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NON-SEA-SALT SULFATE AND NITRATE IN TRADE WIND AEROSOLS AT BARBADOS:
EVIDENCE FOR LONG-RANGE TRANSPORT

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Abstract. From mid-May 1984 through December 1987, more than 1100 daily high-volume bulk aerosol samples were collected during onshore trade winds at Barbados, West Indies. All of these have been analyzed to determine the concentrations of particulate non-sea-salt (nss) sulfate, nitrate, and Saharan dust; 91 of the samples were also analyzed for methanesulfonate (MSA). The mean concentrations (in $\mu\text{g m}^{-3}$) during the period were nitrate, 0.509 ($s=0.389$); nss sulfate, 0.751 ($s=0.602$); mineral dust, 16.0 ($s=21.1$); and MSA, 0.0207 ($s=0.0093$). The concentrations of both nitrate and nss sulfate are significantly correlated with those of Saharan dust, indicating that substantial fractions of both are transported across the tropical North Atlantic in association with the dust. This transport accounts for about 60% of the mean total concentration of each of the anions at Barbados. Our data, combined with those from previous studies, indicate that these dust-related fractions are probably not derived from the Sahara soil material; they are more likely derived from anthropogenic sources. The nitrate-to-nss sulfate ratio in this dust-related fraction changes markedly from the summer to the winter. During the summer the general meteorology and nitrate-to-nss sulfate mass ratio (0.36) are consistent with Europe being the major source region; during the winter the ratio increases by a factor of 4 to 1.44, which may be consistent with a source region over southern West Africa. The ratio of the mean MSA concentration to that of the nss sulfate that is not related to dust transport is 0.066, a value similar to the ratio of MSA to total nss sulfate at relatively pristine stations in the Pacific: Fanning Island and American Samoa. The similarity suggests that this fraction of the nss sulfate at Barbados may be derived predominantly from oxidation of reduced sulfur compounds emitted from the ocean. The mean concentration of nitrate that is not related to dust transport is $0.22 \mu\text{g m}^{-3}$, about double the total mean concentration over the tropical South Pacific ($0.11 \mu\text{g m}^{-3}$) and 40% higher than that over the equatorial Pacific ($0.16 \mu\text{g m}^{-3}$). The major source (or sources) of this nondust-related nitrate is still very uncertain.

Introduction

Research during the last two decades has shown that the long-range transport of materials

from the continents to the oceans can have a major impact on the marine atmosphere. Most studies have focused on mineral dust, since it can be unambiguously ascribed to continental sources. Such studies have often been the cornerstone for our understanding of the mechanisms of transport and for an appreciation of the great distances that are sometimes involved. However, the transport of other continentally derived constituents, such as nitrogen and sulfur compounds, could have a more dramatic effect on tropospheric chemistry and physics. Gaseous nitrogen oxides are major determinants of the oxidation state of the remote troposphere, being directly involved in establishing the level of O_3 and, both directly and indirectly, the level of the OH radical. For the most part, reduced sulfur compounds act as sinks for these oxidants although they do not play a significant role as such. The major oxidation products, HNO_3 and H_2SO_4 , are substantial contributors to the acidity of precipitation and particles in remote, as well as polluted, regions. By reducing the pH of these components, they can cause the volatilization of weaker acids, inorganic as well as organic, and the partial dissolution of minerals, including carbonates and aluminosilicate dust. Nitrate and sulfate readily form aerosols, which affect climate and weather through their interaction with solar and terrestrial radiation and their action as condensation nuclei.

The transport of massive quantities of dust from the Sahara across the tropical North Atlantic (TNA) has been studied extensively during the past 25 years (For reviews of various aspects of Saharan dust phenomena, see Prospero [1981]; Coude-Gaussen [1984]; Middleton et al. [1986]; Pye [1987]; Prospero and Carlson [1981].) Data have been obtained from island stations, from ships, and aboard aircraft across the entire expanse of the region between the coast of Africa and the Caribbean. Much less attention has been focused on the concurrent transport of other materials. Recently, Talbot et al. [1986] have made measurements of numerous constituents in samples collected aboard aircraft near Barbados ($13^{\circ}10'N$, $59^{\circ}25'W$), West Indies (Figure 1). On the basis of this data, the authors concluded that the dust-related nitrate and non-sea-salt (nss) sulfate could account for all of both of these constituents in the marine boundary layer at this location. The authors calculated the fluxes of these species to the underlying ocean on the basis of the assumption that the nitrate and sulfate were derived from the Saharan soil material and hence that the ratios of these constituents to that of dust are essentially constant. In contrast, Savoie [1984] suggested that the concurrence of high concentrations of nitrate, nss sulfate, and dust can be attributed

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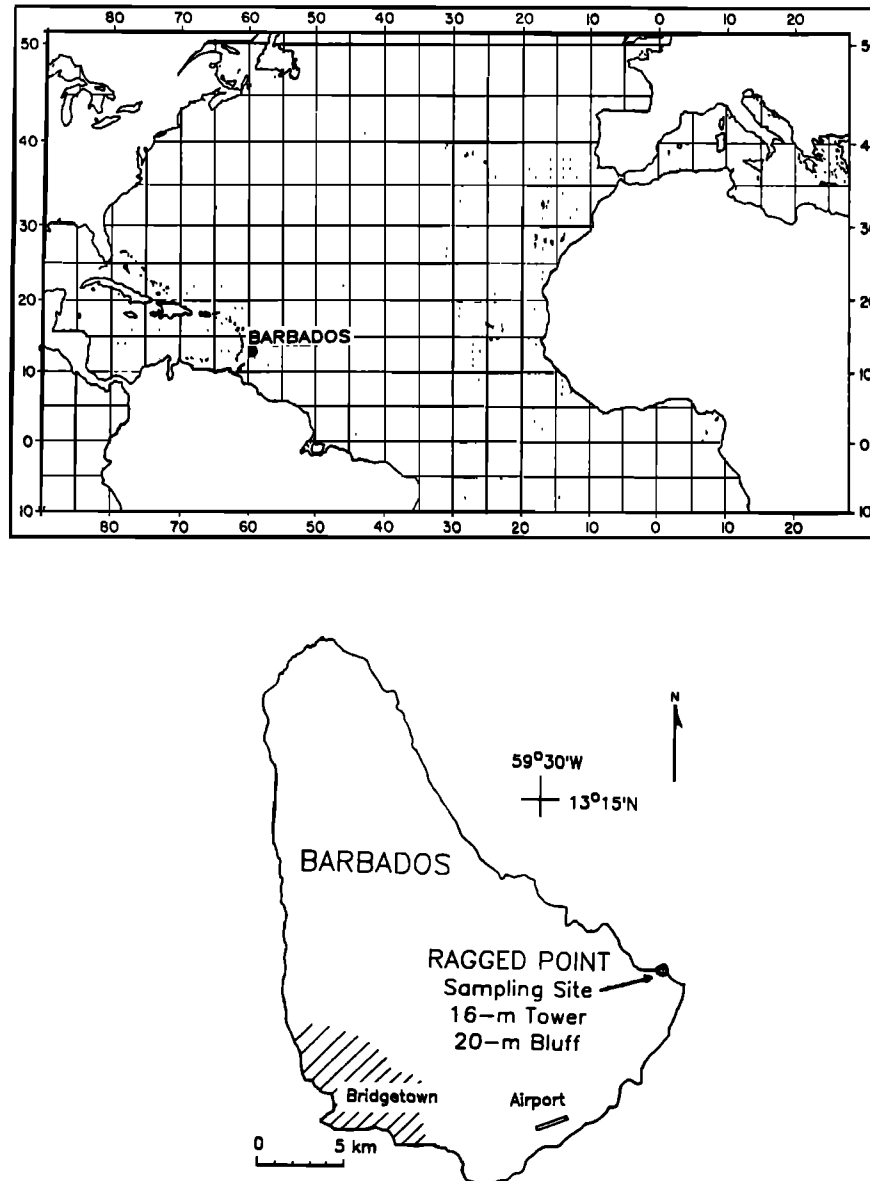


Fig. 1. Map of the North Atlantic, indicating the location of Barbados, and map of Barbados, showing the location of the sampling site.

to the fact that the synoptic events associated with dust storm generation in the Sahara are also conducive to the rapid transport of pollutant materials from Europe and North Africa into the Sahara. The resulting mixture of dust and pollutants is then carried out over the Atlantic. If this latter scenario is correct, then the fluxes over the western TNA are not directly tied to that of the dust, but rather to sources that may be thousands of kilometers more distant.

The resolution of the source issue is important from two standpoints. Prospero and Nees [1986] have reported that the mean dust concentration at Barbados increased by more than a factor of 4 from the late 1960's, prior to the onset of the drought in Africa, to the peak dust concentration in 1984. If the anions are derived from the soil material, then the flux of nitrate

and sulfate to the TNA should have increased by the same factor. On the other hand, if the two anions and/or their precursors are derived from other sources and are simply transported along with the dust, then their concentrations would not be directly tied to that of the dust, and consequently, they could have varied in a completely different manner. Perhaps, more importantly, the nitrate and sulfate derived from soils are principally in the form of neutral salts. In contrast, the nitrogen and sulfur from pollutant sources are oxidized to acids which may or may not be subsequently neutralized by reactions with basic species in the atmosphere.

In this report we present and discuss the concentrations of nss sulfate, nitrate, and mineral dust measured in 1162 daily high-volume bulk aerosol samples, which were collected at

Barbados from mid-May 1984 through December 1987. About one third (91) of the 1985 samples were also analyzed for methanesulfonate (MSA), which is used here as an indicator of the marine biogenic source of nss sulfate. Our primary objectives were to examine more closely the interrelationships which exist among the three major components and to use these results, along with previously published information, to assess more critically the potential sources of the nitrate and nss sulfate.

Sampling and Analysis

The system at Barbados is located on a 20-m bluff at Ragged Point, near the easternmost point of the island. Since May 1984 the samples have been collected on top of a 16-m fold-down tower. The sampler is equipped with wind sensors, which activate the pumps only when the wind is coming off the ocean at speeds greater than 1 m s^{-1} . The trade winds blow very steadily at this site all year long; as a consequence, in-sector times average about 95%.

Daily high-volume bulk aerosol samples were collected by drawing air through $20 \times 25 \text{ cm}$ Whatman 41 filters at a flow rate of about $1.1 \text{ m}^3 \text{ min}^{-1}$, yielding an average sampled volume of about 1500 m^3 . The results from studies using dual in-line Whatman 41 filters with a 1-cm separation distance indicate that these filters have total mass collection efficiencies greater than 90% for nss sulfate and MSA, 95% for nitrate and sea salt [Savoie, 1984], and 99% for mineral aerosol (M. Uematsu, University of Rhode Island, personal communication, 1988). These results are consistent with those obtained by Watts et al. [1987] and Lowenthal and Rahn [1987] for high volume bulk aerosol samples. Although similar efficiencies were reported by Kitto et al. [1988], they presented no details of their sampling conditions or environment.

Quarter sections of the filters were extracted in 20 mL of Milli-Q water. The concentrations of sulfate, nitrate, and MSA in the extraction solutions were determined to within $\pm 5\%$ by ion chromatography and that of sodium was determined to within $\pm 2\%$ by atomic absorption spectroscopy [Saltzman et al., 1983, 1986a; Savoie et al., 1987]. The nss sulfate concentrations were calculated by subtracting 0.2517 times the sodium concentration from the total sulfate concentration. For the average sodium concentration of $4.14 \mu\text{g m}^{-3}$ in the current sample set and an average total sulfate concentration of 1.8, the average standard error in the nss sulfate concentration is about $\pm 0.1 \mu\text{g m}^{-3}$. On the average, the nss sulfate accounts for about 42% of the total sulfate at Barbados.

Although magnesium, potassium, and calcium have not been analyzed in the current sample set, Savoie and Prospero [1980] have previously found that the dust contributes a negligible amount of water-soluble sodium in the near-surface marine boundary layer. The water-soluble magnesium in those samples was also derived almost exclusively from sea salt. In contrast, significant and often substantial fractions of the potassium and, particularly, the calcium were derived from the soil dust.

The insoluble material including the filter matrix was ashed at 500°C to destroy any organic matter. The residual ash, less that of the average blank, is ascribed to mineral aerosol [Savoie et al., 1987]. Previous analyses of the aluminum content of similarly treated Saharan aerosol samples yielded an average Al content of 8%, a value that is consistent with that of average soil material [Prospero et al., 1987]. The standard error in the mineral aerosol concentration is $\pm 10\%$ for concentrations greater than about $1 \mu\text{g m}^{-3}$. Below $1 \mu\text{g m}^{-3}$, the standard error is essentially constant at $\pm 0.1 \mu\text{g m}^{-3}$.

The nitrate values reported here are for total inorganic nitrate, i.e., particulate nitrate plus gaseous HNO_3 . Whatman 41 and other cellulose filters have been shown to be greater than 95% efficient for collecting HNO_3 when the filters are impregnated with NaCl [Appel et al., 1980, 1981; Forrest et al., 1980; Okita et al., 1976]. Teflon-nylon filter packs have also been shown to quantitatively retain inorganic nitrate [cf. Appel et al., 1980, 1981; Spicer et al., 1982]. Results from concurrent sampling with unimpregnated Whatman 41 filters, NaCl impregnated Whatman 41 filters, and Teflon-nylon filter packs indicate that in the near-surface marine boundary layer, the unimpregnated filters are as efficient as the other two systems for collecting total inorganic nitrate [Savoie, 1984]. Concentrations measured with the unimpregnated filters were 1.05 (standard geometric deviation (SGD)=1.12, $n=10$) times those with the filter pack and 1.02 (SGD = 1.11, $n=6$) times those with the NaCl Whatman 41. Neither of these factors differs significantly from 1. The high efficiency for HNO_3 in this environment is probably, at least partially, a consequence of the substantial concentrations of sea salt, which rapidly builds up on the filter and, in effect, impregnates the filter with NaCl and other salts. While Savoie and Prospero [1982] have also concluded that 90% or more of the nitrate in the near-surface marine boundary layer exists in the particulate phase, that conclusion remains to be verified.

Despite their high efficiency for collecting HNO_3 , the NaCl impregnated cellulose filters are very inefficient for collecting SO_2 . Forrest et al. [1980] reported that SO_2 losses on such filters were negligible. Coutant [1977] had previously found that unimpregnated cellulose acetate filters also absorbed negligible amounts of SO_2 . Consequently, there is no reason to believe that our samples are subject to significant artifact sulfate formation by SO_2 retention and its subsequent oxidation.

Results and Discussion

Mean Concentrations

The mean concentrations found at Barbados during the 1984 through 1987 period are summarized in Table 1. Results from the analyses of 19 years of data show that the annual mean dust concentration at Barbados can vary considerably from one year to another [Prospero

TABLE 1. Mean Concentrations of Various Constituents at Barbados From May 1984 Through December 1987 and Those at Several Remote Locations in the Pacific and Indian Oceans

Data Set	N	Nitrate	Nss Sulfate	Dust	MSA
<u>Barbados</u>					
1984-1987	1162	0.509 (0.389)	0.751 (0.602)	16.0 (21.1)	0.0207 (0.0093,91)
<u>Remote Marine Locations</u>					
Fanning Island	175	0.165 (0.084)	0.673 (0.276)		0.044 (0.012 ,29)
Nauru	189	0.163 (0.089)			
Funafuti	161	0.107 (0.070)			
American Samoa	195	0.110 (0.052)	0.347 (0.141)		0.026 (0.012 ,22)
Rarotonga	192	0.117 (0.068)			
Indian Ocean, southern hemisphere air	23	0.159 (0.051)	0.50 (0.17)		0.020 (0.013 , 9)

N, the number of samples, applies to each of the data sets except MSA. For MSA, the number is given as the last value in the parentheses for each data set. Standard deviations of the means are given in parentheses. Concentrations are in $\mu\text{g m}^{-3}$.

and Nees, 1977, 1986]. However, there are very distinct and significant long-term trends that are related to the cycle of rainfall in North Africa, especially to the long-term drought that began in the late 1960's. The drought was especially severe in the early 1980's, and dust concentrations in Barbados reached an all-time maximum of $18.7 \mu\text{g m}^{-3}$ in 1983 and then declined to $16.4 \mu\text{g m}^{-3}$ in 1984. The overall mean for the 1984 through 1987 period is $16.0 \mu\text{g m}^{-3}$, which is 3-4 times greater than those in the mid-1960's, before the drought.

The mean concentrations of nitrate ($0.509 \mu\text{g m}^{-3}$) and nss sulfate ($0.751 \mu\text{g m}^{-3}$) at Barbados are considerably higher than those for remote marine regions. The mean nitrate concentration at Barbados is more than a factor of 4 higher than that at each of three remote stations in the tropical South Pacific, $0.11 \mu\text{g m}^{-3}$ (American Samoa, Funafuti, and Rarotonga), and more than a factor of 2 higher than those from the equatorial Pacific, $0.16 \mu\text{g m}^{-3}$ [Prospero et al., 1985; Savoie et al., 1989a,b] and from southern hemisphere air over the equatorial Indian Ocean [Savoie et al., 1987]. If the nitrate concentrations over those Pacific and Indian Ocean areas are representative of the mean "background" in the tropical marine atmospheric boundary layer in the Atlantic, then the "background" nitrate at Barbados would account for only about 20-30% of the overall mean.

The nss sulfate concentration at Barbados is nearly a factor of 2 higher than that at American Samoa, but only about 10% higher than that at Fanning Island. Over some remote ocean regions both MSA and nss sulfate are believed to be derived primarily from the emission of biogenically produced dimethylsulfide (DMS) from seawater and its subsequent oxidation in the troposphere. The strength of this source can vary both seasonally and geographically [Saltzman et al., 1986a]. At the tropical Pacific

stations, American Samoa and Fanning Island, MSA and nss sulfate exhibit a very consistent relationship, with MSA:nss sulfate ratios of the order of 0.06 to 0.07 [Saltzman et al., 1985]. With regard to continental inputs, American Samoa is among the cleanest sites ever studied; trace metal concentrations there are as low as, and often lower than, those at the south pole [Arimoto et al., 1987; Tuncel et al., 1989]; nitrate concentrations are also among the lowest measured in the marine boundary layer [Savoie et al., 1989b]. In areas strongly affected by the input of sulfate from continental sources, the MSA:nss sulfate ratio is considerably lower than those at the remote sites [Saltzman et al., 1986a,b; Andreae et al., 1988a].

At Barbados the ratio of the means, 0.028, is lower by a factor of about 2 indicating a substantial impact from continentally derived sulfate. The annual mean temperature at Barbados is essentially identical to those at American Samoa and Fanning, 26.4° - 26.5°C [Environmental Data Service, 1968; National Climatic Center, 1982]. Moreover, the monthly mean temperature at Barbados varies over the narrow range from 25.3° to 27.3°C (similar to that at American Samoa). Hence, differences in the MSA:nss sulfate ratios between Barbados and the tropical Pacific stations can not be explained by variations in the MSA yield from DMS oxidation as a function of temperature [Hynes et al., 1986].

The mean MSA concentration at Barbados, $0.0207 \mu\text{g m}^{-3}$, is comparable to that found at several tropical and mid-latitude sites over the Pacific [Ayers et al., 1986; Saltzman et al., 1986a] and Indian Oceans [Saltzman et al., 1983]. This similarity suggests that there is probably nothing particularly unusual about the oceanic productivity in the TNA region. In contrast, the mean at Fanning Island in the equatorial Pacific is about twice as high, $0.044 \mu\text{g m}^{-3}$ [Saltzman et al., 1986a]. The higher concentration in that region is attributed to the increased primary

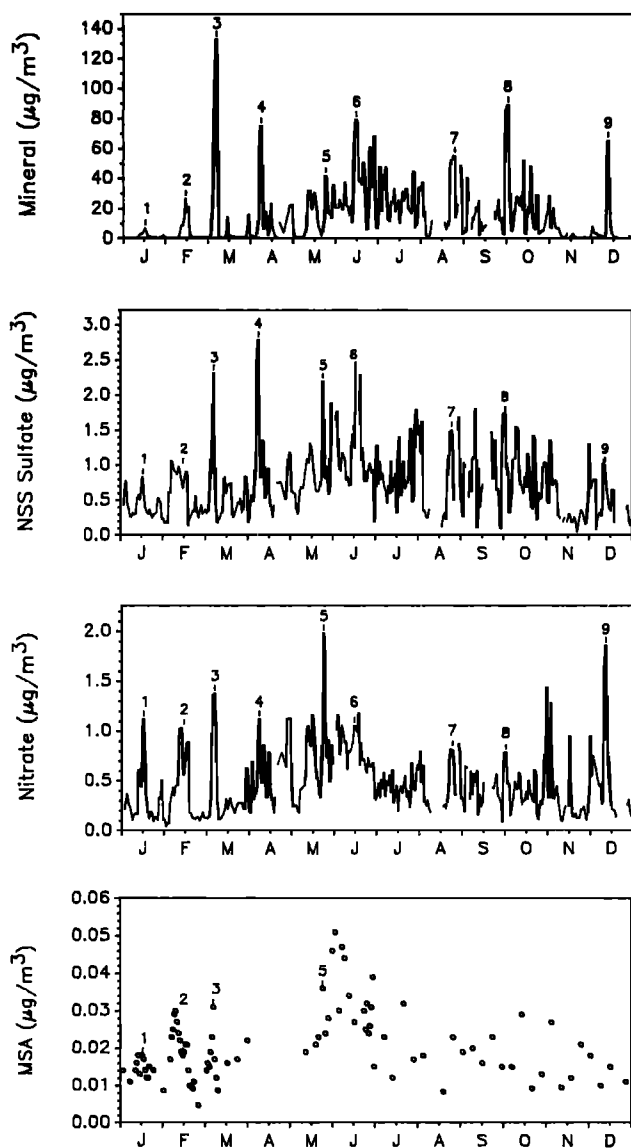


Fig. 2. Daily averaged concentrations of mineral aerosol, nss sulfate, nitrate and methanesulfonate (MSA) in trade wind air at Barbados during 1985. For comparison purposes, some prominent dust peaks are marked, along with the corresponding peaks in the other plots. Note that peaks in concentrations of nss sulfate and nitrate are often but not always coincident with those of Saharan dust.

productivity associated with the upwelling of nutrient-rich water.

Seasonal Variations in Constituent Interrelationships

The results obtained by Talbot et al. [1986] and Savoie [1984] indicate that the high concentrations of nitrate and nss sulfate at Barbados are associated with the transport of dust from Africa. However, the latter study

further indicates that there is no consistent relationship between dust concentration and the concentrations of the two anions; this is true not only at Barbados, but also on several ship cruises and at a station above the trade wind inversion at Tenerife, Canary Islands. The lack of a consistent relationship is also evident in the current study, as the data from 1985 illustrate (Figure 2); Figure 2 also indicates the large day-to-day variations in the concentrations of each of the species. The samples numbered in Figure 2 indicate nine prominent peaks in the dust concentration. Note that relatively high concentrations of nitrate and nss sulfate occurred during these same time periods.

However, a closer examination of the time series plots in Figure 2 reveals that the apparent relationship between the dust and the two major acid species is rather tenuous. As an example, note peak number 2. While high concentrations of the other species are evident during the dust peak, the concentrations were equally high or higher during the preceding week. Comparing peaks 1 and 2, the peak nitrate and nss sulfate concentrations are similar during the two periods, even though the dust concentration during peak 2 is a factor of 3 higher. Similar comparisons can be made with peaks 5 and 6, among others. Note that for peak 6 the mineral dust is the highest concentration measured during June; for nitrate and nss sulfate, peak 6 occurs several days prior to their respective highest concentrations.

In an attempt to clarify the interrelationships between the three major constituents (nitrate, nss sulfate, and mineral dust), scatterplots were generated for each of the monthly data sets for the 3.5-year period. In the process it quickly became obvious that the relationship of nitrate to the other two constituents exhibited a very strong seasonal dependence. There was one relationship which dominated during the summer months from July through October and another which dominated during the winter months from December through February. During the remaining months the relationship varied between the summer and winter extremes, with most samples plotting somewhere in the intermediate region. In contrast, the relationship between nss sulfate and mineral dust was essentially the same for all months of every year.

The regression equations used in all of the succeeding discussions were calculated using the maximum likelihood estimation of a structural relationship [Kendall and Stuart, 1961]. In this method, errors in both the x and y components are taken into account. The minimization is in the sum of the squares of the perpendicular distances from the data points to the regression line after multiplying one of the components by a constant factor, so that the absolute errors in the two components are equal, thus making the error surfaces surrounding the data points circular rather than ellipsoidal.

The scatterplots of nss sulfate versus mineral dust are presented in Figure 3. The regression equations for the summer (July through October) and winter (December through February) did not differ significantly at the 95% confidence level. Hence we present the data from

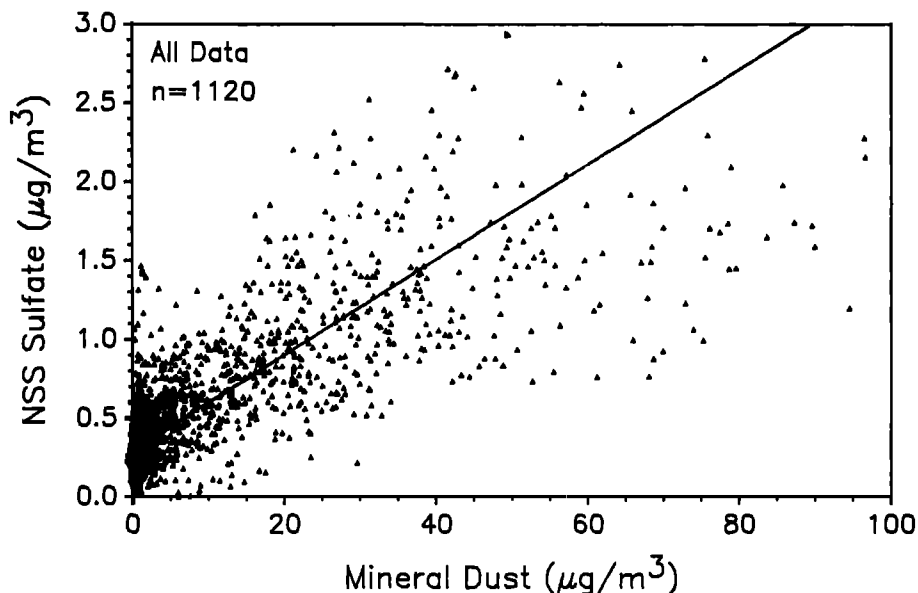


Fig. 3. Scatterplot of the concentrations of non-sea-salt sulfate versus those of mineral dust for all of the samples collected at Barbados from May 1984 through December 1987. The line indicates the regression equation: $nss\ sulfate = 0.0302 (\pm 0.0024) * mineral + 0.30 (\pm 0.06)$; $r=0.75$. See text for explanation of the regression equation.

all of the samples collected during the entire sampling period:

$$\text{summer: } nss\ sulfate = 0.0347 * mineral + 0.25 \\ (\pm 0.0047) \quad (\pm 0.13)$$

$$\text{winter: } nss\ sulfate = 0.0308 * mineral + 0.31 \\ (\pm 0.0062) \quad (\pm 0.07)$$

$$\text{all: } nss\ sulfate = 0.0302 * mineral + 0.30 \\ (\pm 0.0024) \quad (\pm 0.06)$$

(where the plus or minus indicates the 95% confidence interval for a given parameter).

On the basis of these results, the best estimate for the sulfate flux which is associated with Saharan dust transport is obtained by multiplying the dust flux by 0.030. Note that this value is about 2.3 times higher than that used by Talbot et al. [1986] for their flux estimates even though they suggested that the dust could account for all of the nss sulfate. Our estimated slope would certainly have been larger had a zero intercept been assumed. In all cases there is a very significant intercept of about $0.3 \mu g\ m^{-3}$, which indicates that there is a significant source of nss sulfate that is not associated with dust transport from Africa. This feature will be discussed further in the next section.

For nitrate versus mineral dust, the scatterplots for both the winter and summer seasons are presented (Figure 4). The data are presented on the same scales to facilitate visual comparisons. Despite the apparent considerable scatter in the winter data, the correlation coefficient and regression parameters are highly significant (i.e., >95% confidence level). As

mentioned earlier, the regression slopes for the two time periods differ markedly:

$$\text{summer: } nitrate = 0.0125 * mineral + 0.202 \\ (\pm 0.0016) \quad (\pm 0.046)$$

$$\text{winter: } nitrate = 0.0443 * mineral + 0.246 \\ (\pm 0.0083) \quad (\pm 0.096)$$

Even the summer ratio is 2.4 times higher than that used by Talbot et al. and again they had assumed that all of the nitrate was associated with the dust. The difference between the two seasons is clearly significant; the winter slope is more than 3.5 times higher than that during the summer. Moreover, when the two plots are overlain, there is extremely little overlap of the scatterplots for mineral dust concentrations greater than $5 \mu g\ m^{-3}$. As with nss sulfate, the significant intercept of about $0.2-0.25 \mu g\ m^{-3}$ indicates an additional significant source for nitrate and/or its precursors.

Because of the much stronger correlation between nitrate and nss sulfate, the winter summer difference for the nitrate versus nss sulfate relationship is even more dramatic (Figure 5):

$$\text{summer: } nitrate = 0.361 * nss\ sulfate + 0.110 \\ (\pm 0.043) \quad (\pm 0.051)$$

$$\text{winter: } nitrate = 1.44 * nss\ sulfate - 0.22 \\ (\pm 0.19) \quad (\pm 0.11)$$

The winter slope in this case is nearly 4 times higher than that during the summer.

It must be noted at this point that not all of the winter samples exhibit the typical winter relationship. There are rare brief periods

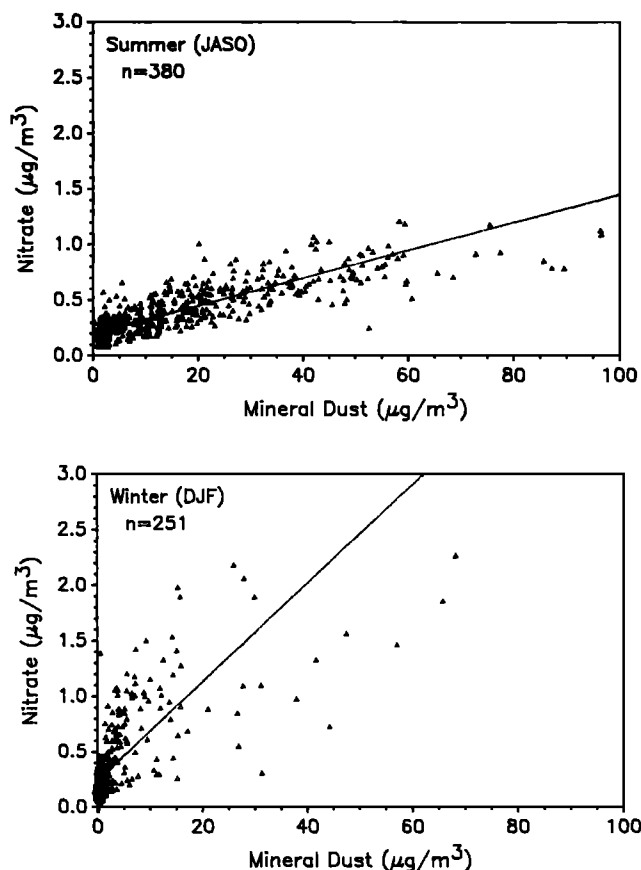


Fig. 4. Scatterplots of the concentrations of nitrate versus those of mineral dust for samples collected at Barbados from May 1984 through December 1987. (a) Summer months, July through September, for which the regression equation is $\text{nitrate} = 0.0125 (\pm 0.0016) * \text{mineral} + 0.202 (\pm 0.046)$, $r=0.80$, (b) Winter months, December through February, for which the regression equation is $\text{nitrate} = 0.0443 (\pm 0.0083) * \text{mineral} + 0.246 (\pm 0.096)$, $r=0.69$. See text for explanation of the regression equations.

(sometimes on the order of a week long) during which the relationship reverts to that typical of the summer months. One such period is noted in Figure 6, in which nitrate is plotted versus nss sulfate for all of the January data from 1985 through 1987. All of the points for the period January 24 through February 1, 1986, fall in the region which is typical of the summer, whereas the vast majority of the data are clearly of the winter variety. Over the course of the 3-month winter period, less than 10% of the data exhibit a summerlike relationship and all tend to occur in several day events rather than in isolated samples. The major cause of these events is still not known but several possibilities are currently being investigated.

Sources of Nitrate and nss Sulfate

Nondust-related fraction. As far as the sources of the nitrate and nss sulfate are

concerned, the nondust-related (NDR) fractions are perhaps most easily assessed. The mean concentrations of the NDR fractions have already been estimated by one method, i.e., the intercepts of the regression equations of the constituent concentrations versus mineral dust. From those results the estimated mean NDR concentration of nss sulfate is $0.304 (\pm 0.058) \mu\text{g m}^{-3}$ and that of nitrate is $0.202 (\pm 0.046)$ to $0.246 (\pm 0.096) \mu\text{g m}^{-3}$, based on the summer and winter regressions, respectively.

An alternative approach is to assume that the total concentrations of nitrate and nss sulfate are each equal to a mean NDR concentration plus a continental component. The continental component is, in turn, assumed to have a constant nitrate/nss sulfate ratio during the summer and a different but also constant ratio during the winter. These ratios are defined by the regression slopes of nitrate versus nss sulfate which have already been presented. Relationships of either to the dust concentration are

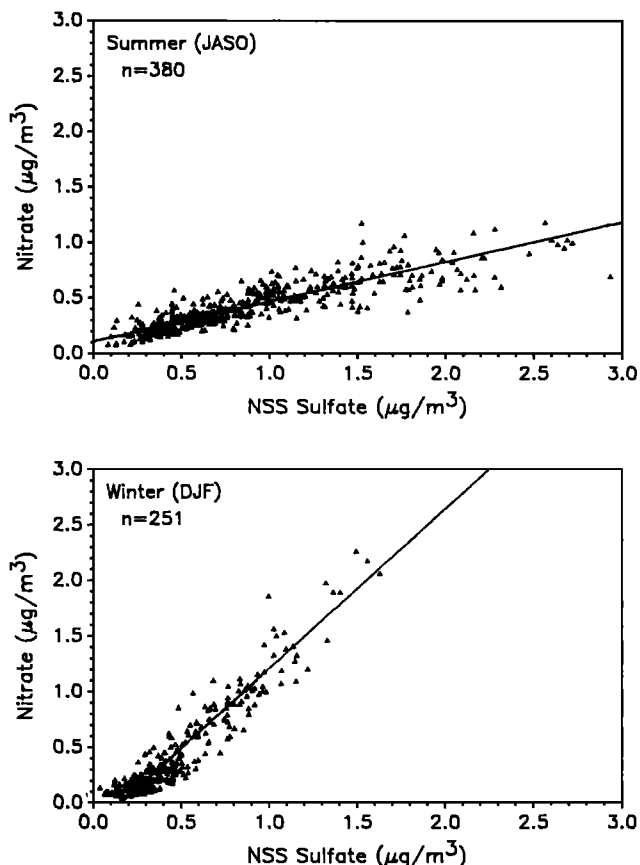


Fig. 5. Scatterplots of the concentrations of nitrate versus those of non-sea-salt sulfate for samples collected at Barbados from May 1984 through December 1987. (a) Summer months, July through September, for which the regression equation is $\text{nitrate} = 0.361 (\pm 0.043) * \text{nss sulfate} + 0.110 (\pm 0.051)$, $r=0.86$, (b) Winter months, December through February, for which the regression equation is $\text{nitrate} = 1.44 (\pm 0.19) * \text{nss sulfate} - 0.22 (\pm 0.11)$, $r=0.94$. See text for explanation of the regression equations.

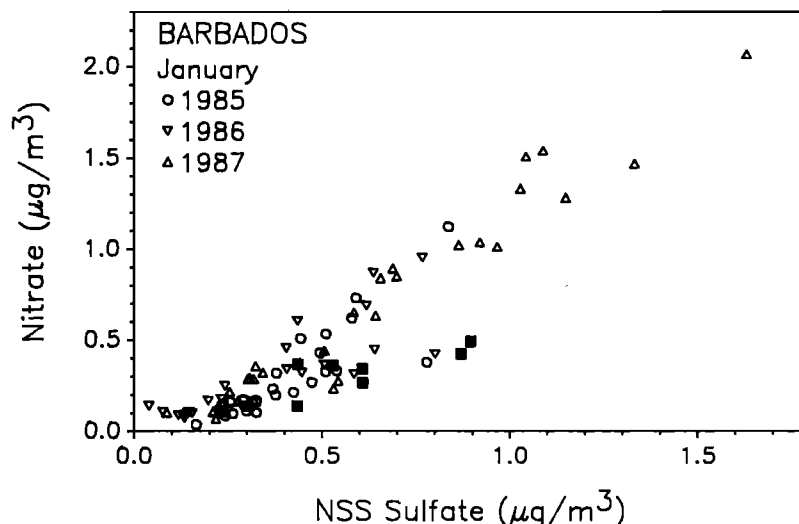


Fig. 6. Scatterplot of the concentrations of nitrate versus those of non-sea-salt sulfate for samples collected at Barbados during January of 1985, 1986, and 1987. The data indicated by solid squares are those samples that were collected from January 24 to February 1, 1986, 7 consecutive days when the nitrate-to-nss sulfate ratio was similar to that normally observed during the summer rather than the winter months.

irrelevant under these assumptions. Since the two constituents are more strongly related to one another (based on the respective correlation coefficients) than they are to the mineral dust, this is perhaps a better approach. The pertinent equation then becomes:

$$\text{NO}_3^-(\text{total}) - \text{NO}_3^-(\text{NDR}) = A * [\text{nss SO}_4^{2-}(\text{total}) - \text{nss SO}_4^{2-}(\text{NDR})]$$

Application of the summer (S) and winter (W) regression parameters then yields the following equations:

$$\text{S: NO}_3^-(\text{NDR}) = 0.110 + 0.361 * \text{nss SO}_4^{2-}(\text{NDR})$$

$$\text{W: NO}_3^-(\text{NDR}) = -0.225 + 1.435 * \text{nss SO}_4^{2-}(\text{NDR})$$

Solving these equations simultaneously yields estimates of $0.223 \mu\text{g m}^{-3}$ for $\text{NO}_3^-(\text{NDR})$ and $0.312 \mu\text{g m}^{-3}$ for $\text{nss SO}_4^{2-}(\text{NDR})$, both of which are essentially identical to those obtained by the first method.

The average NDR nss sulfate accounts for about 40% of the total mean nss sulfate ($0.767 \mu\text{g m}^{-3}$) measured at Barbados during the 3.5-year study period. The primary source of the NDR nss sulfate may well be biogenic. The mean MSA concentration in the 91 samples which have been analyzed thus far is $0.0207 \mu\text{g m}^{-3}$. Hence the ratio of the MSA mean to that of the NDR nss sulfate is about 0.066 to 0.068. These values are very similar to the ratio of 0.065 obtained at the remote Pacific stations of Fanning Island and American Samoa, where the transport of sulfate from the continents is believed to be unimportant [Saltzman et al., 1985].

On the basis of the mean value of $0.22 \mu\text{g m}^{-3}$, the NDR nitrate accounts for about 43% of the total mean nitrate measured at the site from

1984 through 1987. The primary sources for the NDR nitrate are still not known. The mean concentration at Barbados is twice as high as the mean concentration of $0.11 \mu\text{g m}^{-3}$, which occurs over the tropical South Pacific at Rarotonga, American Samoa, and Funafuti and about 40% higher than the mean of $0.16 \mu\text{g m}^{-3}$ over the equatorial Pacific [Savoie et al., 1989a,b]. However, the major source (or sources) of the nitrate even at the relatively pristine Pacific locations is still very uncertain. A major source may be lightning, but the oxidation of organic nitrogen compounds, perhaps proteinaceous material [Mopper and Zika, 1987], may also be important. On the basis of the poor correlation with ^7Be at American Samoa, Savoie et al. [1989b] concluded that stratospheric input was not a major source; however, they conceded that it could be significant. Without knowing the real source of this marine "background" nitrate, it is impossible to even guess how this "background" concentration should vary either temporally or geographically, and hence whether or not the NDR nitrate is a reasonable marine "background" value for the tropical North Atlantic.

The transport of pollutants from North America could potentially contribute to the NDR fractions of both nss sulfate and nitrate. An examination of daily isentropic back trajectories from Barbados (J. Merrill et al., University of Rhode Island, unpublished data, 1988) revealed that many of the air parcels arriving at the site in the marine boundary layer had been over the northern United States about a week before. It is still very unclear what, if any, impact this source might have on the NDR concentrations. On the basis of the trajectories, the air parcels are often at quite high altitude, of the order of 6 km or so, as they cross the continent; however, that by itself does not preclude the incorporation of some pollutants, unless the atmosphere is stably stratified.

Dust-related nitrate and nss sulfate. The origin of the dust-related nitrate and nss sulfate is another problem that has not been completely resolved; however, based on the results mentioned earlier, the dust-related fractions account for about 60% of both the mean total nitrate and nss sulfate at Barbados during the study period. The term "dust-related" is used here to indicate that the nitrate and nss sulfate appear to be transported from Africa along with the Saharan dust. This cotransport is indicated both by the concurrent peaks in the concentrations of all three constituents and the significant correlations between the anions and mineral dust. However, there are several lines of evidence which suggest that the anions are not derived from the soil material itself. First, the additional sulfate present during Saharan dust transport exists primarily in submicron particles [Savoie and Prospero, 1982; Savoie, 1984]. There is no gypsum or other measurable sulfate mineral in unaltered Saharan dust [Glaccum and Prospero, 1980]. X ray analyses of both surface station and aircraft samples indicate that gypsum only occurs in the near-surface samples. In those samples the sea-salt concentrations are sufficiently high to permit solution reactions to occur on the filter during sampling. Scanning electron microscopic studies of the samples clearly reveal that the gypsum occurs as freshly grown "flowers" extending from calcite crystals and not as the aged gypsum crystals one would expect from desert soils [Glaccum, 1978]. Second, even though nitrate and nss sulfate correlate significantly with the dust load, the relationships vary substantially, even within a given outbreak [Savoie, 1984]. In contrast, the mineralogical studies [Glaccum, 1978; Glaccum and Prospero, 1980; Prospero et al., 1981] of Saharan dust indicate that the water-insoluble fraction of the dust has a monotonously consistent composition, both within specific dust outbreaks and from one outbreak to the next.

The long-range transport of pollutants with Saharan dust is favored by a number of factors. Because of the strong vertical convection and lack of precipitation over North Africa, aerosol deposition over the arid desert region would be greatly reduced. Gas-to-particle conversion processes and other reactions may also be limited because of the extremely low relative humidities and the resultant dryness of even hygroscopic particles. Consequently, many gaseous and particulate species are likely to have much longer lifetimes over the desert and, subsequently, over the ocean within the Saharan air layer (SAL), which is largely confined between two inversions at 1.5 and 6 km altitude. Within the marine boundary layer over the ocean, the region of the dust outbreaks is characterized by severely suppressed cloud conditions; consequently, precipitation scavenging will also be greatly reduced, except within the immediate area of the easterly wave disturbances.

If one accepts the premise that the continental source that supplies the major fraction of nitrate and nss sulfate to Barbados is not the Saharan soil material, then there are two major sources which remain as likely candidates: Europe and/or Africa. It is difficult to understand how the input of material

from North America could be correlated with concentrations of Saharan dust, unless the strength of the North American source for some, yet unknown, reason coincidentally varies with the dust transport from Africa. There was certainly no evidence of such covariance from the isentropic trajectories to the marine boundary layer at Barbados.

During the summer the most likely source of the dust-related nitrate and nss sulfate is pollution from Europe and the Mediterranean coastal areas of North Africa. There have been numerous studies of the meteorology associated with dust outbreaks during this time of year [Carlson and Prospero, 1972; Prospero and Carlson, 1981; Karyampudi, 1979, 1986; Westphal et al., 1987]. The genesis of Saharan dust storms normally occurs every 3-5 days, usually in association with easterly waves. Under these conditions, the compression of the isobars over North Africa [Estoque et al., 1986] induce strong northerly winds which are conducive both to the generation of dust storms [Helgren and Prospero, 1987] and to the rapid transport of pollutants from the north into the Sahara and thence into the SAL. Studies have shown that the highest concentrations of numerous pollutants over the Mediterranean are a consequence of transport from Europe [cf. Dulac et al., 1987]. Because of this transport the mean concentrations of nitrate ($1-5 \mu\text{g m}^{-3}$) and nss sulfate ($5-20 \mu\text{g m}^{-3}$) over the Mediterranean Sea are about an order of magnitude greater than those at Barbados [Nguyen et al., 1974; Savoie, 1984] and are certainly high enough to support the dust-related concentrations measured over the TNA. Moreover, the mass ratio of dust-related nitrate to that of nss sulfate during the summer (0.36) is consistent with the mean ratio of about 0.31 over the Mediterranean [Savoie, 1984]. Possible transport from the populated regions along the southern coastal region of West Africa (Ivory Coast, Ghana, Nigeria, etc.) is severely curtailed, as the Intertropical Convergence Zone (ITCZ) generally lies between that region and the Sahara during the summer.

A likely source of the dust-related nitrate and nss sulfate during the winter is the region of West Africa south of the Sahara. During the winter the ITCZ shifts well to the south so that essentially all of West Africa is meteorologically within the northern hemisphere circulation patterns. The southward transport of hot, dry, dust-laden Saharan air produces the so-called harmattan periods over the south coastal region of West Africa [Delmas, 1980; Kalu, 1979; Morales, 1986; d'Almeida, 1986]. At this time of year, the nitrate-to-nss sulfate mass ratio is about 1.44, far too high to be consistent with a European pollutant source. In contrast, the emissions from south of the Sahara may well be compatible with this high nitrate:nss sulfate ratio. Major pollutant sources in that region include wood and biomass burning [Delmas, 1982] which release gases and particulates with far higher nitrogen-to-sulfur ratios than those produced from the combustion of fossil fuels. For example, Delmas [1982] suggested that the nitrogen-to-sulfur mass ratio released during biomass burning in the savannah region of the Ivory Coast is about 10-20. During the Amazon Boundary Layer Experiment conducted during July

and August in central Amazonia of Brazil, the average total nitrate-to-sulfate mass ratio in biomass-burning plumes was significantly greater than 1 [Andreae et al., 1988b]. High concentrations of soot carbon and fine-particle non-sea-salt potassium over the Atlantic between the equator and 10°N during October and November [Andreae, 1983] provide additional evidence for a major source over southern West Africa. If the major winter source is indeed the southern portion of West Africa, then the summerlike ratios which sometimes occur during the winter could simply reflect a temporary shift in the position of the ITCZ or other meteorological conditions in the region.

We hasten to note that while the data are consistent with the source areas which we have suggested, there is currently no other evidence that implicates specific regions. To fully understand the major sources or source regions for the summer and winter dust-related nitrate and nss sulfate clearly requires further study, and in fact, some additional investigations are already in progress. Backward trajectories from the free troposphere above Barbados are being computed under the supervision of J. Merrill at the University of Rhode Island. The objective is to determine (1) whether or not the source region for the material in the SAL can be more clearly defined, and (2) to determine whether or not there are clearly defined differences between the winter and summer seasons and, in the winter period, between the periods of normal winter ratios and those which exhibit summer type ratios. Weather maps are also being checked to assess variations in the location of the ITCZ and potential inflow into the source region of the Saharan dust. Studies of lead isotope ratios may also prove to be extremely useful in resolving this issue.

Summary and Conclusions

There appear to be two major categories of sources for the nitrate and nss sulfate at Barbados. The majority (about 60%) of each constituent is transported to Barbados in association with Saharan dust. On the basis of our current evaluation and the results from previous studies, we conclude that this fraction is ultimately derived from anthropogenic sources and not from Saharan soil material. The nitrate-to-nss sulfate mass ratio from this category changes radically between summer and winter, suggesting that there are probably at least two major source regions. On the basis of the general meteorological conditions, we suggest that Europe is probably the major source region during the summer and that the southern portion of West Africa is a likely major source region during the winter. While these sources are consistent with our current knowledge of the meteorology, considerably more research will need to be done to determine whether or not they are indeed the actual sources.

On the basis of the ratio of the mean MSA concentration to that of the nondust-related nss sulfate, the remaining 40% of the total nss sulfate is believed to be derived primarily from

the emission of reduced sulfur compounds from the ocean and their subsequent oxidation in the atmosphere. The primary source for the remaining 40% of the total nitrate is still uncertain; possibilities include, but are not necessarily limited to, production by lightning, oxidation of organic nitrogen compounds, and stratospheric injection. Trajectory analyses indicate that transport from North America could contribute to the nondust-related fractions; however, there is still too little information to attempt a quantitative estimate of the impact of this source.

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