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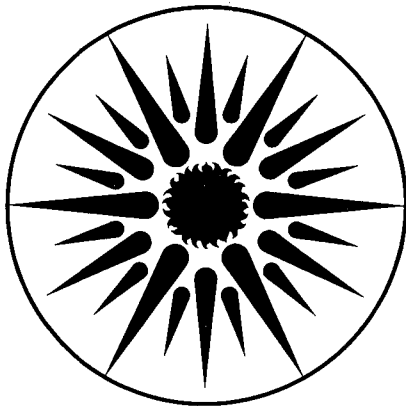
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ABSORPTION OF SOLAR RADIATION IN THE ARCTIC HAZE

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ABSTRACT

We have studied the vertical distributions of combustion-generated graphitic carbon particles and their associated absorption coefficients in the Arctic. These distributions show substantial concentrations of graphitic particles throughout the Arctic troposphere. Vertical profiles in the Norwegian Arctic can show both a strongly layered structure and an almost uniform distribution. Concentrations of graphitic particles within layers can be as large as those found in typical urban areas in the United States. The absorption optical depths associated with these vertical profiles are large enough to cause a substantial change in the solar radiation balance over a highly reflecting surface.

The phenomenon of Arctic haze, first reported by Murray Mitchell in the mid-1950's, is due to small aerosol particles that both scatter (Bodhaine et al., 1981) and absorb solar radiation (Rosen et al., 1981; Heintzenberg, 1982; Patterson et al., 1982), leading to substantial optical depths (Shaw, 1981). From both trace element analysis (Rahn and McCaffrey, 1980; Barrie et al., 1981) and direct identification of large concentrations of combustion-generated graphitic carbon particles (Rosen et al., 1981; Rosen and Novakov, 1983) in the Arctic atmosphere, it is known that the Arctic haze has a major combustion-derived component.

This component has a large optical absorption cross-section (~ 10 m^2/g) in the solar spectral region and could lead to significant heating effects over the high surface albedo polar icecap(Shaw, 1981; Porch and MacCracken, 1982; Cess, 1983). The magnitude of these effects depends largely on the vertical and horizontal distributions of these particles and their concentrations as a function of time of year. One of the major uncertainties in modeling the effects of the Arctic haze on the solar radiation balance has been the limited knowledge of the vertical distribution of the haze and, in particular, the vertical distribution of graphitic carbon particles and their associated absorption coefficients. In this letter and a companion letter (Hansen et al., 1984) , we report on the first measurements of such distributions in the Arctic atmosphere and, to the best of our knowledge, in the atmosphere in general.

During March and April, 1983, a series of flights (AGASP) organized by NOAA explored the vertical and horizontal distributions of the Arctic haze. One of the instruments used in this program was the aethalometer (Hansen et al., 1982), which was developed at Lawrence Berkeley Laboratory and has the capability of determining graphitic carbon concentrations on a real-time basis. This instrument measures the derivative of the optical transmission of particles collected on a highly reflecting filter medium (quartz fiber, Pallflex type 2500 QAO). The method has been shown both experimentally (Rosen et al., 1978; Yasa et al., 1979) and theoretically (Rosen and Novakov, 1983) to respond only to the absorbing component of the aerosol. The instrument is calibrated using the results of Gundel et al. (1984). The filters from the aethalometer can be analyzed after the flight to ensure that the aethalometer

response and the chemical analyses for graphitic carbon from the filter are consistent.

Vertical profiles were obtained by dividing the atmosphere into altitude bins and determining the average aethalometer response in each of these bins over the particular time periods of interest. The bin size was chosen so that the errors due to the minimum detection limit of the aethalometer were small compared to the average aethalometer response in the altitude bin. Error bars shown in the figures are based on three times the minimum detectable limit of the aethalometer and correspond to a change in the transmission of the filter deposit of 0.075%. The error bars are different at different altitudes because they are inversely proportional to the volume of air sampled in a given altitude bin.

Particles were also collected in parallel on Millipore filters (1.2 μm nominal pore size, type RATF) for x-ray fluorescence analysis of elements with $Z > 11$. The flows through both the aethalometer and the Millipore filter were constantly monitored by mass flow meters (Kurz Instruments Model 544). The mass flows were converted into volumetric flows using vertical density profiles obtained from a standard winter Arctic atmosphere (Handbook of Geophysics and Space Environments, 1965).

In Figs. 1 and 2, vertical profiles of graphitic carbon concentrations vs. altitude are shown for AGASP flights 8 and 10 in the Norwegian Arctic. Flight 8 originated in Bodo, Norway, on March 31, 1983, at about 0830 GMT. The flight was essentially in a northerly direction over Bear Island to northern Spitzbergen at $80^{O}N$ $15^{O}E$ and then returned to Bodo. The flight profile included a slow descent from 7 km to .1 km at $74^{O}N$

25°E near Bear Island. The descent started at 1030 GMT and finished around 1145 GMT. The graphitic carbon concentrations as a function of altitude are shown for this descent in Fig. 1. Also shown for comparison are the average ground-level concentrations found at the NOAA-GMCC observatory near Barrow, Alaska, for April, 1982, and the annual average ground-level concentrations found in various urban areas in the United States. It is clear from this figure that the vertical profile has considerable structure with at least three layers located at approximately 1 km, 2.5 km, and 4.5 km and what appears to be a very narrow layer at 1.7 km. The haze layers can have sharp boundaries with dramatic changes in concentration over distances less than 100 m. These bands appear to be superimposed on a background of haze that decreases with altitude. The concentrations within these layers are large, with the the peak concentrations at 1 km as large as typical ground-level concentrations in the United States (Berkeley, Denver, Gaithersburg) and only about a factor of 2.5 lower than those in New York City. The concentrations in this band are about a factor of 10 higher than the 0.15 $\mu g/m^3$ April, 1982, ground-level concentrations at Barrow, Alaska. The flight profile also indicates a relatively clean region at low altitudes, which is consistent with the Barrow results (Hansen et al., 1984) and the limited ground-level measurements made in the Norwegian Arctic.

Flight 10 also originated in Bodo, Norway, but flew west across the Norwegian sea on April 5, 1983, to an area near 70°N 0°E over the open ocean and then returned to Bodo. The plane remained within 70°±2° latitude and ranged from 5°E to 5°W in longitude between 0914 GMT and 1604 GMT. The average vertical profile of graphitic carbon concentrations for this period is shown in Fig. 2a. This profile is quite distinct

from the one shown in Fig. 1: it has much less structure with only one sharp band at 3.75 km superimposed over an almost uniform haze layer, decreasing only slightly with altitude even close to the top of the troposphere. If this profile is plotted vs. the mixing ratio, as in Fig. 2b, it is essentially flat to the top of the troposphere with a very intense band located at about 3.75 km. This profile could indicate significant transport at high altitudes above the planetary boundary layer. However, it may also be indicative of changes in the vertical distribution that have occurred after the air mass has reached the Arctic.

Recent modeling studies of the possible effects of the Arctic haze on the radiation budget of the Arctic indicate rather substantial changes in the heat balance if the optical depth due to absorption is sufficiently large (Porch and MacCracken, 1981; Cess, 1983). One can calculate absorption coefficients and optical depth from the results shown in Figs. 1 and 2 for various models of the Arctic aerosol. In these models, it is important to distinguish between graphitic particles mixed internally or externally with the other nonabsorbing components. Ackerman and Toon (1981) showed that such differences can lead to significant changes in aerosol absorption. We will consider three examples:

Model 1. Graphitic carbon particles internally mixed with all the major submicron aerosol components (i.e., sulfates, organics).

Model 2. Graphitic carbon particles internally mixed with only submicron organic aerosol components.

Model 3. Graphitic carbon particles externally mixed with the other aerosol components.

Of these three, the first two are the most likely combinations. For these, we treat the particles as homogeneous spheres and mix in the various components by volume mixing of dielectric constants (Chylek et al., 1981). In these Mie calculations the indices of refraction of graphitic carbon were chosen to be 1.94–0.66 i (Ackerman and Toon, 1981), with a density of 1.5; and the index of refraction of the nonabsorbing components was taken to be 1.5. The relative concentrations of graphitic carbon and sulfates were determined from chemical analyses of filters collected on the flights over the time periods of the vertical profiles, while the relative concentrations of the organic component were obtained from ground-level measurements in April at Barrow, Alaska, where detailed chemical analysis of the submicron aerosol has been completed. The particle size distribution for the submicron aerosol is assumed to be log-normal with r_{gv} = 0.2 μm and σ_g = 2, which are obtained from Whitby (1978) for the urban aerosol and are consistent with the results of Heintzenberg (1982) for the Arctic aerosol. For the external mixture, we use the photoacoustic measurements of Roessler and Faxvog (1979) and Szkarlat and Japar (1981) who obtain an average value of 8.3 m^2/g for a wavelength of 0.5 μ.

For the vertical profile in flight 8, the three models yield specific absorption coefficients respectively of 18.1 m^2, 12.0 m^2, and 8.3 m^2 per gram of graphitic carbon. The corresponding results for flight 10 are 14.6 m^2, 12.0 m^2, and 8.3 m^2 per gram of graphitic carbon. Vertical profiles of the absorption coefficient using aerosol model 1 are shown for flights 8 and 10 in Figs. 3 and 4. The absorption coefficients for the other models can be determined from these figures from ratios of specific absorption coefficients. These absorption

coefficients can be integrated over altitude to determine absorption optical depths for the various flights and aerosol models. For flight 8, the three aerosol models yield optical depths at 0.5 μ of 0.053, 0.035, and 0.024; and for flight 10, the corresponding optical depths are 0.036, 0.029 and 0.020. It is interesting to compare these optical depths to those obtained by Valero et al. (1983) using a net flux radiometer. Such a comparison can be made for the vertical profile from flight 8 shown in Fig. 1, where Valero et al. (1983) have completed their analyses. They obtain a best-fit optical depth due to absorption of 0.06, which is very close to the largest value we report and could suggest that the aerosol may be completely internally mixed as represented in Model 1.

These optical depths reported here are large enough to produce significant changes in the radiation balance over a highly reflecting surface (Porch and MacCracken, 1981; Cess, 1983). This is illustrated by the calculations of Porch and MacCracken (1981) and of Cess (1983), who have modeled the Arctic aerosol for an absorption optical depth of 0.021 (which is close to our minimum estimate), cloud-free conditions, a zenith angle of 72°, and a surface albedo of 0.8. For these parameters, they obtain a change in the noontime solar radiation balance at the top of the troposphere of about 20 W/m^2. When averaged over the day for March 15 at 70°N, the surface-atmosphere energy absorption would increase by about 7 W/m^2 (Porch and MacCracken, 1981; Cess, 1983). These changes are substantial and would correspond to an increase in the energy absorbed by the earth-atmosphere system of approximately 5% of the incident solar flux at the top of the troposphere. Correspondingly larger changes would be expected for the internally mixed aerosol

models. These first estimates of the effects of graphitic carbon on the Arctic radiation balance are based on the flight profiles shown in Figs. 1 and 2, which are obviously only over small geographical regions and in narrow time frames. Similar vertical and horizontal profiles in other areas of the Arctic will be available in the future.

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FIGURE CAPTIONS

Figure 1. Vertical profile of graphitic carbon concentrations
expressed as nanograms per cubic meter on March 31, 1983, at ~ 74°N
25°E. Shown for comparison are the annual average ground-level graphitic
carbon concentrations at various urban locations in the United States
and the average April, 1982, ground-level values at the NOAA-GMCC obser-
vatory near Barrow, Alaska.

Figure 2a. Vertical profile of graphitic carbon concentrations
expressed in nanograms per cubic meter on April 5, 1983, at ~ 70°N 0°E.

2b. Same as the above but with graphitic carbon concentrations
expressed as a mixing ratio.

Figure 3. Vertical distribution of absorption coefficients obtained
from the graphitic carbon profile shown in Fig. 1 using aerosol model 1.
The corresponding absorption coefficients for models 2 and 3 can be
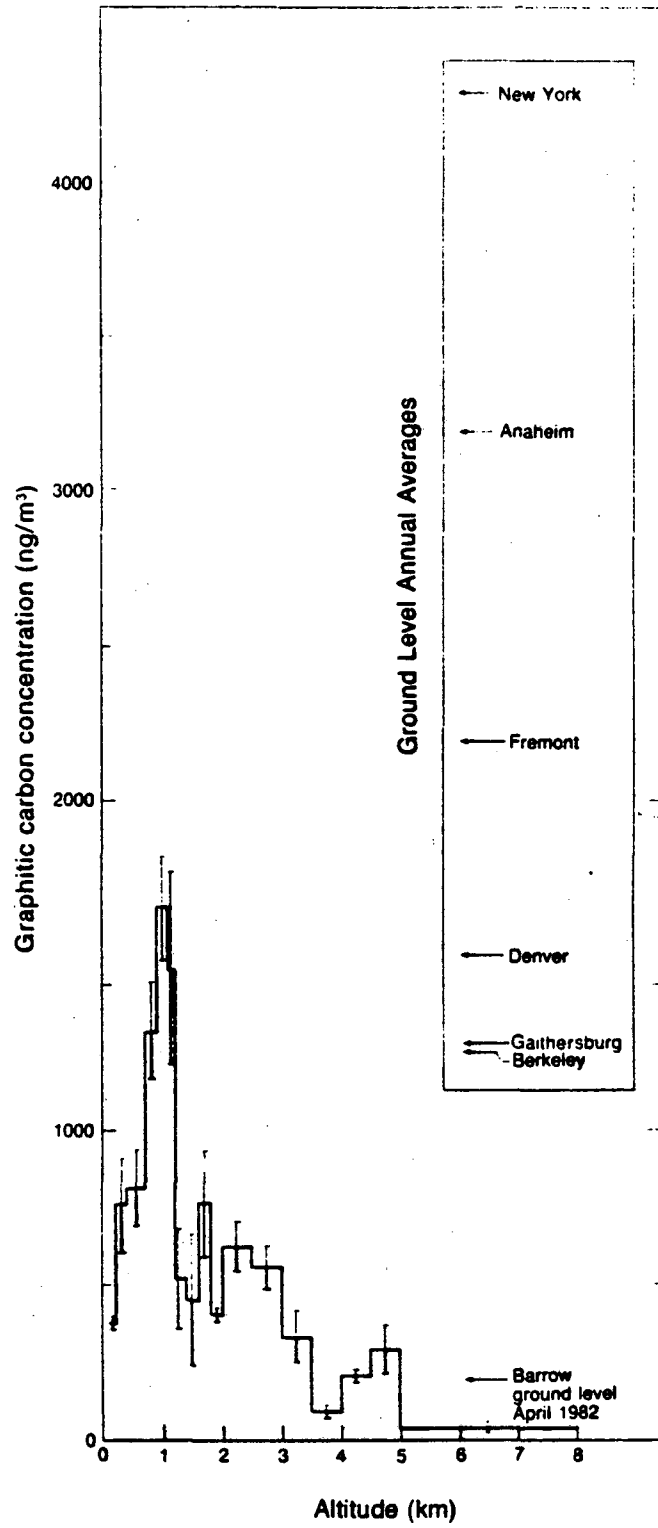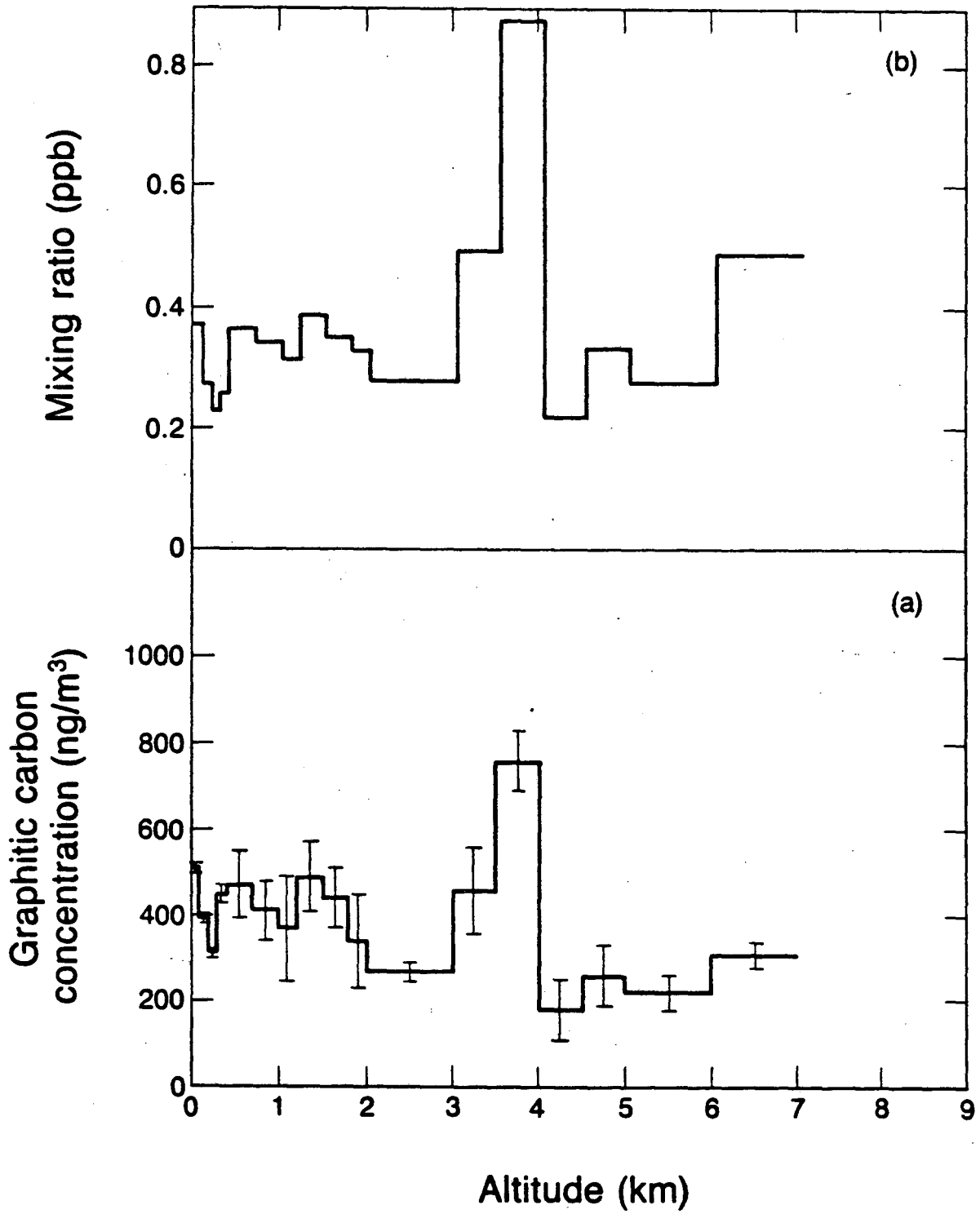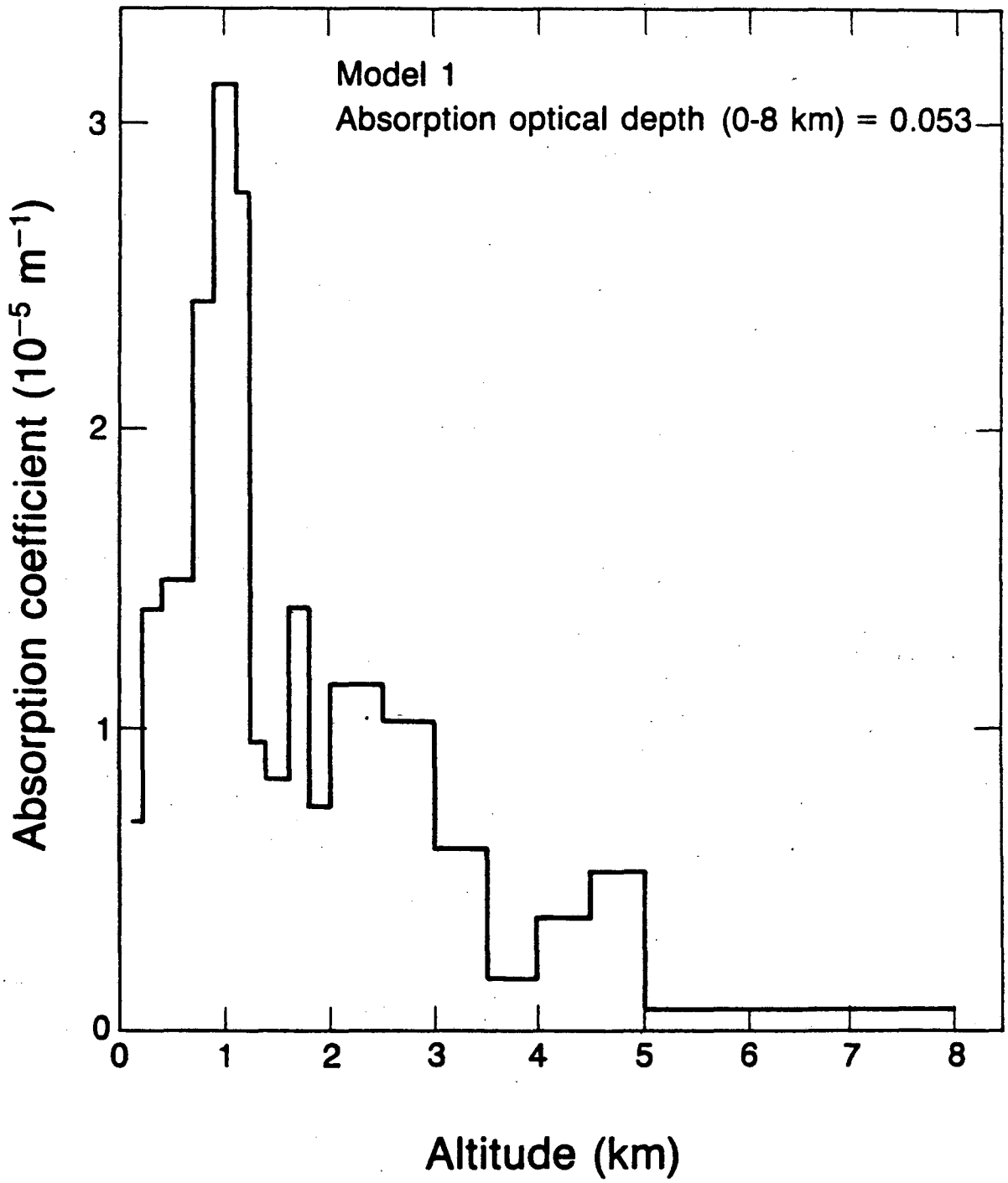obtained by multiplying the distribution by factors of 0.66 and 0.46
respectively.

Figure 4. Vertical distribution of absorption coefficients obtained
from graphitic carbon profile shown in Fig. 2 using aerosol model 1.
The corresponding absorption coefficients for models 2 and 3 can be

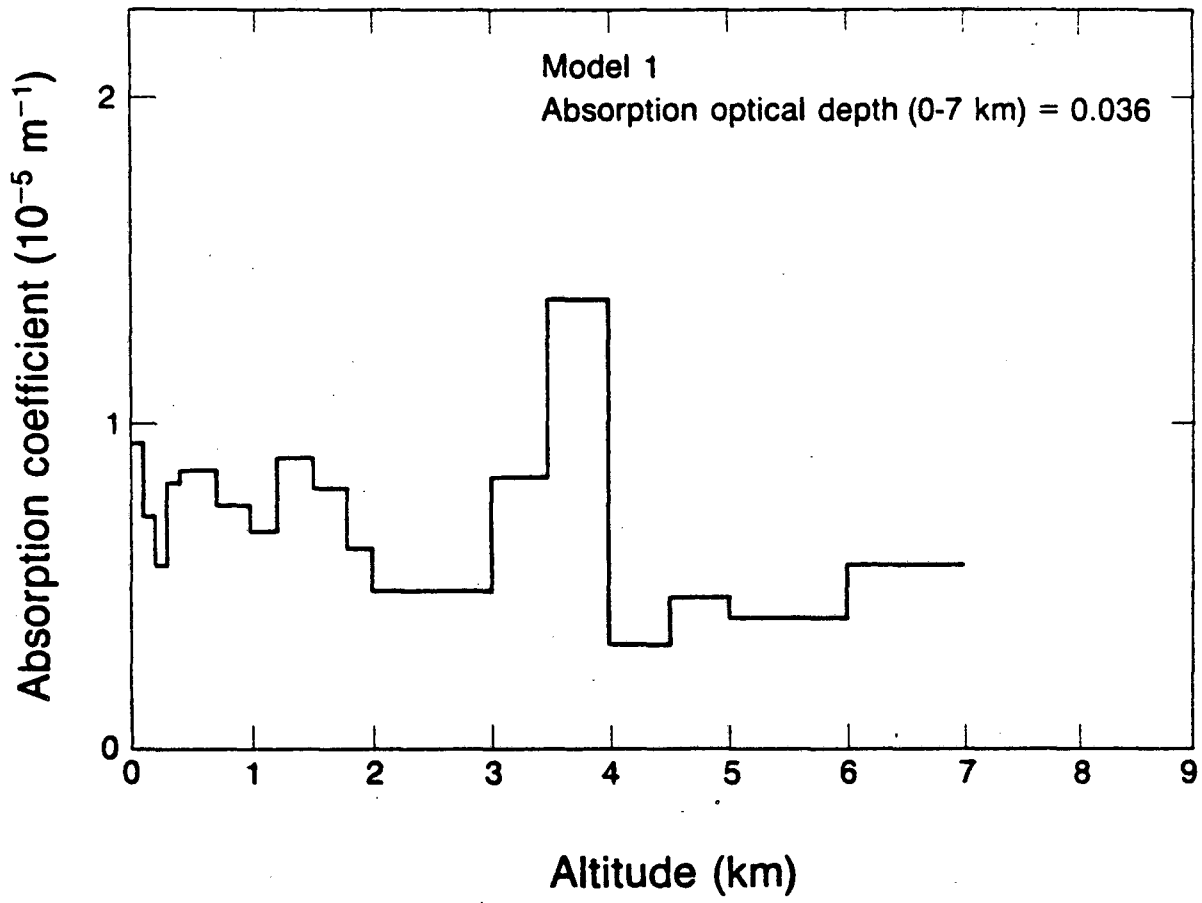obtained by multiplying the distribution by factors of 0.82 and 0.57 respectively.

Figure 1

Figure 2

Figure 3

XBL 8312-4317

Figure 4

XBL 8312-4318