

Dominance of mineral dust in aerosol light-scattering in the North Atlantic trade winds

X. Li, H. Maring, D. Savoie, K. Voss & J. M. Prospero

University of Miami, Rosenstiel School of Marine and Atmospheric Science, 4600 Rickenbacker Causeway, Miami, Florida 33149, USA

ATMOSPHERIC aerosols can affect climate by scattering and absorbing solar radiation¹⁻³. Most recent studies of such effects have focused largely on anthropogenic sulphate aerosols, which are believed to exert a substantial cooling influence². Mineral dust aerosols have been largely ignored, because it was thought that their scattering efficiency and concentrations were too low to have a substantial effect on climate. Here we report measurements of the light-scattering properties of North African dust delivered to Barbados by the North Atlantic trade winds. Although the mass scattering efficiency of the dust is only about a quarter of that of non-seasalt sulphate over the North Atlantic², the annual-mean dust concentration in Barbados trade-wind air is 16 times that of non-seasalt sulphate⁴. The net scattering by mineral dust is therefore about four times that by non-seasalt sulphate aerosols. African mineral dust should therefore be the dominant light-scattering aerosol throughout the tropical and subtropical North Atlantic region. Our observations suggest that mineral dust could be an important climate-forcing agent over this ocean region and in other regions where dust concentrations are high^{7,8}.

Climate models incorporating aerosols suggest that anthropogenic non-seasalt sulphate (n.s.s. SO_4^{2-}) aerosols in the Northern Hemisphere scatter as much as $1-2 \text{ W m}^{-2}$ of incoming solar radiation back to space^{2,4}, yielding a cooling rate comparable to the heating rates attributed to greenhouse gases^{4,9}. As a consequence, climate-related aerosol measurement programs have focused largely on anthropogenic n.s.s. SO_4^{2-} .

In many regions, however, mineral dust is a common aerosol component^{7,8}. Large quantities of dust are transported from sources in North Africa across the tropical Atlantic, as documented by measurements made continuously on Barbados ($13^\circ 15' \text{ N}$, $59^\circ 30' \text{ W}$) since 1965^{3,10,11}. Satellite imagery shows that dust out-breaks take about a week to reach the Caribbean¹²; the dust clouds typically cover a large area in a latitude band $10-20^\circ$ wide, reaching to altitudes of 3-6 km (refs 12-14). Dust is the dominant aerosol constituent in this region during the spring, summer and early autumn when dust events are most common^{3,10}.

We measured aerosol physical and chemical properties from 4

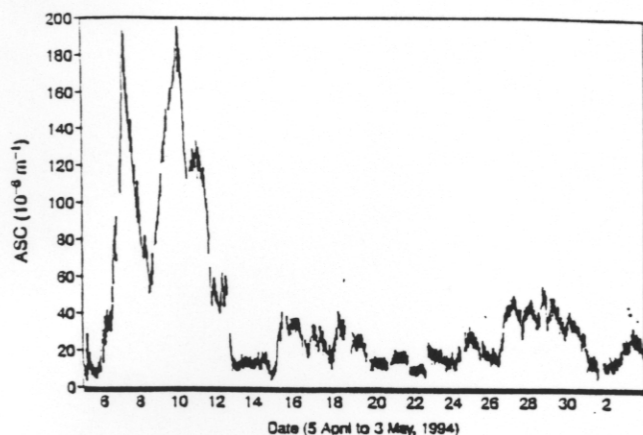
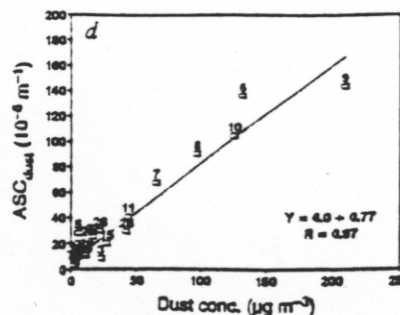
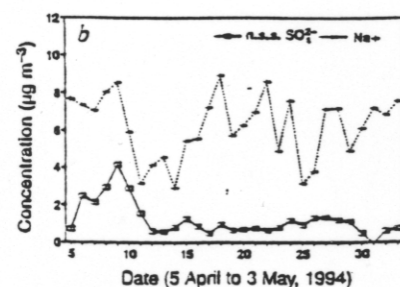
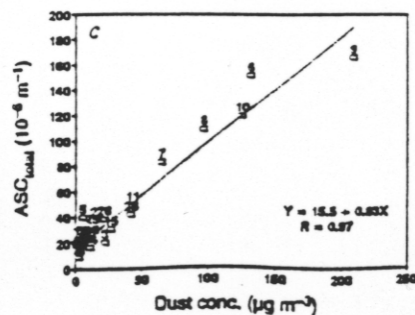
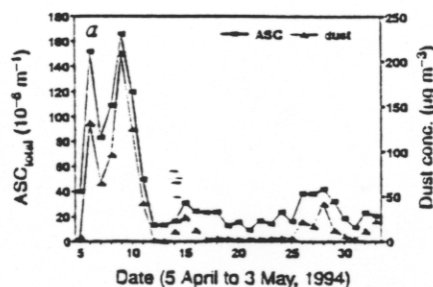


FIG. 1 Time series of ASC_{total} (five-minute averages) measured at Barbados, 5 April to 3 May, 1994 using a Radiance Research Inc. M903 nephelometer at 530 nm.

METHODS. The instrument zero was set daily by flushing with particle-free (filtered) air; it was calibrated by flushing with CO_2 ($ASC = 2.14 \times 10^{-5} m^{-1}$ at $20^\circ C$). Measurements were conducted at the top of a 16-m aluminum tower located on a 30-m-high bluff on the windward coast; the nephelometer sampled directly from the ambient air. To minimize relative humidity (RH) effects on particle growth, the inlet air was heated to ensure that the RH was below 40%; all ASC data in this work apply to these low-RH conditions. A 10- μm -diameter (50% cut-off) impactor was inserted between the heater and the nephelometer to eliminate large particles (principally locally derived sea salt). We periodically alternated nephelometer measurements made with and without the impactor; in most cases the difference in scatter was too small to be detected by the nephelometer, which suggests that the dominant light-scattering aerosols were $<10 \mu m$ diameter. Because light scattering from large particles has a strong forward component and because of the truncated viewing angle of nephelometers, a fraction of the large-particle scatter is not seen by the detector. We estimated the truncation error by Mie scattering calculations using our measured size distributions, and obtained a value of 1.25 which has been incorporated in all ASC values reported here.

April to 3 May 1994. The aerosol scattering coefficient (ASC), an important climate-related property of aerosols², was measured with an integrating nephelometer¹⁵ (results shown in Fig. 1). From 6 to 10 April, dust concentrations (Fig. 2a) were consistently above $65 \mu g m^{-3}$; the value on 9 April, $209 \mu g m^{-3}$, was the highest

FIG. 2 Relationship between aerosol concentrations and the aerosol scattering coefficients (ASC). a, Time series of daily average of ASC_{total} and mineral dust concentration; b, Time series of daily average n.s.s. SO_4^{2-} and sea salt concentration; c, scatter plot of daily average ASC_{total} against mineral dust concentration; d, scatter plot of daily average ASC_{dust} against mineral dust concentration. In c and d, numbers in the figure refer to the sample date. Daily aerosol samples were collected at a flow rate of $0.8 m^3 min^{-1}$ using Whatman-41 filters which have essentially 100% collection efficiency for marine aerosols²⁸. In Miami, filters are extracted with water; Na^+ is determined by flame atomic absorption and Cl^- , NO_3^- and SO_4^{2-} by suppressed ion chromatography²⁹. Non-seasalt (n.s.s.) SO_4^{2-} is calculated as total SO_4^{2-} minus Na^+ times 0.2517, the $SO_4^{2-}:Na^+$ mass ratio in sea water. Mineral dust is determined by ashing the extracted filter at $500^\circ C$ for several hours and weighing the residue. To minimize contamination from local sources, pumps were activated only when winds blew directly from the ocean, a condition that was satisfied essentially 100% of the time during the sampling period.



ever measured in 30 years at this site¹¹ although daily mean values over $100 \mu g m^{-3}$ are not uncommon. The maximum daily average ASC_{total} ($166 \times 10^{-6} m^{-1}$) coincided with the dust maximum on 9 April (Fig. 2a).

Sun photometer measurements of aerosol optical thickness (AOT), a measure of the attenuation by aerosols of direct solar radiation through the atmosphere, yielded high AOT values through the period, seldom dropping below 0.2 (at 550 nm). The highest AOTs (maximum 0.72) occurred during the large dust event of 8–10 April. The high values of AOT are consistent with past observations^{13,14} that the dust layers are very deep, typically exceeding 3 km.

At times, winds from the central Atlantic brought relatively dust-free air to the site and ASC values dropped sharply (Fig. 2a); nonetheless, substantial quantities of dust were always present. The minimum daily average ASC_{total} ($9.0 \times 10^{-6} m^{-1}$) occurred on 21 April when the dust concentration was $3.3 \mu g m^{-3}$. The concentrations of other aerosol species varied considerably although not nearly as much as that for dust (Fig. 2b). During the very dusty period in early April, the dust concentrations were 30–53 times greater than those of n.s.s. SO_4^{2-} and 9–24 times those of Na^+ ; thus, the ASC_{total} during this period should be dominated by dust scatter.

A scatter plot of daily average ASC_{total} against dust concentration is shown in Fig. 2c. Linear regression yields:

$$ASC_{total}(10^{-6} m^{-1}) = (15.5 \pm 2.3) + (0.83 \pm 0.04) \times [dust] \quad (1)$$

where [dust] is the dust concentration in units of $\mu g m^{-3}$; $r = 0.97$. The slope of the equation provides an estimate of the MSE of dust (MSE_{dust}), $0.83 m^2 g^{-1}$. There is considerable debate about the appropriate definition of MSE, and the procedures that must be used to measure it^{2,16,17}; our approach yields an MSE value that is an ensemble average of the effects of the dominant aerosol component and the minor species that are closely associated with it. The substantial positive intercept in equation (1) suggests that there are other aerosol species that contribute to the value of ASC_{total} independent of dust and associated species. We can estimate the light scatter due to dust alone (ASC_{dust}) by subtracting from ASC_{total} the scattering due to other components:

$$ASC_{dust} = ASC_{total} - \sum(MSE_i \times C_i) \quad (2)$$

where MSE_i and C_i represent the MSE and concentration of aerosol species, i , other than dust. Table 1 shows the mean aerosol

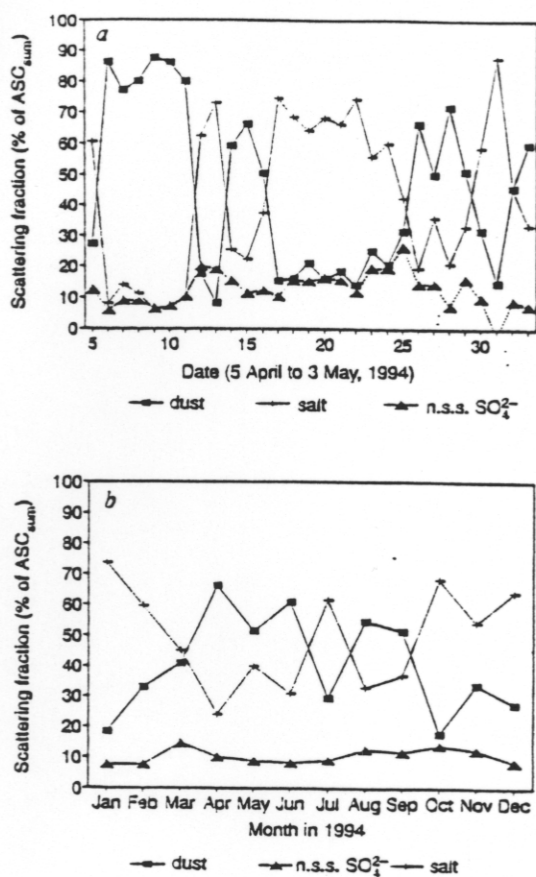


FIG. 3 The relative contribution of mineral dust, sea salt and n.s.s. SO_4^{2-} to the ASC_{sun} at Barbados. a, Daily values for the time period of the experiment, 5 April to 3 May, 1994; b, monthly mean values calculated for the year 1994.

concentrations of the principal species during the period of the experiment, for all of 1994 and for the period 1984–94. On all timescales, the major aerosol components are mineral dust, n.s.s. SO_4^{2-} and sea salt. Although NO_3^- is a substantial aerosol component, its size distribution follows that of sea salt¹⁸; thus it will not have a significant impact on light scatter relative to sea salt. Ammonium is a minor constituent whose size distribution follows that of n.s.s. SO_4^{2-} ; its scattering effects are apportioned accordingly. Other species are present but they should have a minor effect on light scatter; black carbon concentrations were low, and Saharan dust contains only 3.3% organic matter¹⁹. On this basis in equation (2) we need to account principally for scattering due to n.s.s. SO_4^{2-} and sea salt. We estimated the MSE of sea salt (MSE_{SS}) using data collected when dust concentrations were below $5 \mu\text{g m}^{-3}$; for this sample set the mean seasalt concentration was 7.5 times that of dust. Linear regression analyses of ASC against sea salt yielded estimates of MSE_{SS} that fell in the range $0.3\text{--}0.5 \text{ m}^2 \text{ g}^{-1}$.

We could not estimate the MSE of n.s.s. SO_4^{2-} (MSE_{SO_4}) at Barbados with the procedure used for dust and sea salt because

sulphate concentrations were always very low compared to dust and sea salt. There are no published estimates of MSE_{SO_4} for the tropical North Atlantic; as a first approximation we use a value obtained near the Azores², $2.3 \text{ m}^2 \text{ g}^{-1}$, which falls in the low end of the range of previous estimates^{2,16,17} for n.s.s. SO_4^{2-} . A low value for MSE_{SO_4} is consistent with the observation (based on cascade impactor studies during the experiment) that the size distribution of sulphate is shifted to relatively larger sizes because of the presence of sea salt and mineral dust and the consequent deposition of n.s.s. SO_4^{2-} on the surfaces of these larger particles.

Using equation (2), an intermediate value of $\text{MSE}_{\text{SS}} = 0.4 \text{ m}^2 \text{ g}^{-1}$ and $\text{MSE}_{\text{SO}_4} = 2.3 \text{ m}^2 \text{ g}^{-1}$, we calculated ASC_{dust} for the daily samples; a scatter plot of ASC_{dust} against dust concentration is shown in Fig. 2d. The regression equation of the line is:

$$\text{ASC}_{\text{dust}} (10^{-6} \text{ m}^{-1}) = (6.0 \pm 2.2) + (0.77 \pm 0.04) \times [\text{dust}] \quad (3)$$

with [dust] in units $\mu\text{g m}^{-3}$. The slope of the equation yields the MSE_{dust} $0.77 \text{ m}^2 \text{ g}^{-1}$. Because of the extremely high dust concentrations, the slope is relatively insensitive to the values of MSE_{SO_4} and MSE_{SS} ; a matrix of test values with MSE_{SS} ranging from 0.3 to $0.5 \text{ m}^2 \text{ g}^{-1}$ and MSE_{SO_4} , 2–5 $\text{m}^2 \text{ g}^{-1}$, yielded a maximum range of 0.73–0.79 $\text{m}^2 \text{ g}^{-1}$ for MSE_{dust} . The highest test value for MSE_{SO_4} ($5 \text{ m}^2 \text{ g}^{-1}$) is the same as that used for the global mean by Charlson *et al.*².

There may be other aerosol scatterers present during dusty periods but their effects, if any, are masked by the extremely high concentrations of dust and, thus, they will have little impact on the derived MSE for dust. Our results are consistent with those obtained in an extensive study of dust optical properties in the western United States by White *et al.*²⁰, to our knowledge the only other study that directly measures dust scatter using similar techniques.

We can estimate the relative contributions of dust, n.s.s. SO_4^{2-} and sea salt and associated aerosol components to aerosol light scatter at Barbados in the boundary layer (reduced to relative humidity $\leq 40\%$) with the equation:

$$\text{ASC}_{\text{sun}} (10^{-6} \text{ m}^{-1}) = 0.4[\text{sea salt}] + 2.3[\text{n.s.s. SO}_4^{2-}] + 0.77[\text{dust}] \quad (4)$$

where ASC_{sun} represents the sum effect of the scattering of the principal species in square brackets (units, $\mu\text{g m}^{-3}$). On a daily basis for April 1994 (Fig. 3a), dust was the dominant scatterer during 15 of the 29 days of the experiment. During days when dust was about $10 \mu\text{g m}^{-3}$ or less, sea salt was the dominant scatterer, with a comparable contribution from sulfate and mineral dust.

Studies carried out by us over the past 30 years at Barbados show that the chemical and physical properties (including size distribution and mineralogy) of dust are relatively constant during the year except in the winter when dust concentrations are generally low. On this assumption, we estimate the relative scatter for dust, n.s.s. SO_4^{2-} and sea salt for the year 1994 using monthly mean concentrations for these species (Fig. 3b). The contribution from n.s.s. SO_4^{2-} was relatively constant throughout 1994 ($\sim 10\%$ of ASC_{sun}) while that from dust was highly variable, from about 60% during the spring and summer to 20% during the winter. On the basis of the ten-year mean values (Table 1) of aerosol concentrations, the mean scattering contributions to ASC_{sun} from dust, sea salt and n.s.s. SO_4^{2-} are 56%, 33% and 11%, respectively.

Although sea salt is an important light-scattering aerosol, its concentration drops sharply above the marine boundary layer which is typically at an altitude of 1–1.5 km in the western tropical Atlantic; in contrast, continentally derived n.s.s. SO_4^{2-} and dust aerosols in the trade winds are carried in a layer that typically extends to 3–6 km altitude^{13,14,21}. Thus, averaged over the atmospheric column, the net scattering effect of sea salt will be substantially reduced relative to dust and n.s.s. SO_4^{2-} .

TABLE 1 Mean aerosol concentrations at Barbados

Averaging period	dust ($\mu\text{g m}^{-3}$)	n.s.s. SO_4^{2-} ($\mu\text{g m}^{-3}$)	Sea salt ($\mu\text{g m}^{-3}$)	NO_3^- ($\mu\text{g m}^{-3}$)	NH_4^+ ($\mu\text{g m}^{-3}$)
April 1994	30.90	1.19	20.18	0.79	0.13
1994 total	11.16	0.69	21.11	0.62	0.12
1984–94	12.89	0.78	16.50	0.53	0.11

On a global scale, mineral dust is one of the major aerosol components in the atmosphere with emission rates estimated^{7,22,23} to be in the range of about 1,000–3,000 Tg yr⁻¹ while that for n.s.s. SO₄²⁻ aerosol is ~250 Tg yr⁻¹ of which 140 Tg yr⁻¹ is from anthropogenic sources²³. Although it is not presently possible to determine the role of dust in light scatter on a global scale on the basis of dust mass emission estimates because of the large uncertainties regarding the variability in the physical properties of the dust, it is clear that dust has the potential to play an important role in radiative processes and climate²⁴.

Although mineral dust is clearly a natural product, the rates of emission of this dust seem to have greatly increased²⁵ owing to drought compounded by land-use practices²⁵. Measurements of mineral dust made on Barbados since 1965^{10,11} show that dust concentrations in the trade winds are correlated with rainfall deficits in the sub-Saharan region. Indeed, the large dust episode in early April 1994 is clearly traceable to the coast of sub-Saharan Africa in satellite (Meteosat and Goes) imagery. Thus, a substantial fraction of the mobilized dust in the atmosphere might properly be considered to be an anthropogenic pollutant. These various factors suggest that the radiative effects of mineral dust could be important in climate processes, and that the effects of dust could complicate the attempts to assess human effects on climate^{24,27}. □

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