Long-term measurements of the transport of African mineral dust to the southeastern United States: Implications for regional air quality

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Abstract. Continuous daily aerosol sampling carried out at a coastal site in Miami, Florida, for the past 23 years shows that large quantities of African mineral dust are periodically carried into Florida every summer, yielding daily concentrations in the range of about 10 µg m\(^{-3}\) to 100 µg m\(^{-3}\). Dust events typically last several days or longer. The maximum dust concentration occurs in July (monthly mean, 16.3 µg m\(^{-3}\)), but relatively high concentrations are also observed in June (8.4 µg m\(^{-3}\)) and August (9.8 µg m\(^{-3}\)). There is considerable year-to-year variability that is apparently linked to various meteorological factors including climate conditions in North Africa as manifested by drought. Satellite data show that African dust incursions are synoptic-scale events; consequently, they will impact a large region of the southern and eastern United States. The incursion of dust events over this large region, coupled with inputs from local emissions, could have important implications regarding regional air quality.

1. Introduction

Studies of ambient aerosols generally fall into two broad categories. Climate-oriented aerosol research generally focuses on the properties of aerosols that relate to radiative forcing either directly through the scatter and absorption of solar radiation or indirectly through the role of aerosols on cloud formation and cloud properties. Such studies tend to focus on aerosols that are significant on global and synoptic scales. A second category of research focuses on those aerosol properties that might relate to air quality issues, including health. Although it is recognized that long-range transport can impact on local air quality (for example, the transport of sulfur oxides from the midwestern United States to the Northeast and to Canada), it is generally assumed that the composition and concentration of airborne particulate matter in populous regions is largely controlled by emissions from proximate sources. In this paper, I present the results of 23 years of measurements in Miami which show that during summer months, large quantities of African dust are carried into the southern Florida region; as a result, African dust is the dominant aerosol constituent during the summer.

In July 1997 the Environmental Protection Agency (EPA) established a new standard for suspended particles [Federal Register, 1997] which applies to particles 2.5 µm diameter and smaller (henceforth the particulate matter (PM) 2.5 standard). The PM 2.5 standard specifies an annual mean of 15 µg m\(^{-3}\) and a 24 hour mean of 65 µg m\(^{-3}\) based on the 98th percentile of the frequency distribution averaged over 3 years. The EPA, which is required by the Clean Air Act to set standards for air quality at levels that protect public health with an adequate margin of safety, selected a size threshold of 2.5 µm diameter for the new standard based on studies that show that particles less than this diameter can efficiently penetrate into the lungs [Wilson and Spengler, 1996]. Previously, the standard had focused on particles having a diameter of 10 µm and smaller. The PM 10 annual mean standard is retained at 50 µg m\(^{-3}\); the 24-hour standard remains at 150 µg m\(^{-3}\), but it is based on the 98th percentile of the frequency distribution averaged over 3 years. The work presented here suggests that during dust events the concentration of dust, coupled with particles from local emissions, could conceivably yield aerosol concentrations that approach or exceed the EPA’s new standards. Because African dust incursions are synoptic-scale events the concentrations measured in Miami should be representative of a very large region of the southern and eastern United States. Thus dust events could affect compliance with EPA standards across this broad region.

The objective of this paper is to present a general overview of dust transport to southern Florida and to place it in the context of our current knowledge about African dust transport to the western North Atlantic Ocean. I also include data on the concentration of other species: non-sea-salt (nss) SO\(_4\), NO\(_3\), Na\(^+\), and NH\(_4\); the discussion of these data is limited to the general features of temporal trends in comparison to that of dust. The paper concludes with a discussion of the implications of dust transport with regard to air quality issues, including human health.

2. Procedures

We have sampled airborne aerosols in Miami (Figure 1) almost continuously since 1974 on the campus of the University of Miami, Rosenstiel School of Marine and Atmospheric Sciences (RSMAS) which is located at the water’s edge on the southeast end of Virginia Key (25°45’N; 80°15’W), an island about 4 km east of the mainland. Hight-
volume bulk aerosol samples are collected daily except over weekends and holidays when midday samples are taken. Samples are collected by drawing air through 20 x 25 cm Whatman 41 (W-41) filters at a flow rate of about 1.1 m³ min⁻¹, yielding average sampled volumes of about 1500 m³; the flow rate is measured by monitoring the pressure drop across a calibrated sharp-edged orifice plate. In the marine boundary layer the collection efficiency of W-41 filters is greater than 99% for nss SO₄²⁻ [Pszenny et al., 1993], 95% for NO₃⁻ marine aerosols, and 95% for dust [Saviole, 1984; Arimoto et al., 1990]. The sampling protocol has evolved over the duration of the sampling program in an effort to minimize the effects of local sources on samples. Initially, the sampler was operated continuously during the daily sampling period. In 1978, to minimize the impact of sources on the mainland during offshore wind conditions, a wind-direction sensor was added to the system, so the sampler was activated only when the wind blew from the open-ocean sector (approximately NE through east to south). The only land to the east of the site is the island of Key Biscayne, a small residential community with a population of 8000. Most of the island is set aside as park areas, which are largely covered with trees and shrubbery; much of the coast is densely covered with mangrove. There are no industrial activities on the island. The only substantial source of pollution would be emissions from automobiles. In 1987 a wind velocity sensor was added to the pump control system; the sampler was only activated when the wind-direction condition was met and when the wind speed was greater than 1 m s⁻¹. The low-speed cutoff criterion is intended to eliminate conditions when winds are light and variable. During the entire course of the program the sampling system has been placed on the roofs of various several-story-high buildings on the RMSAS campus. Beginning in 1987 the filter has been mounted on a 16-m fold-over tower on top of a 12-m-high building located 7 m above mean sea level and 10 m from the water's edge; the pump and associated flow-measuring and control equipment are housed at the base of the tower. To further minimize errors due to blanks and small sample sizes, I exclude data from samples that have a running time of 10% or less (i.e., sampling time divided by the total time that the filter was in place in the system). Short running times indicate that the winds were largely out of sector (i.e., come from over land) or that winds were weak and erratic. As shown below, there is no evidence of substantial impacts from local soil emissions.

Filters were first extracted with deionized water and the extracts were analyzed for major soluble inorganic ions: Na⁺ by flame atomic absorption; Cl⁻, NO₃⁻, SO₄²⁻ by suppressed ion chromatography [Saviole et al., 1980]; and NH₄⁺ by automated colorimetry. Nss SO₄²⁻ is calculated as follows: [total SO₄²⁻] - [Na⁺ x 0.2517], where 0.2517 is the SO₄²⁻/Na⁺ mass ratio in bulk seawater. The extracted filters were then placed in a muffle furnace for about 14 hours (overnight) at 500°C; the ash residue weight (less filter blank) is assumed to be mineral dust. The ash blanks yield a standard error in the mineral dust concentration that is essentially constant at ±0.1 µg m⁻³ for concentrations less than 1 µg m⁻³; at higher levels the standard error is about ±10%. The filter ash weight underestimates the true dust concentration because of the loss of soluble soil minerals during the extraction procedure and during heating, the volatilization of bound water and various species (e.g., some halides), and the breakdown of some components (e.g., carbonates). A scatterplot of AI concentration, measured by neutron activation [Arimoto et al., 1995; R. Arimoto, personal communication, 1997] against filter ash weights from approximately 1349 dust-laden filter samples collected at Barbados, yields an AI concentration of 10.4%. The average crustal abundance of AI in soils is in the range of 6%-8% [Taylor and McLennan, 1985]. Using the measured Al-filter ash percentage, 10.4%, and assuming an average Al concentration in soil dust of 8%, we obtain an adjustment factor of 1.3; all the dust data presented herein are adjusted using this factor. While we do not directly measure organic matter, work by others [Lepple and Brine, 1976] shows that African dust contains only about 3% organics.

3. Results

Initially, I focus on the daily aerosol record for the period 1989-1996 when the advanced wind sensor control system was in place and the filter was located on a 16-m tower on top of a three-story building at the water's edge. These samples are least likely to be impacted by sources on mainland Florida. Figure 2 shows the daily concentration data for mineral dust, nss SO₄²⁻, NO₃⁻, and sea-salt aerosol (calculated Na⁺ x 3.256, the ratio of dissolved salts to Na⁺ in seawater). Table 1 presents information on the statistical characteristics of the concentration distributions obtained from the daily measurements. The data (Figure 2a) show a clear seasonal periodicity with the maximum dust concentrations in June, July, and August. As will be shown in detail in section 4, this temporal pattern is consistent with the previous studies that show that during the summer months, large quantities of African dust are routinely carried across the tropical North Atlantic by easterly trade winds to the Caribbean region [Carlson and Prospero, 1972, Prospero and Carlson, 1972; Karyampudi and Carlson, 1988; Westphal et al., 1987, 1988; Prospero, 1996a, b].

The seasonal dust cycle in Miami is markedly different from the seasonal pattern of the other major aerosol species: nss SO₄²⁻, NO₃⁻, and Na⁺ (i.e., sea salt aerosol). Nss SO₄²⁻ (Figure 2b) and NO₃⁻ (Figure 2c) are largely attributable to pollution sources. The highest nss-SO₄²⁻ and NO₃⁻ concentrations occur in spring when the peninsula is often under the influence of
persistent northeast winds that can bring polluted air masses from the midlatitudes. Under typical synoptic meteorological conditions pollutants emitted from sources in the eastern United States are transported out to sea and, subsequently, brought into the Florida region by northeast winds. Alternatively, pollutants are carried behind cold fronts that penetrate to south Florida in the winter and spring. In Figure 2d, sea-salt concentrations show a strong seasonal cycle that peaks in the winter-spring in response to the stronger winds in those seasons.

The daily concentrations of dust and nss SO\(_4^{2-}\) during 1993 are shown in greater detail in Figure 3. There were several well-defined dust episodes that affected south Florida in 1993. African dust events usually occur in association with a steady southeast trade wind that is characteristic of the climatology of Florida in the summer [Henry et al., 1994]. Dust episodes usually extend over several days or more; given the persistence of the trade-wind flow, this suggests that the scale of the dust events is of the order of several hundred to 1000 km; this scale is consistent with satellite depictions of aerosol distributions over the western North Atlantic and the Caribbean [Husar et al., 1997; Herman et al., 1997]. Thus measurements of aerosol concentration and composition in Miami should be representative of a very large region. As an example, on August 4-5, 1993 (Figure 3), dust concentrations reached 97 \(\mu g \cdot m^{-3}\). Coincidentally, the State of Florida's air quality monitoring network (which adheres to a one-day-in-six sampling cycle) was operating on the same day; 40 monitoring sites from the panhandle to the Florida Keys reported unusually high concentrations of total suspended particles, over 80 \(\mu g \cdot m^{-3}\) and as high as 113 \(\mu g \cdot m^{-3}\). Isentropic air mass trajectories [Merrill, 1994] show that during most of the month of August, trajectories consistently arrive from the southerly sectors, over the tropical Atlantic they hook to the east, toward Africa. The pattern of these dust-bearing trajectories is consistent with the general large scale dust transport patterns observed on Barbados (13.17\(^\circ\)N, 59.43\(^\circ\)W) [Savoie et al., 1989, 1992] and on Bermuda (32.27\(^\circ\)N, 64.87\(^\circ\)W) [Arimoto et al., 1992, 1995]. These same patterns are observed in Miami every year in conjunction with dust events.

During dust events the concentration of nss SO\(_4^{2-}\) is substantial (typically several \(\mu g \cdot m^{-3}\)) but lower than winter-spring concentrations (Figure 3); also, the ratio of dust to nss SO\(_4^{2-}\) is high. Although the summertime concentrations of nss SO\(_4^{2-}\) (and NO\(_x\)) are relatively low, they are higher than concentrations normally associated with unimpacted ocean regions. Studies on Barbados [Savoie et al., 1989, 1992] and

**Table 1. Daily Aerosol Concentration Statistics, 1989-1996**

<table>
<thead>
<tr>
<th>Date</th>
<th>Dust(^a)</th>
<th>Sea Salt(^a)</th>
<th>NO(_x)</th>
<th>nss SO(_4^{2-})^(^b)</th>
<th>NH(_x)</th>
<th>Total(^a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>4.72</td>
<td>8.67</td>
<td>1.95</td>
<td>2.23</td>
<td>0.72</td>
<td>18.21</td>
</tr>
<tr>
<td>Maximum</td>
<td>120.65</td>
<td>58.00</td>
<td>25.61</td>
<td>16.84</td>
<td>10.50</td>
<td>133.34</td>
</tr>
<tr>
<td>Quartile 75%</td>
<td>3.26</td>
<td>10.60</td>
<td>2.26</td>
<td>2.81</td>
<td>0.96</td>
<td>20.00</td>
</tr>
<tr>
<td>Median</td>
<td>1.19</td>
<td>7.72</td>
<td>1.38</td>
<td>1.65</td>
<td>0.49</td>
<td>15.15</td>
</tr>
<tr>
<td>Quartile 25%</td>
<td>0.53</td>
<td>5.29</td>
<td>0.91</td>
<td>1.02</td>
<td>0.28</td>
<td>11.77</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.03</td>
<td>0.44</td>
<td>0.27</td>
<td>0.08</td>
<td>0.00</td>
<td>1.47</td>
</tr>
<tr>
<td>Count</td>
<td>1503</td>
<td>1525</td>
<td>1526</td>
<td>1519</td>
<td>1523</td>
<td>1522</td>
</tr>
</tbody>
</table>

\(^a\)Concentrations, \(\mu g \cdot m^{-3}\)

\(^b\)Negative values deleted from data.

\(^c\)Sea salt computed from Na concentration x 3.256.

\(^d\)Statistics calculated from the daily "total" aerosol concentration.
on Tenerife (Canary Islands, 28.30°N, 6.50°W) [Prospero et al., 1995] show that the nss SO$_4^{2-}$ and NO$_3^-$ associated with African dust are most likely derived from sources in Europe. The synoptic situations associated with dust storms over Africa favor the transport of European air masses across the Mediterranean and into North Africa where pollutants become mixed with suspended dust. On Barbados, about half of the nss SO$_4^{2-}$ and NO$_3^-$ is believed to be anthropogenic [Savoie et al., 1989, 1992]. In Miami during the summer, subtle shifts in meteorological conditions can result in rapid changes in relative concentrations. For example, the very large nss-SO$_4^{2-}$ peak observed at the end of August 1993 (Figure 3) is associated with the shift to isentropic back trajectories that trace back to the Gulf of Mexico and then north into the central United States. On August 25, trajectories shifted once again to the east, and nss SO$_4^{2-}$ dropped sharply.

Figure 4 shows for the period 1989 to 1996 the daily aerosol concentrations for the sum of all major inorganic species: mineral dust, sea salt, nss SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$. Most concentration values fall in the range of about 10-20 µg m$^{-3}$. On a day-to-day basis the highest total aerosol concentrations occur during the summer, and these are associated with dust incursions. The dominance of dust aerosols during the summer months is seen in Figure 5, which shows the monthly mean aerosol concentrations for the major species for the period 1989-1996; Table 2 presents the means of the eight years of monthly means along with the maximum and minimum monthly means for that period. The maximum dust concentration occurs in July with a mean of 16.3 µg m$^{-3}$ (range of July means, 8.12-29.9 µg m$^{-3}$). Relatively high concentrations are also observed in June (8.41 µg m$^{-3}$) and August (9.83 µg m$^{-3}$), and minor amounts in September.

Figure 4. Total daily concentrations of major inorganic aerosol constituents in onshore winds at Miami, 1989-1996; the sum of nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, mineral dust, and seasalt (calculated from Na$^+$ concentration multiplied by 3.756, the mass ratio of total salts in seawater to that of Na$^+$).
When dust is not the dominant species, sea-salt aerosol is dominant with monthly means in the range 6.64-12.44 μg m⁻³. As can be seen in Figure 5, concentrations are high in the winter and spring and dip to relatively low (but still substantial) values in the summer at the time when dust concentrations peak. The sea-salt concentrations measured at the Miami site are not strongly influenced by local sources of salt spray such as surf zones. As previously stated, the sampling site is located on a tower on a building at the water’s edge on Virginia Key; Key Biscayne lies east of the site, across a narrow channel, between Virginia Key and the Straits of Florida. On occasion during the winter there is substantial surf action on the east shore of Key Biscayne. However, wave action is seldom very heavy because of the protection afforded by the Bahama Islands to the east; the short fetch over the Straits of Florida precludes the buildup of large waves. Because of these factors and the location of the filler at a height of 30 m above sea level, sea-salt aerosol concentrations measured at the site are probably fairly representative of those carried inland by the wind.

The monthly mean nss-SO₄²⁻ concentrations ranged from 0.85 to 5.0 μg m⁻³ (Table 2) over the period 1989-1996. As shown in Figure 5 and Table 2, monthly mean nss-SO₄²⁻ concentrations tend to increase steadily in the early part of the year, reaching a maximum in May (8-year May mean, 3.23 μg m⁻³; range of May means, 1.78-4.92 μg m⁻³) and a minimum in January (1.45 μg m⁻³). The seasonal cycle of NO₃⁻ is similar to that of nss SO₄²⁻. The individual monthly mean NO₃⁻ concentrations range from 0.87 to 3.83 μg m⁻³. The maximum is in April (8-year mean, 2.23 μg m⁻³; range of April means, 1.35-3.15 μg m⁻³); the minimum, 1.49 μg m⁻³, is in November. As previously stated, the high nss-SO₄²⁻ and NO₃⁻ concentrations are associated with trajectories that carry pollutants from the eastern United States. Nonetheless, it is clear from Figure 5 that on a mass basis, NO₃⁻ and nss SO₄²⁻ are relatively minor constituents compared to sea salt and mineral dust.

The annual means for the major aerosol species during each year, 1989-1996, are shown in Figure 6, along with the average for the entire period. Except for dust, the annual means fall in a narrow range: nss SO₄²⁻, 1.71-2.47 μg m⁻³; NO₃⁻, 1.52-2.04 μg m⁻³; sea salt, 7.79-10.63 μg m⁻³. In contrast, dust concentrations vary by a factor of about 2, 2.90-6.31 μg m⁻³. This variability is related to a variety of factors that will be discussed below.

4. Discussion

4.1. General Characteristics of Dust Events

There are a variety of physical observations that clearly distinguish the summer dust events from other types of aerosol episodes. Intense dust events are usually associated with the occurrence of dense haze and a large aureole around the Sun. The dust haze has a yellow-brown color that is distinctly different from the bluish-gray cast of pollution hazes, which are usually associated with the high concentrations of nss SO₄²⁻ and NO₃⁻ in the spring. Aerosol filters collected during summer dust events have a distinct red-brown coloration; in contrast, during other seasons the filters typically are gray [Prospero et al., 1987]. Rain collected during intense summer dust events can have a turbid appearance. When filtered, these rains yield a distinct “cake” of red-brown mud [Prospero et al., 1987] which, when dried and rubbed with the fingers, produces an extremely fine, rouge-like powder. Indeed, brief showers often leave a clearly visible deposit of reddish-brown dust spots on cars [Prospero et al., 1987]. In contrast, during winter rain events, there is very little sediment, and the sediment is often gray colored and gritty, characteristics that are associated with particles derived from pollution sources and from local soils which often have a high content of calcium carbonate. It should be noted that summer is the rainy season in Florida with about 50% of the annual rainfall occurring in the period June-September [Henry et al., 1994]; consequently, soils are well wetted and vegetation coverage is dense. In contrast, winter is the dry season. It is also the agricultural season when fields are tilled and planted; during winter, intense cold fronts, often carrying high winds, pass through the area. Thus we would
Table 2. Mean of Monthly Mean Aerosol Concentrations and Maximum and Minimum Monthly Mean Values, Miami, 1989-1996

<table>
<thead>
<tr>
<th></th>
<th>Mineral Dust</th>
<th>Sea Salt</th>
<th>NO₃</th>
<th>sea SO₄</th>
<th>NH₄</th>
<th>Total Aerosol</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean</td>
<td>Max</td>
<td>Min</td>
<td>Mean</td>
<td>Max</td>
<td>Min</td>
</tr>
<tr>
<td>Jan.</td>
<td>0.86</td>
<td>1.58</td>
<td>0.57</td>
<td>10.65</td>
<td>13.04</td>
<td>8.21</td>
</tr>
<tr>
<td>Feb.</td>
<td>1.05</td>
<td>2.30</td>
<td>0.57</td>
<td>10.41</td>
<td>14.93</td>
<td>6.45</td>
</tr>
<tr>
<td>March</td>
<td>1.57</td>
<td>2.64</td>
<td>0.84</td>
<td>10.10</td>
<td>11.54</td>
<td>9.14</td>
</tr>
<tr>
<td>April</td>
<td>1.61</td>
<td>2.34</td>
<td>1.12</td>
<td>10.45</td>
<td>13.70</td>
<td>7.13</td>
</tr>
<tr>
<td>May</td>
<td>2.48</td>
<td>4.71</td>
<td>1.00</td>
<td>7.72</td>
<td>10.04</td>
<td>5.62</td>
</tr>
<tr>
<td>June</td>
<td>8.41</td>
<td>16.43</td>
<td>1.46</td>
<td>7.21</td>
<td>9.00</td>
<td>5.46</td>
</tr>
<tr>
<td>July</td>
<td>&gt;16.30</td>
<td>&gt;29.86</td>
<td>&gt;8.12</td>
<td>7.57</td>
<td>8.62</td>
<td>5.66</td>
</tr>
<tr>
<td>Aug.</td>
<td>9.83</td>
<td>22.59</td>
<td>5.25</td>
<td>6.96</td>
<td>8.59</td>
<td>5.45</td>
</tr>
<tr>
<td>Sept.</td>
<td>3.90</td>
<td>9.71</td>
<td>1.11</td>
<td>6.64</td>
<td>10.13</td>
<td>4.48</td>
</tr>
<tr>
<td>Oct.</td>
<td>0.99</td>
<td>1.95</td>
<td>0.55</td>
<td>8.15</td>
<td>12.87</td>
<td>4.97</td>
</tr>
<tr>
<td>Nov.</td>
<td>0.81</td>
<td>1.53</td>
<td>0.29</td>
<td>&gt;12.44</td>
<td>&gt;19.82</td>
<td>8.28</td>
</tr>
<tr>
<td>Dec.</td>
<td>0.88</td>
<td>1.58</td>
<td>0.53</td>
<td>8.79</td>
<td>11.57</td>
<td>7.56</td>
</tr>
</tbody>
</table>

Within each column, maximum values are shown by >, minimum values are shown in italics. All concentrations are in units μg/m³.

*Negative values deleted from data.

bSea salt computed from Na concentration x 3.256.

cStatistics calculated from the daily "total" aerosol concentration.
expect local soil dust sources to be most active in winter; yet dust loads are at a minimum during the winter (Figures 2, 3, 5). Some winter dust events are seen (e.g., in December 1993, Figure 3), but the dust concentrations are quite low compared to summer events and the ratio of nss SO$_2$ to dust is very high compared to summer values.

Several lines of evidence link Miami dust to African sources. First of all, measurements of aerosol properties and the associated meteorology have clearly established that large amounts of dust are transported out of Africa and across the tropical North Atlantic each year [Carlson and Prospero, 1972; Prospero and Carlson, 1972; Karyampudi and Carlson, 1988; Westphal et al., 1987, 1988; Prospero, 1996a, b]. Many studies have taken place in Barbados, West Indies (13°15'N, 59°30'W; see Figure 1), where the University of Miami aerosol group has been carrying out a continuous sampling program since 1965 [Prospero and Nees, 1986; Savoie et al., 1989; Li et al., 1996], using the same equipment and protocols as those used in Miami. The seasonal pattern of dust concentrations in Miami is similar to that at Barbados, as shown in Figure 7, for the period 1989-1996. The major differences between the records from the two sites are that the dust concentrations are consistently higher at Barbados and that the dust transport season on Barbados is longer than that in Miami, where transport starts later in the year and ends earlier. Note also that the winter dust concentrations at Barbados are generally greater than those at Miami, although there is usually one month each winter where concentrations are comparable. The low winter concentrations measured in Miami relative to Barbados confirm that local and regional soil sources are not important in south Florida. As a result of these various factors, the annual mean concentration at Barbados is about 2.5 times that in Miami [Prospero, 1994b].

Various satellite products [Ott et al., 1991; Husar et al., 1997; Herman et al., 1997] enable us to trace individual dust outbreaks from the time they emerge from the coast of Africa until they enter the Caribbean about one week later. The AVHRR (advanced very high resolution radiometer) aerosol optical thickness (AOT) product [Husar et al., 1997] shows a "plume" of high values of AOT over the tropical Atlantic, extending from the coast of Africa to the Caribbean. The TOMS (total ozone mapping spectrometer) UV observations of absorbing aerosols are especially useful because the system is only responsive, in effect, to two types of aerosols, mineral dust and smoke [Herman et al., 1997] and because the system can detect aerosols over both land and water surfaces. Figure 1 shows the frequency of occurrence (days per month) of moderate-to-high absorbing aerosol values over North Africa and the North Atlantic during the month of July 1984 [see Herman et al., 1997, Figure 6]. The TOMS absorbing aerosol "plume" visually links the Miami and the Barbados dust to sources in West Africa. Both TOMS [Herman et al., 1997] and AVHRR [Husar et al., 1997] show that the African dust plume undergoes a seasonal displacement reaching its northernmost position in July and August and its southernmost position in the low latitudes of the North Atlantic in the winter. The seasonal oscillation of the plume is consistent with the seasonal cycle of dust concentrations as measured in Miami and Barbados [Husar et al., 1997; Chiapello et al., 1998] as shown in Figure 7. The fact that the dust "season" in Barbados is longer than that in Miami reflects the fact that Barbados lies in the principal latitude of the plume transport as clearly depicted in AVHRR and TOMS.

### 4.2. Long-Term Trends

Summer dust transport has been a persistent feature throughout the 23 years of measurements in Miami, 1974-1996, as shown by the monthly mean dust concentrations in Figure 8. Nonetheless, there were very substantial variations in dust concentration over this period. Concentrations were consistently high during the period 1983-1987. The early 1980s was a time of severe drought in North Africa. Previous
work has shown that summer dust concentrations measured in the trade winds at Barbados are anticorrelated with rainfall in the sub-Saharan (Sahel) region of North Africa [Prospero and Nees, 1986; Prospero et al., 1993]. The highest monthly mean dust concentrations were obtained in Miami in July 1983 (35.8 μg m⁻³), the year following the onset of one of the most intense El Niño events in recent history; a similar sharp increase in dust was also observed on Barbados [Prospero and Nees, 1986]. The longer-term variability of dust transport could be linked in a complex way to other climate variables such as the North Atlantic Oscillation [Moulin et al., 1997]. Dust concentrations decreased after 1993 possibly as a consequence of near-normal rainfalls in the Sahel [Halpert et al., 1996].

Figure 7. Monthly mean concentrations of mineral dust at Miami and Barbados, 1989-1996.

Figure 8. Monthly mean mineral dust concentrations measured in Miami, 1974-1996. Note that during the early part of the record, the sampler was not sectored for wind direction. Thus winter mineral dust concentrations in the early years are probably influenced by local sources; the summer concentrations should be essentially unaffected by local sources because of the strong easterly flow and the rainy conditions that prevail.
4.3. Implications for Air Quality

When placed in the context of the meteorology and climatology of the region, it is clear that dust events are large-scale phenomena that can affect a very large region. This conclusion is supported by other short-term aerosol studies. As a part of a large study of mercury deposition in Florida, a network of aerosol, gas and precipitation sampling stations was established at sites extending from the panhandle to the Florida Keys [Landing et al., 1995]. The deposition rate of Al in precipitation in 1993-1994 ranged from 0.062 to 0.148 g m⁻² yr⁻¹. Assuming an average crustal abundance of 8% Al, these are equivalent to dust deposition rates ranging from 0.78 to 1.9 g m⁻² yr⁻¹; the average, 1.14 g m⁻² yr⁻¹, is essentially identical to that obtained by Prospero et al. [1987] in 1983-1984, 1.25 g m⁻² yr⁻¹. Almost all the deposition of soil dust takes place during the summer months, which is also the rainy season in Florida.

Recently, Perry et al. [1997] assessed the temporal and spatial variability of PM 2.5 soil dust particle concentrations (based on the measured concentration of Al, Ca, Fe, Si, and Ti in twice-weekly daily samples) in a network of approximately 70 sites located in national parks and wilderness areas during the period 1992-1995. The highest individual PM 2.5 soil concentrations were associated with sites in the eastern United States during the summer, not in the arid southwest as one might expect. Furthermore, there was a large-scale coherence in the temporal variability of the high PM 2.5 values, suggesting that they were associated with synoptic-scale processes. The elemental composition of the samples in large PM 2.5 events was distinctly different from nonevent samples; furthermore, the composition of the samples at sites in the eastern United States during these events was identical to samples collected in the Virgin Islands. These observations are consistent with the hypothesis that the high PM 2.5 episodes were associated with incursions of African dust. Indeed, the progress of some of these dust incursions could be followed in the data as they moved from the Caribbean and Gulf of Mexico into the southern states and across the northeastern United States. Independently, in a retrospective study of data from a field program held in central Illinois in the summer of 1979, Gazz and Prospero [1996] noted the occurrence of unusually high concentrations of Si, Al, and other crustal elements. Back trajectories from Illinois showed flow from the Gulf of Mexico. Concurrent mineral dust measurements at Miami show that there was a strong influx of North African dust at the same time. High concentrations of African dust are routinely observed on Bermuda during the summer [Arimoto et al., 1992, 1995]. The transport of African dust to the western Atlantic has been taking place on geological timescales; for example, the composition of soils on Bermuda is consistent with African dust sources and shows little evidence of inputs from North America [Herwitz et al., 1996].

The frequent occurrence of high concentrations of dust over such large regions has implications regarding air quality and compliance with EPA’s air quality standards for PM 2.5 particles. Measurements of size distributions made at various sites in the western Atlantic suggest that a large fraction of the dust mass is 2.5 μm diameter or less. Hardy et al. [1976] made size distribution measurements with a five-stage cascade impactor at three sites in the Miami area during July 8-18, 1974. Data from the University of Miami site show that African dust concentrations were high during much of the Hardy study, ranging from 2.3 to 33.0 μg m⁻³ (average 14.7 μg m⁻³). The dust event was not recognized as such by Hardy at the time of their field study, nor in the ensuing publication, although they do comment on the surprisingly high concentration of crustal material which they attribute to local sources such as road dust. The Fe/Ti ratio in their samples was close to the crustal ratio, about 0.1 [Taylor and McLennan, 1985]. The size distribution of Fe (computed from Table 1, Hardy et al.) suggests that 32% of the mineral dust mass was less than 2 μm diameter, and 73% was below 4 μm diameter; the Ti data yield 44 and 81%, respectively. Li-Jonas and Prospero [1998] present the results of size distribution (cascade impactor) measurements made on Barbados in April 1994 during which time four large African dust events occurred [Li et al., 1996], 43% of the dust mass was less than 2.5 μm aerodynamic diameter and 18% less than 1.25 μm diameter. Thus it appears that about a third to a half of the African dust mass over the western Atlantic would conform to EPA’s PM 2.5 criterion. Past studies have shown that the mass median diameter of mineral dust over the oceans is typically 2-3 μm [Duce, 1995], for example, Asian dust over the central Pacific [Arimoto et al., 1985].

EPA’s new PM 2.5 standard specifies a 24-hour mean of 65 μg m⁻³ (based on the 98th percentile of the frequency distribution averaged over 3 years). If a third to a half of the African dust mass entering the south Florida area is less than 2.5 μm diameter, as suggested above, then many dust events will yield daily PM 2.5 concentrations that would constitute a substantial fraction of the 24-hour standard. Note in Figure 7 that in every year there was one or more dust events that yielded concentrations over 50 μg m⁻³ (which implies PM 2.5 dust concentrations over about 25 μg m⁻³); in 1993 and 1995 there were events that yielded dust concentrations over 100 μg m⁻³ (i.e., PM 2.5 dust over 50 μg m⁻³). African air masses would, in effect, bring into the region very high PM 2.5 “background” aerosol concentrations. Although emissions from local sources in themselves might be relatively small compared to the PM 2.5 standard, in combination with the advected dust (and sea salt) aerosols the 24-hour standard could be challenged. Because dust events occur in the summer months, the time of year when pollution episodes are frequent and intense in the eastern United States, there is a high probability of the simultaneous occurrence of dust and pollution events.

These same issues are relevant to the PM 10 standard (annual mean, 50 μg m⁻³; 24-hour standard, 150 μg m⁻³, based on the 99th percentile). The annual mean “total” aerosol was 18.21 μg m⁻³ (Table 1) of which dust contributed 4.72 μg m⁻³. Thus dust does not contribute much to the total aerosol concentration based on the annual mean standard. From the standpoint of the 24-hour standard, there were periods (1993 and 1995) when concentrations were in the range of 120 μg m⁻³ and above (Figure 4). Thus it is conceivable that the PM 10 24-hour standard might be challenged as well as the PM 2.5 standard.

In assessing compliance the EPA has the authority to exclude air quality data that are affected by “exceptional events” caused by natural sources [Federal Register, 1997]. In this category the EPA [Nichols, 1996] specifically identifies volcanic eruptions, wild-land fires, and high-wind events. While the last category implicitly addresses the possible impact of soil dust, the description of this category as “high wind events” makes it clear that the EPA assumes that soil dust will be derived from local sources and that a direct
association can be made on the basis of the occurrence of high wind speeds. None of these categories directly addresses the impact of the long-range mineral dust which, in the case of African dust, is not associated with high wind speeds. Therefore in order to account for the impact of such events, it will be necessary to develop a set of diagnostic indicators for African dust. The mineral composition of the dust might serve as an indicator. Dust collected in the western Atlantic is identical to that collected off the coast of Africa (Glaccum and Prospero, 1980); the dominant constituents are clay minerals and quartz. Other possible indicators are dust elemental composition (Glaccum, 1978; Perry et al., 1997; Gates and Prospero, 1996; Arimoto et al., 1992, 1995; Harwitz et al., 1996), meteorological characteristics of air masses (Carlson and Prospero, 1972; Gates and Prospero, 1996), and various satellite products (Husar et al., 1997; Herman et al., 1997). Indeed, using satellite imagery and atmospheric transport models, it should be possible to predict the areas that will be impacted by specific dust events.

African dust events could impact on other aspects of air quality. As shown here, dust events occur during the summer months, the time of year when conventional photochemical pollution episodes are common in the eastern United States; such events are characterized by high concentrations of SO2, various oxides of nitrogen and volatile organic compounds that together lead to the production of ozone, and the formation of particles that contain acid species such as SO3 and NO3. The presence of high concentrations of dust could affect the photochemical evolution of polluted air masses. Modeling studies suggest that because of the large surface area of the suspended dust and the presence of neutralizing species (e.g., CaCO3) in dust, the particles could serve as a sink for reactive species [Zhang et al., 1994; Deniener et al., 1996]. Li-Jones and Prospero [1998] showed that in the presence of African dust, as much as 70% of the NSS sulfate can be associated with supramicrometer particles. Recently, Dickerson et al. [1997] presented evidence that aerosols, by modifying the radiative balance in the atmosphere, can have a pronounced effect on photochemical smog formation; they predict that UV-absorbing aerosols such as mineral dust should sharply reduce the rate of ozone production. It should be possible to test the Dickerson et al. hypothesis by studying the evolution of photochemically produced species during the course of the day in the presence of an African dust event in urban areas of the southeastern United States.

4.4. Health Issues

Finally, it should be recognized that the EPA standards for airborne particulate materials are largely based on epidemiological studies of specific at-risk communities in regions that experience relatively high concentrations of anthropogenic particles and other airborne pollutants. These studies suggest that increased illness and morbidity are associated with increased concentrations of airborne particles. Because of the complexity of particle-related health effects, it has not been possible to unambiguously identify specific cause-effect relationships for specific components in aerosols or validate mechanisms by which low concentrations of particles could cause cardiopulmonary toxicity [Uteil and Samet, 1996]. Andur [1996] and Uteil and Samet [1996] review various hypotheses regarding mechanisms; they present evidence that suggests that aerosols coated with first-row transition metals are especially efficient in producing an inflammatory response in the lungs. In particular, they suggest that surface-complexed iron can generate hydroxyl radicals in lung tissue and that these radicals have acute lung toxicity. In this regard, it should be noted that African dust particles collected on Barbados have a total Fe content of 3.4% [Zhu et al., 1997]. The particles are heavily coated with iron which accounts for the characteristic red-brown color of filters collected during dust events. Under acid conditions, 6% of the total Fe content of dust is easily dissolved [Zhu et al., 1997]; thus it might be expected that a substantial fraction of the Fe (and presumably other first-row transition elements) on dust could be readily mobilized in the lung when the particles are deposited on lung tissue. Nonetheless, in the case of African dust, I am not aware of any studies that relate dust concentrations to health effects. Thus it is not possible to anticipate what effect, if any, African dust itself might have on human health in the areas impacted in the United States.

5. Conclusions

Over a 23-year period, African dust was the dominant component in aerosols collected in Miami during summertime onshore winds. These results, coupled with other shorter-term studies and with satellite measurements, clearly establish that African dust events affect a large area of the southern and southeastern United States. Thus African air inversions bring high "background" aerosol concentrations which, when coupled with local emissions, could cause problems with respect to compliance with the EPA standard for particulate matter, especially the PM 2.5 24-hour standard. There is no evidence that African mineral dust, in itself, constitutes a health problem. However, a large fraction (a third to a half) of the dust mass does fall into the "respirable" size range, and consequently, African dust can efficiently penetrate into the human respiratory system. It remains to be seen if the composition of the dust (i.e., the presence of high concentrations of silicates, iron, and other metals) is such that inhaled particles would produce a physiological effect.

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References


PROSPERO: AFRICAN DUST TRANSPORT TO THE EASTERN UNITED STATES


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