temperature combustion; it cannot be formed, significantly transformed, or destroyed by atmospheric processes. It is therefore a direct tracer for combustion emissions.

Both chemical and optical methods for determining the black carbon content of aerosol samples collected on filters have been developed and are in common use at many laboratories. However, these measurements require that the aerosol be collected on the filter before analysis. It is therefore not generally possible (nor practicable, given the large number of analyses required) to obtain time resolutions of better than 1 to 2 hrs in most locations. Variations in emission rates and in transport and meteorology may occur more rapidly than this. It is therefore desirable to make real-time measurements of as many pollutants as possible, including black carbon. The aethalometer performs this latter measurement in real time, and its results are presented here for the 10-day period of the Carbonaceous Species Methods Comparison Study (CSMCS) conducted at Citrus College, Glendora, California, from August 12-21, 1986.

Experimental Details

The aethalometer operates by measuring the light transmission through a quartz fiber filter while air is being drawn through the filter (Hansen et al., 1982, 1984). The absorption of light in the visible spectrum by ambient aerosols is primarily due to the presence of microcrystalline graphitic carbon. When suspended as a submicron aerosol and not surrounded by a droplet of water or other adsorbed optically scattering material, the optical absorption cross section of this material is approximately $8 \text{ m}^2/\text{g}$ in the wavelength range 500-600nm (Roessler and Faxvog, 1979). When collected on a fibrous filter, multiple internal reflections enhance the optical absorption approximately threefold and nullify the effects of scattering from the particles (Rosen and

Novakov, 1983). Under these circumstances, the attenuation of light transmitted through the filter is due to the absorbing "black" carbon component of the aerosol and is not affected by light-scattering components such as organics, sulfate, and nitrate. The optical measurement on a filter-collected sample can be calibrated to provide a quantitation of aerosol black carbon (Gundel et al., 1984), with the relationship

$$\Delta \ln I = \sigma \Delta S .$$

The reduction of transmitted light intensity $\Delta \ln I$ is proportional to the incremental surface loading ΔS of black carbon on the filter, with the coefficient σ being the optical attenuation cross section. For quartz fiber filters, this coefficient has the value of $25.4 \pm 1.7 \text{ m}^2/\text{g}$ measured at a wavelength of 632 nm.

In the aethalometer, the ambient air with black carbon concentration $[\text{BC}] \text{ g/m}^3$ is drawn through one portion of the filter at velocity $V \text{ m/s}$. The increment of black carbon collected in time Δt will result in a decrease in the light transmission of

$$\Delta \ln I = \sigma [\text{BC}] V \Delta t .$$

This aerosol is collected on one portion of the filter above the "signal" photodetector; another photodetector located under a noncollecting portion of the filter acts as a reference. The filter is uniformly and diffusely illuminated; when ambient air is passed through the filter, the gradual accumulation of black carbon causes the "signal" intensity to decrease relative to the "reference" intensity. The electronics and microcomputer convert the detectors' outputs into an attenuation rate $\Delta \ln I / \Delta t$ from which the aerosol black carbon concentration can be calculated. The spectral output of the incandescent lamp and the response of the detectors gives an effective measurement spectral distribution with peak intensity between 500 nm and 700 nm. Previous work has shown that the absorption spectrum of ambient aerosols is essentially uniform and shows very little structure in the visible range (Rosen et al., 1978).

The collecting spot area of the filter was 1.14 cm^2 , and the air flow rates used ranged from 4 to 8 liters/min. The filter was changed usually every 6 hrs, resulting in a flow through the active filter area of between approximately 1 and $2.5 \text{ m}^3/\text{cm}^2$. With black carbon concentrations in the range of $2 \text{ to } 5 \mu\text{g}/\text{m}^3$, this resulted in filter loadings on the collecting area of $2 \text{ to } 10 \mu\text{g}/\text{cm}^2$.

Air flow through the aethalometer was monitored with a mass flow meter (Model 544, Kurz Instruments Inc.). This unit had a nominal range of 0-150 SLPM, with the most sensitive scale displaying 0-5 SLPM. Due to the fairly large black carbon concentrations encountered during the study, it was necessary to operate the aethalometer at low flow rates to avoid overloading the filter in the 6-hr measurement period. Under these conditions, the flow could not be read to an accuracy better than $\pm 5\%$. Upon return to the laboratory, the mass flow meter calibration was checked against a standard gas meter to correct for any offset or nonlinearity at these low flow rates. Over a 6-hr period, the flow rate was frequently observed to change by as much as 5% to 10%. Although these changes were accounted for in the analysis of the data, we consider that flow-rate uncertainties constitute the largest single error contribution to the overall measurement, limiting the reported black carbon concentration measurements to an accuracy of $\pm 10\%$.

The aethalometer was set up at the south end of the CSMCS platform, with its inlet at 8 ft above ground level. After some initial problems with flow rate variation and instability, it yielded good data on a 1-min basis for more than 95% of the time.

Results and Discussion

The results are available in three forms: (1) average black carbon concentrations for periods of usually 6 hrs from analyses of the filters, (2) hourly average black carbon concentrations, and (3) continuous black carbon concentration measurements on a time base of 1 minute.

Filter analyses. Thirty-four filters were collected by the aethalometer, mostly covering 6-hr periods with changes at 0200, 0800, 1400, and 2000 local time. The total optical attenuation of

the filter deposit as determined by the instrument is calculated from the ratio of initial to final optical transmissions, corrected for reference beam changes. This gives the black carbon loading per square centimeter of the filter, and hence the mean ambient black carbon aerosol concentration during the sampling period. The results are presented in Fig. 1. The filters were subsequently analyzed in the laboratory for black carbon content by a laser transmissometer that has been calibrated against the solvent extraction/quantitative thermal analysis for black carbon. The comparison of these two analyses confirms the calibration of the aethalometer head.

Hourly averages. A portion of the hourly average data is shown in Fig. 2, covering Monday, 8/18/86, through Wednesday, 8/20/86. These averages are calculated from the continuous aethalometer data.

One-minute data. The 1-min black carbon concentrations are calculated directly from the aethalometer data base and smoothed by means of a first-order autoregressive filtering algorithm of the form

$$S_t = [N \times S_{t-1} + C_t] / (N + 1),$$

where S_t is the smoothed value at time t , C_t is the raw data value, S_{t-1} is the previous minute's smoothed value, and N is the time constant. A value for N of 1 was used when processing the majority of the data.

Over the duration of the study period, 0800-8/12/86 to 0800-8/21/86, the system recorded 12252 1-min data points, representing a 94.5% coverage of the time. Most of the time out was due to data recording problems on 8/14/86 and 8/15/86. From 2000-8/15/86 to 0800-8/21/86 (i.e., 5-½ study period "days"), the coverage was 99.3%. This is the normal coverage expected of the instrument, with the total time out of about 10 min per day being the time necessary for four filter changes.

The hourly data format provides a convenient overview of the black carbon concentrations.

Hourly mean concentrations varied from 0.6 to 9.6 $\mu\text{g}/\text{m}^3$. With the exception of Sunday, 8/17/86, all the days show a clear peak in black carbon concentration in the early morning. We believe that this is due to vehicle activity in the delivery area for a shopping mall that adjoined the study site, with trucks often parked with motors idling less than 50 m from the aethalometer location. No other systematic diurnal variations are seen in the data.

Examination of the 1-min data clearly shows the impact of local plumes on the black carbon concentration. Figure 3 shows excerpts of the 1-min data for the early morning hours for Sunday, 8/17/86, through Thursday, 8/21/86. A clear event is seen every working day morning: on Wednesday morning, the 1-minute measurements of aerosol black carbon concentration rose from a steady value of approximately 3 $\mu\text{g}/\text{m}^3$ to a peak value of 9.5 $\mu\text{g}/\text{m}^3$ within 4 min. These weekday events in the data record all coincide with the following observer-noted occurrence: early morning every day except Sunday, a refuse truck would arrive in the adjacent lot (about 50 m from the sampling platform) and would drive back and forth servicing containers for a few minutes. Smaller individual events could also be identified. Late at night, two or three automobiles would occasionally park in a corner of the adjacent lot for a few minutes with their motors running. An observer-correlated event in the data record at 2212 on 8/18/86 is shown in Fig. 4. Similar events were seen in the data other days. Although some of these peaks consist of increases of only 20% to 30% above the preceding and following concentration levels, we feel that they represent the definite impact at the measurement location of plumes from individual sources, or from localized groups of sources.

Conclusions

During the August, 1986, Carbon Species Measurement Comparison Study conducted in the inland region of the California South Coast Air Basin, we performed measurements of aerosol black carbon in real time. The results from the filter analyses show 6-hr average concentrations ranging from 1.1 $\mu\text{g}/\text{m}^3$ to 6.5 $\mu\text{g}/\text{m}^3$, with the highest concentrations deduced from the filter

collected during the period 8 am-2 pm. Collecting the real-time data into hourly averages shows considerably more structure, with concentrations typically stable throughout the day, somewhat diminished overnight, but showing maxima between 7 am and 10 am every working day morning. Examination of the minute-by-minute data shows definite events in which individual vehicles in an adjacent property emitted plumes contributing up to $5 \mu\text{g}/\text{m}^3$ black carbon above the background level. The aethalometer has the sensitivity and time resolution to clearly identify these individual events. A measurement of aerosol black carbon is a tracer for the impact of combustion emissions at a site at any time scale. Over periods of hours, the measurements reflect fuel use patterns, local meteorology, and local to regional transport. On a time base of minutes, these scales are correspondingly reduced to those of individual source (e.g., vehicle) activity in the immediate neighborhood. Fourier analysis of the data might show definite frequency regimes (e.g., time scales of 1 week, 1 day, 1 hour, 1 minute) that could be associated with specific temporal features of source strengths and meteorology.

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Figure Captions

- Figure 1 Average aerosol black carbon concentrations for the periods covered by each filter (usually 6 hr).
- Figure 2 Excerpt of hourly average aerosol black carbon concentrations calculated from the continuous aethalometer data. Clear peaks are seen each morning.
- Figure 3 Excerpts of 1-min data for aerosol black carbon concentrations in the early morning hours of Sunday, 8/17/86, through Thursday, 8/21/86. The data for the working days cover the times at which a refuse truck was active in an adjacent lot: clear peaks in the black carbon concentration are seen to be contributed by this nearby source. The truck did not visit the site on Sunday.
- Figure 4 Excerpt of the 1-min aerosol black carbon data for the late evening of 8/18/86. Two automobiles parked in the adjacent lot for a few minutes with their motors running.

August 1986

XBL 881-9524

Figure 1

XBL 881-9522

Figure 2

Figure 3

XBL 881-9523

Figure 4

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