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OPTICAL ABSORPTION BY AEROSOL BLACK CARBON AND DUST IN A DESERT REGION OF CENTRAL ASIA

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Abstract—In September 1989 a joint U.S.S.R.–U.S. study of the causes and effects of desert dust on the environment was conducted in the Tadzhik S.S.R. in Soviet Central Asia. The objectives of the study included measurements of optical absorption by suspended material, both windblown dust and aerosol "black" carbon. This latter material is a combustion effluent, prevalent in emissions from poorly controlled burning, with a long atmospheric lifetime and a large cross-section for the absorption of visible radiation. The measurements obtained from the analysis of filter samples indicate that only during periods of active dust production was there a significant contribution of dust to total absorption. At other times, the presence of black carbon from local and regional sources accounted for approximately 90° of the total aerosol optical absorption. The conclusions are that fuel combustion may produce a greater optical impact on the atmosphere in less-developed areas of the world than that arising from the effects of desert dust production.

INTRODUCTION

A joint U.S.S.R.–U.S. study of the causes and effects of desert dust in the context of anthropogenically induced desertification was conducted in the Tadzhik S.S.R. in Soviet Central Asia in September 1989. This study involved the participation of many research groups and organizations on both sides (Gillette *et al.*, 1993, this issue), under the provisions of the 1988 Protocol of Working Group 8 of the U.S.–U.S.S.R. Bilateral Agreement on Environmental Protection. Three sites were set up for the study of atmospheric physical and chemical parameters in the arid valley of the Kafirnigan River, in the southeast portion of the Tadzhik S.S.R. Synoptic, local and micro-meteorological parameters were measured, photometric measurements were taken, and satellite images of the region were obtained for the study period. The objective from the point of view of atmospheric optics is to produce a representation of optical absorption and scattering as a function of dust loading, consistent with photometric and satellite data, that may be used as input to models calculating the impact of anthropogenic desertification on atmospheric radiative transfer. Other U.S. investigators were also involved in measurements of aerosol optical properties (Patterson *et al.*, 1993, this issue). In this paper we concentrate on measurements of optical absorption and their interpretation in terms of the presence of two species: "dust" and aerosol "black" carbon.

Aerosol "black" carbon (BC, sometimes referred to as "elemental" carbon, EC) is a universal component of the effluent from the combustion of carbonaceous fuels (Wolff, 1984; Goldberg, 1985). Its emission factor, defined as the mass of aerosol BC emitted per unit mass of carbon consumed in the fuel, is highly variable and depends to a great degree on the nature of the fuel and the combustion process (Hansen *et al.*, 1978). In general, the higher the temperature and the better controlled the combustion, the lower the BC emission factor. This factor has been measured as ranging from less than 10^{-6} for emissions from industrial-scale boilers powered by natural gas, to greater than 10^{-6} for the burning of a pile of asphaltic roofing material as encountered in a residential conflagration (Dod *et al.*, 1989). More typical values for the average emissions from an area-wide source are around 10^{-4} for an urban mix of sources in a developed country, increasing by perhaps one order of magnitude to around 10^{-3} for regional emissions in a less-developed area.

Aerosol black carbon has an absorption cross-section of approximately 10 $m^2\ g^{-1}$ in the visible spectrum, making it by far the strongest absorber commonly encountered in the atmosphere (Roessler and Faxvog, 1980). It has a long atmospheric residence time, and has been detected at the most remote locations on the globe (e.g. at the South Pole, Hansen *et al.*, 1988). Typical concentrations of BC range from $>10\ \mu g\ m^{-3}$ in some urban areas, to 0.1–1 $\mu g\ m^{-3}$ in rural areas of the U.S., to 0.01–0.1 $\mu g\ m^{-3}$ in remote regions of the Northern Hemisphere. The optical absorption coefficient of desert dust is generally measured to be at least two orders of magnitude smaller, i.e. in the region of 0.03–0.1 $m^2\ g^{-1}$ (Patterson *et al.*, 1977; Royer *et al.*, 1983). Thus, dust loadings must exceed BC concentrations by at least a factor of 100 if the dust is to make a significant contribution to the overall optical absorption. In remote desert locations devoid of human impact this situation may indeed be the case: however, the long-range transport of aerosol black

carbon from regions of copious fuel combustion may change this situation if the study area is downwind of anthropogenic activities.

EXPERIMENTAL DETAILS

Aerosol samples were collected over periods of one to a few hours using a low-volume sampler at two locations during the study period. From 6 to 18 September, the sampler was set up at Site 2, a rural location at the village of Isenbai. From 18 to 21 September, the sampler was moved to Site 3, in the grounds of the Tadzhik Physical–Technical Institute in the city of Dushanbe. This city has a population of approximately 300, 000 people, with copious fuel combustion emissions visible and a distinct air pollution problem. The Site 2 location was an approximately 70 km distance from Dushanbe. Further details of the site characteristics are presented by Gillette *et al.* (1993, this issue).

Samples were collected on pre-fired quartz fiber filters at an air flow rate of approximately 0.5 $m^3 h^{-1}$. The aerosol was concentrated onto a central spot on the filter that was defined by interchangeable masks. Normally, masks of 0.5, 1 and 2 cm^2 were used. The aerosol inlet configuration involved no radius turns, and so it was hoped that losses of large dust particles would be minimized. After collection, the samples were stored in Petri slides and subsequently returned to Berkeley for analysis. The deposits of carbon and dust are both chemically and mechanically stable, and it is our experience that no substantial losses or errors are introduced during storage or transit.

The filter samples were analysed by a combination technique employing an optical transmission attenuation measurement before and after heat treatment in a furnace. We measure the reduction in intensity of a broad spectrum light beam projected through the aerosol sample collected on the filter, relative to the intensity transmitted through an adjacent blank area of the filter. Quartz filters have a deep mat of optically scattering fibers that produces multiple reflections of light incident on the trapped particles. This has two consequences: first, it nullifies the effects of optical scattering by the particles on the net transmission of light through the sample (thus ensuring that the measurement of absorption is not subject to interference by aerosol scattering); second, the multiple reflections enhance the absorption signal by a factor of approximately two (Rosen and Novakov, 1983; Gundel *et al.*, 1984). A measurement of optical attenuation may therefore be interpreted as a measurement of the surface loading of absorbing aerosol species unaffected by scattering aerosol species, and converted to a concentration given the sampled air flow volume. For samples taken at non-dusty locations, this absorption is attributed entirely to aerosol black carbon. However, in dusty areas such as the target of the present study, it is necessary to consider the contributions of both the BC and the dust. The optical attenuation of the original sample is

$$\mathrm{ATTEN}(1) = \mathit{Sigma}(\mathrm{BC}) * [\mathrm{BC}] + \mathit{Sigma}(\mathrm{Dust}) * [\mathrm{Dust}] \quad (1)$$

where *Sigma* denotes the absorption cross-section of each species, [BC] represents the mass per unit area of black carbon in the sample spot on the filter, and [Dust] represents the mass per unit area of dust on the filter.

We now employ another useful feature of quartz fiber filters: they may be fired to a high temperature without losing structural integrity or changing their optical properties. If the sample is heated to a temperature of approximately 600°C in air for a couple of minutes, the carbon content of the aerosol sample will be completely burned away, and the BC content of the sample will be reduced to zero (Gundel *et al.*, 1984). However, the dust loading will generally not be affected, as dust is a refractory material. Assuming that the optical properties of the dust are not greatly changed by the firing process, the optical attenuation of the sample after furnace firing will be

$$\mathrm{ATTEN}(2) = \mathit{Sigma}(\mathrm{Dust}) * [\mathrm{Dust}] \quad (2)$$

with no contribution from carbon. We observe that a sample that was originally of a dark grey coloration will lose the blackness after firing, with a small amount of reddish-brown coloration remaining.

Subtracting the above two measurements allows us to calculate the amount of aerosol black carbon in the mixed sample, and also to calculate the contribution of each species to the overall optical absorption of the original total aerosol. It is convenient to express the absorption due to dust in terms of a "black carbon equivalent", i.e. the amount of BC that would be needed to produce the same amount of absorption. If the absorption coefficient of dust is 1000 times smaller than that of black carbon, then a dust absorption component "equivalent" to 1 $\mu g\, m^{-3}$ BC represents an actual dust loading of 1 $mg\, m^{-3}$.

Ninety-one samples were collected and analysed for optical transmission attenuation before and after firing in a furnace. The optical absorption component that burned off was attributed to aerosol black carbon: that remaining after firing was attributed to dust. The dust absorption was expressed as "equivalent" BC for ease of comparison.

Light winds were observed predominantly from Dushanbe (down-valley in direction) from the beginning of the experiment until 16 September (Smirnov *et al.*, 1993, this issue). During the dust storm of 16 September, stronger winds directed up-valley towards Dushanbe were measured. Generally, observers noted an abundance of smoldering fires which resulted in a smokey atmosphere under the light wind conditions in the valley from 6 to 15 September.

RESULTS AND DISCUSSION

Figure 1 shows optical absorption results for the samples collected at Site 2 (Isenbai village, 70 km distant from Dushanbe), from 6 to 18 September 1989. The BC concentrations generally ranged from 1 to 8 $\mu g\, m^{-3}$: dust absorption expressed as "equivalent BC", ranged from 0.1 to 1 $\mu g\, m^{-3}$ BC equivalent, i.e. 0.1–1 $mg\, m^{-3}$ of dust if the dust absorption cross-section is 1000 times smaller than that of black carbon. Large excursions are seen in the black carbon data, but relatively little variation was seen in the dust component of optical absorption.

The percentage contribution of dust to the total optical absorption of the samples is plotted in Fig. 2 for these data. The results show a clear periodicity with a diurnal cycle peaking in the afternoon and having a minimum in the early morning hours. This finding is shown in more detail in Fig. 3, which displays the mean dust contribution to total optical absorption for each hour (local time). From 3 to 10 a.m., the dust contribution is from 6 to 8%, with little variability for the same hour over the 12-d period. Note that this period did not include the dust storm of 16 September. This implies that aerosol black carbon (e.g. local or regional "smoke") contributed from 92 to 94% of the total optical absorption during these hours in this two-week period of light winds. From 3 p.m. onwards, the dust contribution increased by a factor of from two to three times, to values from 12 to 15%,

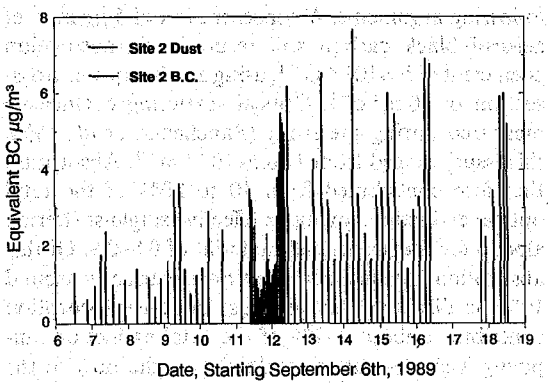

Fig. 1. Optical absorption of particulate material collected on time-integrated filter samples at the rural location, Site 2, from 6 to 18 September 1989. The filters were analysed before and after firing in a furnace, to separate the combustible black carbon and the refractory dust absorption which is shown as "equivalent BC", i.e. the amount of BC that would be required to produce the same optical absorption (see text). The upper points show the BC concentrations, the lower points the dust "equivalent" BC.

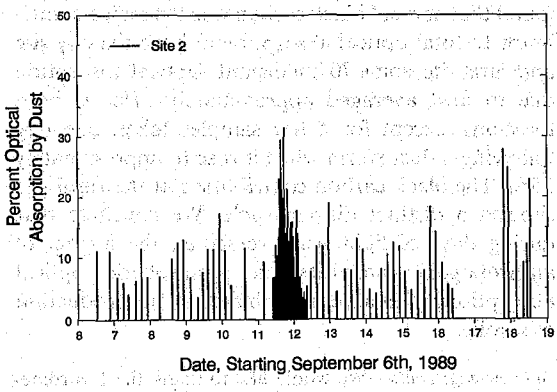

Fig. 2. The percentage contribution of dust to the total optical absorption of the samples collected at Site 2 from 6 to 18 September 1989. Note the diurnal periodicity.

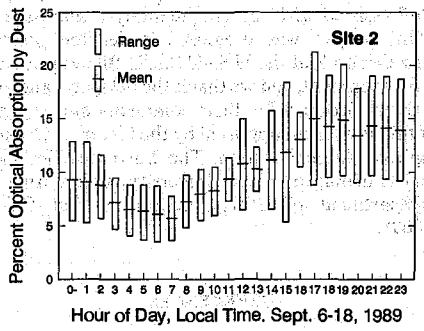

Fig. 3. Percentage contribution of dust to total optical absorption for all samples collected at the rural location, Site 2, from 6 to 18 September 1989, grouped by hour of day (local time). The result from each time-integrated sample is distributed equally over the hours that the sample covered. The mean is shown by the central line, the S.D. by the extent of the bars.

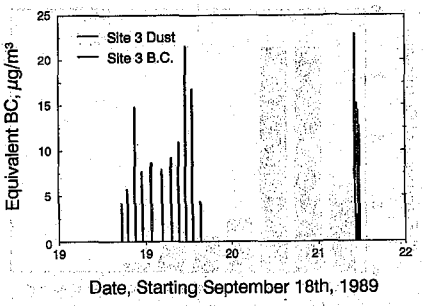

Fig. 4. Aerosol black carbon and dust "equivalent" BC concentrations for samples collected at Site 3 in the city of Dushanbe from 18 to 21 September 1989. The higher dust optical absorption on 21 September is attributed to a dust storm that occurred the previous day.

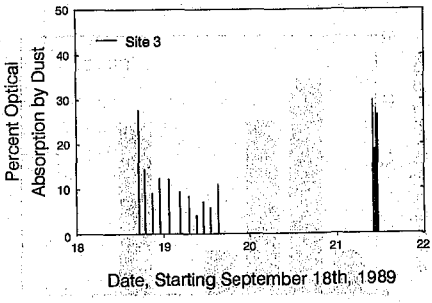

Fig. 5. Percentage contribution of dust to the total optical absorption of the samples collected at Site 3. Note the high contributions of residual dust after the dust storm of 20 September.

with considerably greater variability. This result is probably correlated with anthropogenic activities that may raise dust by vehicular traffic during the day, and the accumulation of pollutants (i.e. smoke) at night.

The data from Site 3 in the city of Dushanbe are shown in Fig. 4 and 5. Note first that the scale of total optical absorption is greatly increased: BC concentrations ranged from 4 to 20 $\mu g\ m^{-3}$. The dust contribution to total absorption was similar to Site 2, except for the set of samples taken at the end of the study period on 21 September 1989. A dust storm occurred on 20 September, although samples were not taken that day. We see that the dust contribution to absorption increased on 21 September, possibly due to residual dust transported into the city. There are insufficient data from this location to determine any diurnal cycle in the dust vs black carbon contributions to optical absorption.

Further analysis is presented in Fig. 6 and 7, which show distributions of the dust contribution to total aerosol optical absorption at the two locations. At Site 2, the analysis of 76 samples showed that dust contributed $11.3 \pm 6.2\%$ of the total absorption (arithmetic mean $\pm$ S.D.). A few samples showed elevated dust

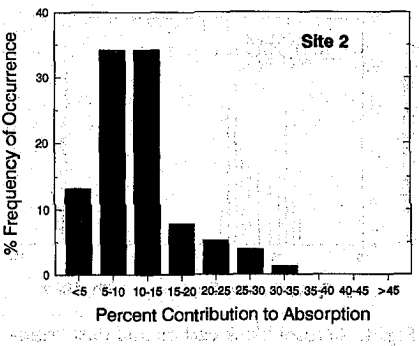

Fig. 6. Distribution of the percentage contribution of dust to total optical absorption for all the samples collected at the rural location, Site 2. For the total number of 76 samples, the mean contribution of dust to optical absorption was $11.3 \pm 6.2\%$.

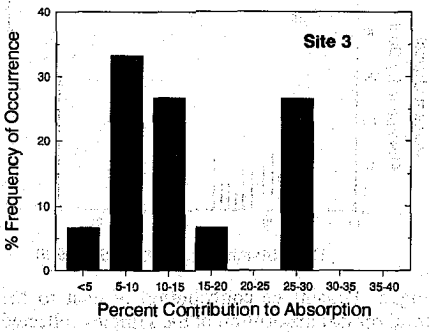

Fig. 7. Distribution of the percentage contribution of dust to total optical absorption for the samples collected at Site 3 in the city of Dushanbe. For the first 11 samples, the mean contribution of dust to optical absorption was $11.2 \pm 6.0\%$, almost identical to that for the samples collected at the rural location. For the last four samples taken one day after a dust storm on 20 September, the mean contribution of dust was $25.8 \pm 4.1\%$. These last samples are represented by the right-most bar of the histogram.

contributions, over 20% but these cases were the exception. In Dushanbe, the analysis of the first 11 samples showed an almost identical result, giving a dust contribution of $11.2 \pm 6.0\%$. The final four samples, taken on 21 September (the day following the dust storm), gave a mean dust contribution of 25.8 $\pm 4.1\%$ to the total optical absorption. These results show that aerosol black carbon due to fuel combustion is the dominant source of optical absorption by suspended material in this study, for the sampling periods spanning times outside of the actual occurrence of dust storms. Unfortunately, we are not able to compare these results with measurements during dust storms.

The effect of this absorption on optical path extinction in the atmosphere may be estimated by the following arguments. A concentration of 5 $\mu g\ m^{-3}$ of aerosol black carbon will result in an absorption coefficient of $50*10^{-6}\ m^{-1}$, using an absorption cross-section of $10\ m^2\ g^{-1}$. Optical scattering coefficients measured during the study (Panchenko *et al.*, 1993, this issue) ranged from 1 to $5*10^{-4}\ m^{-1}$. Absorption therefore contributed from 10 to 20% of the total optical extinction, giving an effective single-scattering albedo for the suspended material of 0.8–0.9. Of this absorption, the dust contribution was usually around 10% as discussed above: thus, the dust absorption contributed about 1–2% of the total extinction, implying a single-scattering albedo for the dust in the range of 0.98–0.99.

CONCLUSIONS

We participated in a desert-dust study in an arid region of Soviet Central Asia, and analysed total aerosol samples collected on quartz fiber filters with a procedure that allows a separation of the absorption due to (refractory) dust from that due to (combustible) aerosol "black" carbon, a common pollutant. We found that aerosol black carbon was the major contributor to total optical absorption at both the city site and rural site, some 70 km distant. Optical absorption due to dust averaged approximately 11% at both locations, except for a few samples taken one day following a dust storm when it rose to approximately 25%. The black carbon contribution at the rural site showed a distinct diurnal cycle. We conclude that during days of light wind velocities, the impact of anthropogenic activities on atmospheric optical absorption in this area is dominated by the production of smoke.

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