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GRAPHITIC CARBON TO LEAD RATIO AS A TRACER FOR THE SOURCES OF THE ARCTIC AEROSOL*

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Recent studies at the GMCC site at Barrow, Alaska, indicate the presence of large aerosol concentrations¹⁻³ which appear to be characteristic of the Arctic region as a whole.⁴ If one excludes the obvious natural aerosol components (e.g., sea salt and soil), the predominant components of the Arctic aerosol are carbon- and sulfur-containing particles. These particles occur at concentrations comparable to those found in urban areas in the continental United States.^{1,2}

There is considerable indirect evidence that a major part of the Arctic aerosol is produced from combustion processes. However, the strongest and most direct substantiation of this fact is provided by the identification of large concentrations of graphitic carbon in the Arctic.¹ These graphitic species, which have a structure similar to carbon black, can only be produced by combustion processes. The concentration of these particles at the GMCC

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sampling station at Barrow during the winter and spring approaches that found in urban areas¹ (i.e., the concentrations are only a factor of 4 less than the average values found in Berkeley, California, and Gaithersburg, Maryland, and a factor of 10 less than those found in New York City).

Associated with these graphitic species are large optical absorption coefficients ($\approx 10 \text{ m}^2/\text{g}$), which could produce a significant perturbation of the heat balance over the Arctic. Preliminary model calculations indicate that the present spring concentrations of graphitic particles could produce a change in the heat balance that is comparable to doubling the CO₂ concentrations.⁵⁻⁷

The sources of the Arctic aerosol are presently unknown. Part of the aerosol may be due to local Alaskan sources, but there is considerable evidence that a major fraction is due to long-range transport from other industrialized regions, ^{2,8,9} with Eurasia suggested as a major source area.² In this paper we report on the use of the graphitic carbon-to-lead ratio as a tracer for the sources of the Arctic aerosol.

An aerosol sampler has been operating at the GMCC station near Barrow since October 1979, collecting parallel filter samples on 47-mm quartz fiber and Millipore substrates at a flow rate of approximately 1.5 cfm. In this paper, we report on measurements of graphitic carbon and particulate lead concentrations from October 1979 through May 1980. The graphitic carbon concentrations were obtained using the LBL laser transmission technique¹⁰ with an empirical calibration curve developed for urban particulates in the United States.¹¹ Independent measurements of the graphitic carbon concentration by an optico-thermal analysis technique are under way. The lead concentrations were determined using x-ray fluorescence analysis.

Natural concentrations of particulate lead are very low, and it is usually

assumed that in polluted environments its major source is from the burning of leaded gasoline by automobiles. Typical ground level lead concentrations in urban atmospheres in the United States are $\simeq 1 \, \mu g/m^3$. In the Arctic the concentrations are much smaller, as shown in Fig. la, but are probably also combustion generated since the enrichment factors relative to the earth's average crustal abundance are large. For comparison the graphitic carbon concentration as a function of the time of year is shown in Fig. 1b. Although both aerosol components show their largest concentrations during the winter and spring, their seasonal variations are quite distinct. While the graphitic carbon concentrations are roughly constant from December through April, the lead concentrations show a rather sharp peak in November and December and relatively low values for the rest of the year. As a result of these differences in behavior, the relative concentrations of graphitic carbon to lead change substantially throughout the year, as shown in Fig. 2. These variations could represent the contributions of different source regions during different times of year or represent a seasonal variation in the fuel mix of given source regions. It should be emphasized that the results reported here are only for one year, and there could be considerable interannual variation due to variability in the air trajectories that arrive at Barrow from year to year.

Interpretations of these results are based on several crucial assumptions that need further investigation, including

1. The graphitic carbon to lead ratio does not change significantly during transport due to different deposition rates for particulate lead and graphitic carbon.

2. Source regions may be characterized by ground-level graphitic carbon to lead ratios (i.e., these ratios do not change appreciably with altitude).

It is interesting to compare the graphitic carbon to lead ratios in the Arctic to typical values in the Western and Eastern United States. As shown in Fig. 2, the values in the Arctic are considerably larger than those found in the United States, especially during the March-April time period. During this time period, the average values are 10 times that found in the Eastern United States and 30 times that found in the Western United States. If we use the graphitic carbon to lead ratio in the Western United States to represent a source region dominated by automotive emissions, these results could imply that source regions of the Barrow aerosol during the March-April time frame do not have a large automotive component relative to other sootgenerating combustion sources. For example, such a region could have a large diesel, wood-burning, or stationary source component relative to automobiles. Furthermore, these results could imply that the United States or regions with similar fuel mixtures do not contribute significantly to the Arctic haze during these periods of high graphitic carbon to lead ratios.

Acknowledgment

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Figure Captions

Figure 1a. Particulate lead concentration as a function of the time of year for 1979-1980.

1b. Graphitic carbon concentration as a function of the time of year for 1979-1980.

Figure 2. Ratio of graphitic carbon to particulate lead concentration as a function of the time of year for 1979-1980. Also shown are the range of values for the Western United States (Anaheim, Berkeley, and Fremont, California) and the eastern United States (New York, New York; Argonne, Illinois; and Gaithersburg, Maryland). The range of values is defined as those values within one standard deviation of the average.



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Figure 1b.



Figure 2

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