



# Global Biogeochemical Cycles

## RESEARCH ARTICLE

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### Key Points:

- Spring dust transport to South America comparable to Caribbean in summer
- African dust events at Cayenne and Amazonia linked to emissions from Sahel
- WHO PM<sub>10</sub> air quality standard is frequently exceeded because of African dust

### Supporting Information:

- Readme
- Figure S1
- Figure S2
- Figure S3

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## Characterizing the annual cycle of African dust transport to the Caribbean Basin and South America and its impact on the environment and air quality

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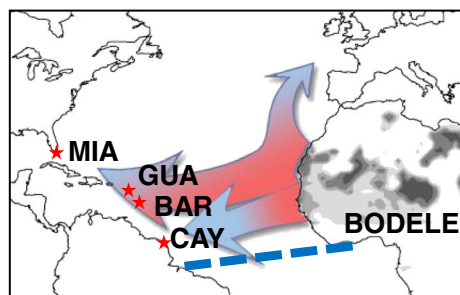
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**Abstract** Decades of aerosol measurements on Barbados have yielded a detailed picture of African mineral dust transport to the Caribbean Basin that shows a strong seasonal cycle with a maximum in boreal summer and a minimum in winter. Satellite aerosol products suggest that in spring, there is a comparable transport to northeastern South America. Here we characterize the complete annual cycle of dust transport to the western Atlantic by linking the Barbados record to multiyear records of airborne particulate matter less than 10  $\mu\text{m}$  diameter (PM<sub>10</sub>) measured in air quality programs at Cayenne (French Guiana) and Guadeloupe. Comparisons of PM<sub>10</sub> at these sites with concurrent dust measurements at Barbados demonstrate that high PM<sub>10</sub> levels are almost entirely due to dust. Cayenne PM<sub>10</sub> peaks in spring in a cycle which is consistent with satellite aerosol optical depth and suggests that the Sahel is the dominant source. The persistent transport of dust during much of the year could impact a wide range of environmental processes over a broad region that extends from the southern United States to the Amazon Basin. Finally, the average 24 h PM<sub>10</sub> concentrations at Cayenne and Guadeloupe frequently exceed the World Health Organization air quality guideline. Thus soil dust PM<sub>10</sub> could be a significant, but generally unrecognized, health factor at western Atlantic sites and also in other relatively remote regions affected by long-range dust from Africa. Because dust emissions and transport are highly sensitive to climate variability, climate change in coming decades could greatly affect a wide range of biogeochemical processes and human health in this region.

## 1. Introduction

Mineral dust plays an important role in many atmospheric and ocean processes that relate to climate and Earth biogeochemical processes [Shao *et al.*, 2011]. Africa is the world's largest dust source, emitting an estimated 800 Tg yr<sup>-1</sup>, about 70% of the global total [Huneeus *et al.*, 2011]. Consequently, there is much interest in the factors that affect dust emissions in Africa, the impact that they might have on the environment, and their ultimate fate. A large fraction of these emissions are carried across the west coast of North Africa to the western Atlantic. Long-term measurements on Barbados [Prospero and Lamb, 2003; Prospero and Mayol-Bracero, 2013] and Miami [Prospero, 1999] have documented the great variability of dust transport to the Caribbean Basin on time scales ranging from days to decades. Various studies have examined how this variability might be linked to meteorological and climate processes [e.g., Prospero and Lamb, 2003; Mahowald *et al.*, 2011; Evan *et al.*, 2011; Guo *et al.*, 2013] and to the impact of humans [Mahowald *et al.*, 2010; Ginoux *et al.*, 2012].

The Barbados record shows a strong seasonal cycle with a maximum in boreal summer and a minimum in winter. The seasonality is consistent with satellite measurements of aerosol optical depth (AOD) which in summer show huge plumes of high-value AOD extending from the coast of Africa to the Caribbean Basin, the Gulf of Mexico, and the southern United States [Hsu *et al.*, 2012; Chin *et al.*, 2014; Yu *et al.*, 2013; Kim *et al.*, 2014]. At Barbados, AOD has been correlated to dust concentrations as measured at the surface [Smirnov *et al.*, 2000]. These same satellite products suggest that there is a comparable transport to the western Atlantic in winter and spring, but that it is largely confined to the latitudes south of Barbados, with the plume axis crossing the coast of South America in the region of French Guiana and Surinam [Ben-Ami *et al.*, 2012; Chin *et al.*, 2014; Huang *et al.*, 2010; Adams *et al.*, 2012; Yu *et al.*, 2013]. However, the interpretation of satellite AOD products is complicated by the presence of smoke from biomass burning which is very intense in North Africa in winter



**Figure 1.** North African dust source areas and transport paths to the western Atlantic. The most active dust source regions in North Africa are identified by the gray-scaled areas which are indicative of the frequency of occurrence of intense dust events, detected as high concentrations of absorbing aerosols by the Total Ozone Mapping Spectrometer [Prospero *et al.*, 2002]. The Bodélé Depression stands out as one of the most active sources. The main seasonal transport paths across the Atlantic are schematically depicted by broad arrows, the more northerly one representative of summer paths and the southerly one of the winter-spring transport route. The climatological position of the Intertropical Convergence Zone (ITCZ) in February is shown with a dashed line.

and spring in regions proximate to strong dust sources [Adams *et al.*, 2012]. Evidence that plume AOD might be strongly linked to desert dust was provided by a brief study at Cayenne in the late 1970s [Prospero *et al.*, 1981] which measured high mineral dust concentrations in spring. Since that time there have been no studies in this region that could be readily linked to the present-day satellite and modeling products. The spring transport has received increased attention in recent years because of the possible impact of African dust on soil nutrient balance in Amazon Basin soils [Bristow *et al.*, 2010; Swap *et al.*, 1992; Ben-Ami *et al.*, 2010; Abouchami *et al.*, 2013], on regional radiation and cloud processes [Prenni *et al.*, 2009; Martin *et al.*, 2010], and on nutrient deposition to the equatorial Atlantic [Baker *et al.*, 2010, 2013; Ussher *et al.*, 2013]. Based largely on satellite and modeling studies, transport to Amazonia has been linked to a specific source, the Bodélé Depression in northern Chad [Ben-Ami *et al.*, 2010, 2012] one of the most persistently active and intense dust sources on Earth [Washington *et al.*, 2009]. If Bodélé emissions do indeed play such an important role, it would serve as an example of a major source-receptor impact persistently acting over a great distance, about 9000 km.

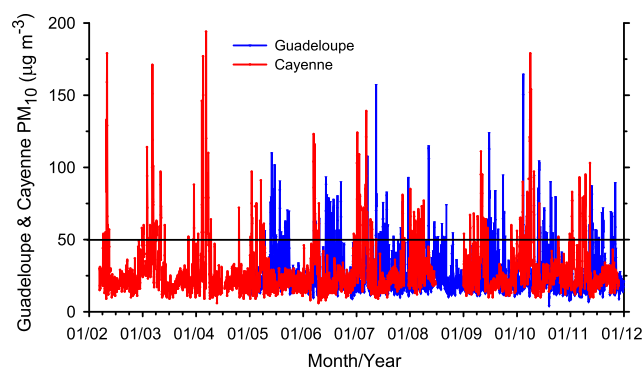
Here we present a multiyear record of daily aerosol measurements made at Cayenne, French Guiana (04.95°N, 52.31°W), and Guadeloupe, Lesser Antilles (16.23°N, 61.53°W), as part of a French national air quality program using instruments that continuously monitor concentrations of particles that have an aerodynamic diameter less than 10  $\mu\text{m}$ , commonly referred to as  $\text{PM}_{10}$ .

Our primary objective in this work is to use the existing multiyear  $\text{PM}_{10}$  air quality data to provide a key to characterizing dust transport to South America. In this way we develop a picture of the complete annual cycle of transport to the western Atlantic and the temporal and spatial relationships in this transport across the region. To this end we first present  $\text{PM}_{10}$  data sets from Cayenne and Guadeloupe and we discuss them in a regional context. Because the  $\text{PM}_{10}$  measurements are not speciated, we assess the contribution of dust to  $\text{PM}_{10}$  by comparing daily values at these sites with concurrent measurements at Barbados where the dust component is specifically measured. Guadeloupe is located 400 km northwest of Barbados and Cayenne is 1200 km to the southeast (Figure 1). As we will show, in summer dust events frequently impact Guadeloupe and Barbados simultaneously and in spring occasional events impact all three sites thereby enabling a direct comparison. By linking the 10 year record at Cayenne with that at Barbados, we provide the basis for assessing the longer-term variability of transport to the region. We then discuss the general significance of large-scale transport to this region in terms of various environmental processes and also the possible impact of  $\text{PM}_{10}$  dust transport on human health in the region. Finally we discuss the implications of climate change for future transports.

## 2. Methods and Environmental Setting

Aerosol sampling at Cayenne and Guadeloupe is carried out under the aegis of Les Associations Agréées de Surveillance de la Qualité de l'Air (AASQA), a national organization that oversees air quality monitoring in each of the France's administrative regions.  $\text{PM}_{10}$  concentrations are measured using the Thermo Scientific Tapered Element Oscillating Microbalance (TEOM) models 1400ab and 1400-FDMS. The TEOM is widely used in air quality programs in Europe and the United States and qualifies as a U.S. Environmental Protection Agency equivalent method (<http://www.epa.gov/ttn/amtic/criteria.html>). Measurements are made continuously and stored as 15 min averages which were used to calculate a 24 h daily averages which start at midnight local time. In Cayenne,  $\text{PM}_{10}$  measurements began in 2002 and on Guadeloupe in 2005 (Figure 2).

Cayenne is the capital of French Guiana, an overseas region and department of France. Instruments are located near the center of the city which has a population of 76,000. Cayenne has a tropical monsoon climate



**Figure 2.** Daily average  $\text{PM}_{10}$  concentrations ( $\mu\text{g m}^{-3}$ ) at Cayenne and Guadeloupe. The horizontal line depicts the World Health Organization Air Quality Guideline (3) for 24 h mean  $\text{PM}_{10}$  concentrations,  $50 \mu\text{g m}^{-3}$ .

which is characterized by a long wet season, nominally from December through June, which is also the period when dust transport is most active. The climate in Cayenne is largely controlled by the movement of the Intertropical Convergence Zone (ITCZ) which reaches its maximum northerly limit in August and its southerly limit in February and March, months when Cayenne experiences very steady northeast winds [Molinié and Pontikis, 1995] and when, as we will show,  $\text{PM}_{10}$  and dust transport are typically at the annual maximum.

On Guadeloupe sampling is carried out at a site on the southern outskirts of Pointe-à-Pitre, which lies south of the major population centers on Guadeloupe (total population 210,715) with the exception of Le Gosier (population 26,550) which lies to the southeast of the site. Winds are predominantly from the east except when disturbances pass through the region. The rainy season begins in August, after the peak of the dust season, and extends through November.

There was concern that the measurements made at Cayenne and Guadeloupe were impacted by local sources. In French Guiana, this issue was addressed by making concurrent measurements with identical instrumentation at two sites located outside the urban area. These measurements are discussed in the supporting information and shown in Figures S1 and S2. Because the  $\text{PM}_{10}$  peak heights are the same at both sites, we conclude that local aerosol impacts are not significant.

On Barbados, measurements are made at a field station located at Ragged Point, an elevated promontory on the east coast ( $13.165^{\circ}\text{N}$ ,  $59.432^{\circ}\text{W}$ ) [Prospero and Lamb, 2003]. Daily aerosol samples are collected using  $20 \times 25$  cm Whatman 41 filters which have a collection efficiency for dust that is greater than 95% [Kitto and Anderson, 1988; Arimoto *et al.*, 1990]. At Miami, the filters are extracted with deionized water and ashed in a muffle furnace at  $500^{\circ}\text{C}$ . The ash residue weight (less filter blank) is assumed to be mineral dust. A scatterplot of Al concentration, measured by neutron activation [Arimoto *et al.*, 1995], against filter ash weights (corrected for losses during extraction and ashing) from 1349 dust-laden Barbados filter samples yields an Al concentration of 8% which is consistent with the range of average abundances in crustal materials 6%–8% [Taylor and McLennan, 1985]. Using this method for measuring dust, a large suite of elements in Barbados aerosol samples yield concentrations that are close to crustal abundances [Trapp *et al.*, 2010]. With respect to the comparison of Barbados dust measured in this way with  $\text{PM}_{10}$ , we note that previous studies of size distributions on Barbados using cascade impactors have shown that the mass median diameter of dust is  $2.3 \mu\text{m}$  [Arimoto *et al.*, 1997] and that over 90% of the total dust mass is less than  $10 \mu\text{m}$  diameter [Li-Jones and Prospero, 1998]. Similar results were obtained in comparisons of speciated  $\text{PM}_{10}$  measurements against bulk dust concentrations in South Florida during dust events [Prospero *et al.*, 2001]. Thus, we would expect that  $\text{PM}_{10}$  measurements at Cayenne and Guadeloupe should capture essentially all the airborne dust mass along with the mass added by other aerosols, mostly sea salt.

### 3. Results

#### 3.1. $\text{PM}_{10}$ Time Series

$\text{PM}_{10}$  concentrations at Cayenne and Guadeloupe (Figure 2) follow a clear seasonal cycle at both sites but they are out of phase by several months: at Cayenne high values of  $\text{PM}_{10}$  occur from January to May; at Guadeloupe from May to September. During these peak periods,  $\text{PM}_{10}$  at Cayenne and Guadeloupe frequently exceeds the World Health Organization [2006] 24 h air quality guideline (AQG),  $50 \mu\text{g m}^{-3}$ , demarked by the line in Figure 2. At Guadeloupe,  $\text{PM}_{10}$  exceeded the AQG on 258 days out of a total of 2799 days (9.2%). At Cayenne, the AQG was exceeded 246 days out of a total of 2765 days (9.0%) (Table 1).

**Table 1.** Sampling Statistics and Frequency of Days With  $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$  at Cayenne and Guadeloupe

Location		Annual Statistics of $\text{PM}_{10}$ Concentrations											
Cayenne	Year	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	Mean <sup>a</sup>
	Sample Days	275	340	337	356	365	329	358	362	350	326	230	345.6
	Mean $\text{PM}_{10}$	—	29.0	26.9	23.4	25.4	31.0	—	27.4	28.5	28.5	—	27.5
	Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	17	41	31	16	20	49	24	31	28	30	29	30.8
	% Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	6.2	12.1	9.2	4.5	5.5	14.9	6.7	8.6	8.0	9.2	12.6	9.0
Guadeloupe	Sample Days	—	—	—	354	358	357	355	365	354	334	322	349.9
	Mean $\text{PM}_{10}$	—	—	—	27.3	27.9	27.8	24.9	24.8	27.5	24.4	28.0	26.6
	Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	—	—	—	28	42	41	15	25	43	21	43	32.3
	% Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	—	—	—	7.9	11.7	11.5	4.2	6.8	12.1	6.3	13.4	9.2
		Frequency of Occurrence $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$ per Month <sup>b</sup>											
Cayenne	Month	1	2	3	4	5	6	7	8	9	10	11	12
	Sample Days	264	237	305	296	326	296	312	331	330	335	317	279
	Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	50	34	106	60	38	7	0	0	0	3	6	12
	%, Month	18.9	14.3	31.5	20.3	11.9	2.4	0.0	0.0	0.0	0.9	1.9	4.3
Guadeloupe	Sample Days	248	216	248	229	248	238	246	232	206	242	230	216
	Days $\text{PM}_{10} \geq 50 \mu\text{g m}^{-3}$	1	6	28	6	34	53	61	33	18	12	5	1
	%, Month	0.4	2.3	11.3	2.6	13.7	22.3	24.8	14.2	8.7	5.0	2.2	0.5

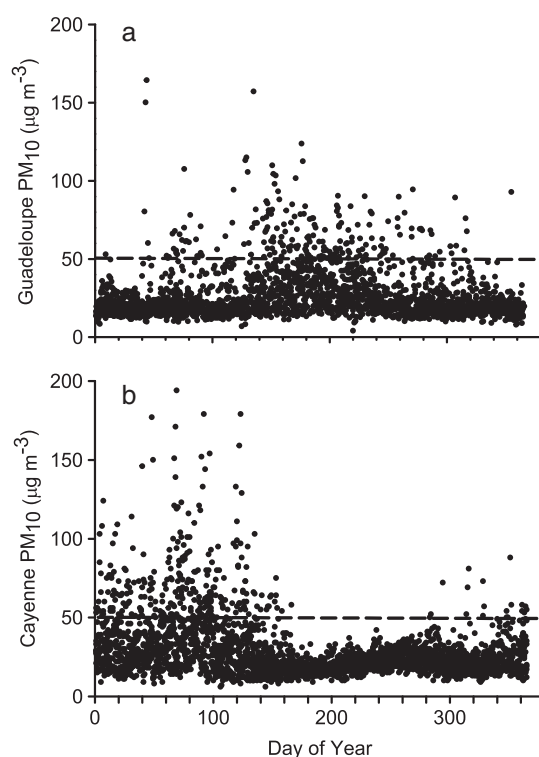
<sup>a</sup>Based on years with essentially complete data: Cayenne, 2003–2007, 2009–2011; Guadeloupe, 2005–2012.

<sup>b</sup>Based on all months with essentially complete data: Cayenne, 2002–2012; Guadeloupe, 2005–2012.

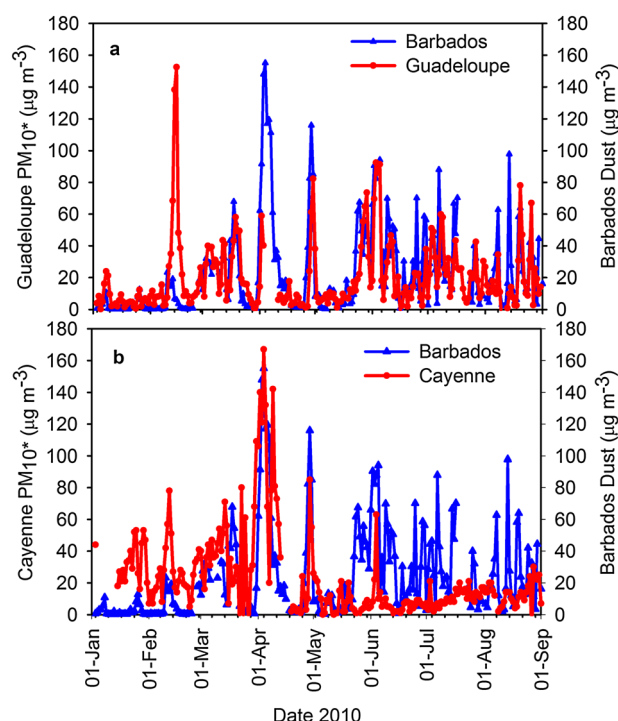
The seasonal cycle at Guadeloupe and Cayenne is more clearly seen in Figure 3 which shows all daily mean  $\text{PM}_{10}$  plotted against the day-of-the-year (DOY). The seasonal maxima are consistent with satellite-based aerosol measurements which show a seasonal to-south-to-north shift of trans-Atlantic dust plumes as the year progresses [Hsu et al., 2012; Adams et al., 2012; Yu et al., 2013; Chin et al., 2014; Ridley et al., 2012; Ben-Ami et al., 2010, 2012]. At Guadeloupe (Figure 3a), most values fall in a narrow band over much of the year, except

during the period from May through August (DOY 120 to 240) when African dust transport is most likely to be present, based on comparisons to Barbados. Except for those months,  $\text{PM}_{10}$  is mostly in the range  $10\text{--}20 \mu\text{g m}^{-3}$ .

At Cayenne (Figure 3b),  $\text{PM}_{10}$  values fluctuate widely from December through May, ending abruptly at about DOY 140–160, in response to the expected advection of African aerosol. This seasonality and the  $\text{PM}_{10}$  concentration levels are consistent with the Cayenne dust measurements reported in Prospero et al. [1981] in the late 1970s and with the satellite climatology of high-value AOD plumes from African as cited above. In contrast,  $\text{PM}_{10}$  yields a very coherent pattern from about early June (DOY 160), when the ITCZ moves over the region and when winds become southeasterly, until late November (DOY 340) when the ITCZ retreats south. In June and July,  $\text{PM}_{10}$  values are at an annual minimum, falling within a narrow range, roughly  $10\text{--}20 \mu\text{g m}^{-3}$ . After this low period, Cayenne  $\text{PM}_{10}$  steadily increases during the summer with values mostly in a narrow range; values reach a maximum in September in the range  $20\text{--}30 \mu\text{g m}^{-3}$ . At this time of year, southeast winds often carry mineral dust and



**Figure 3.** Scatterplot of all Cayenne (2002–2011) and Guadeloupe (2005–2011)  $\text{PM}_{10}$  daily values against the day-of-the-year. The horizontal line marks the WHO  $\text{PM}_{10}$  AQG value,  $50 \mu\text{g m}^{-3}$ .



**Figure 4.** Daily PM concentrations ( $\mu\text{g m}^{-3}$ ) at Cayenne and Guadeloupe compared to mineral dust concentrations at Barbados in 2010. To facilitate the comparison, the measured PM<sub>10</sub> concentrations are adjusted for a nominal background of  $12 \mu\text{g m}^{-3}$  and designated as PM<sub>10</sub>\*. (a) Guadeloupe PM<sub>10</sub>\* and Barbados mineral dust. (b) Cayenne PM<sub>10</sub>\* and Barbados mineral dust. The large peak in early February at Guadeloupe in Figure 4a is linked to the eruption of Soufrière on Montserrat, 90 km to the northwest of Guadeloupe.

at this time of year; these dust events often extended over a broad latitude band which in some cases spans all three sites, a latitudinal distance of 1200 km.

### 3.2. Linking PM<sub>10</sub> to Dust

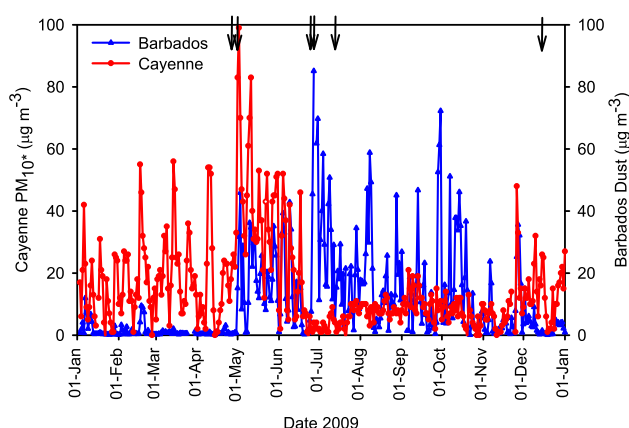
Because the routine PM<sub>10</sub> air quality protocol only requires mass measurements, samples are not analyzed for species composition. Consequently, we cannot directly ascribe the Cayenne and Guadeloupe PM to specific components such as dust. However, we can assess the impact of dust through comparisons to daily dust measurements on Barbados [Prospero and Lamb, 2003]. To facilitate the visual comparison of PM<sub>10</sub> with Barbados dust measurements, we adjust the PM<sub>10</sub> data by subtracting a nominal “background” concentration. At Guadeloupe during winter and much of the spring (Figure 3a) and at Cayenne (Figure 3b) during the quiescent period from about DOY 175 to 225, PM<sub>10</sub> values show a lower bound at about  $12 \mu\text{g m}^{-3}$ . We use this value as the PM<sub>10</sub> background value which we would expect to be comprised largely of sea-salt aerosol based on measurements at Barbados [Li-Jones and Prospero, 1998]. The adjusted-background PM<sub>10</sub> values are henceforth shown as PM<sub>10</sub>\*.

To demonstrate the PM<sub>10</sub> can be related to dust, we compare in Figure 4a the daily PM<sub>10</sub>\* record at Guadeloupe in 2010 with that of dust at Barbados. The Guadeloupe record begins with an extremely large peak in early February which is linked by satellite images and other reports to the 11 February eruption of Soufrière on Montserrat 90 km to the northwest ([http://en.wikipedia.org/wiki/Soufriere\\_C3%A8re\\_Hills](http://en.wikipedia.org/wiki/Soufriere_C3%A8re_Hills)). Over this same time period, a large multiday dust/PM<sub>10</sub>\* event was observed on Barbados and Cayenne but atmospheric Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) back trajectories [Draxler and Rolph, 2003] suggest that these were due to dust transport from Africa, not from the eruption.

From early March to early May, all three sites measured essentially identical dust-PM<sub>10</sub>\* concentrations. Especially notable is the extremely large dust event in early April [Jung et al., 2013], one of the largest in

biomass burning aerosol from northeast Brazil. The shift to Brazilian sources is reflected in a distinct change in dust mineralogy from that measured during the spring period [Prospero et al., 1981]. At both Cayenne and Guadeloupe during their respective dust seasons, daily PM<sub>10</sub> values frequently exceed the World Health Organization (WHO) AQG,  $50 \mu\text{g m}^{-3}$  (Figure 3). At Cayenne in the peak month, March, the daily average WHO AQG is exceeded on 34.8% of the days and at Guadeloupe in July on 24.8% of the days (Table 1).

Note that at Cayenne, the highest PM<sub>10</sub> concentrations are most frequently measured in a period centered at about DOY 75, i.e., mid-March. At this time, a similar period of increased frequency of high PM<sub>10</sub> events is also observed in Guadeloupe; this brief active period at Guadeloupe is distinctly separated from the onset of the summer dust transport season which begins around DOY 125, i.e., early April. Later we will show that some of the high PM<sub>10</sub> values measured at Cayenne and Guadeloupe (and also dust at Barbados) in the March period are linked to huge dust outbreaks seen along the coast of Africa



**Figure 5.** Daily Cayenne  $PM_{10}^*$  and Barbados mineral dust concentrations, 2009. The arrows at the top of the figure identify specific dust events discussed later in the text.

recent years; the Guadeloupe instrument captured the beginning of the event but failed shortly thereafter. Over the remainder of the record in Figure 4a, both the Barbados and Guadeloupe sites yielded much the same picture from the standpoint of phasing and concentrations although there are some differences (e.g., late May and mid-August) that are linked to differences in the path followed by the main plume through the islands as seen by satellite. At Cayenne (Figure 4b),  $PM_{10}^*$  is generally concordant with Guadeloupe and Barbados from early March to mid-May at which time  $PM_{10}^*$  levels drop sharply. The good

agreement between  $PM_{10}^*$  values at Guadeloupe and Cayenne with dust at Barbados during the March–May period suggests that large values of  $PM_{10}^*$  are almost entirely due to African dust. This is consistent with previous studies on Barbados [Li-Jones and Prospero, 1998; Arimoto *et al.*, 1997] and Puerto Rico [Maring *et al.*, 2003] which showed a rapid decrease in particle concentrations at sizes above about 7  $\mu m$  diameter. Indeed, it is notable that few dust particles above 10  $\mu m$  are present in air masses leaving the west coast of Africa [Maring *et al.*, 2003; Schepanski *et al.*, 2009; Müller *et al.*, 2010]. The general agreement in dust and  $PM_{10}$  measurements at Guadeloupe, Barbados, and Cayenne during March and April suggests that over much of this period dust covered much of the western tropical Atlantic in broad plumes. As we will show later, the degree of agreement over this spring time period is not always as good as in 2010, a consequence of year-to-year changes in dust source activity and shifts in transport winds.

### 3.3. Linking Seasonal Trends in $PM_{10}$ and Dust to Transport

The seasonal shift in dust transport patterns can occur quite rapidly as shown in the records from Cayenne and Barbados in 2009 (Figure 5). We use HYSPLIT back trajectories (henceforth, BTs) (Figure 6) to look for changes in source regions and transport paths that might explain these changes. BTs [Draxler and Rolph, 2003] are calculated for altitudes that are based on the past studies of African dust transport [e.g., Carlson and Prospero, 1972; Prospero and Carlson, 1972; Reid *et al.*, 2003] and on dust vertical distributions over the Atlantic [Adams *et al.*, 2012; Campbell *et al.*, 2012; Koffi *et al.*, 2012; Huang *et al.*, 2010; Tsamalis *et al.*, 2013] made with satellite lidar measurements with the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) instrument aboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations [Winker *et al.*, 2013]. During summer, dust transport occurs mainly in the Saharan Air Layer (SAL), a hot, dry dust-laden layer located above the top of the marine boundary layer (MBL) [Carlson and Prospero, 1972]. Over the western Atlantic, the SAL tops out at about 3–4 km as shown in CALIOP [Huang *et al.*, 2010; Adams *et al.*, 2012; Koffi *et al.*, 2012; Tsamalis *et al.*, 2013; Kim *et al.*, 2014] and in ground-based lidar profiles on Barbados [Campbell *et al.*, 2012]. In winter and spring, transport takes place mainly below about 2.5 km with the core between 1.5 and 2.0 km [Huang *et al.*, 2010; Tsamalis *et al.*, 2013]. In our BTs, we use two altitudes: 500 m, in the MBL, and 2000 m, near the nominal core of the SAL.

In Figure 5 we see four distinct changes in aerosol concentrations over time. First, from January through May, there is steady dust transport to Cayenne and essentially none to Barbados until early May when concentrations increased sharply at both sites. In Figure 6a we show BTs for 26 April 2009 (marked by an arrow in Figure 5) shortly before the large jump in dust concentrations. The 2000 m BT from Cayenne tracks across the southwest coast of North Africa and into the Sahel region. In contrast, Barbados BTs hook to the central North Atlantic and turn westward across North America, a pattern that is common for this time of year. In Figure 6b, BTs are shown for 30 April 2009 (arrow), the day when concentrations peak at both sites; both 2000 m BTs track back to the Sahel and Sahara regions. Note that the 500 m MBL BTs do not cross the coast of Africa in Figure 6a or 6b. This is a persistent feature of MBL BTs in this series of examples and also in

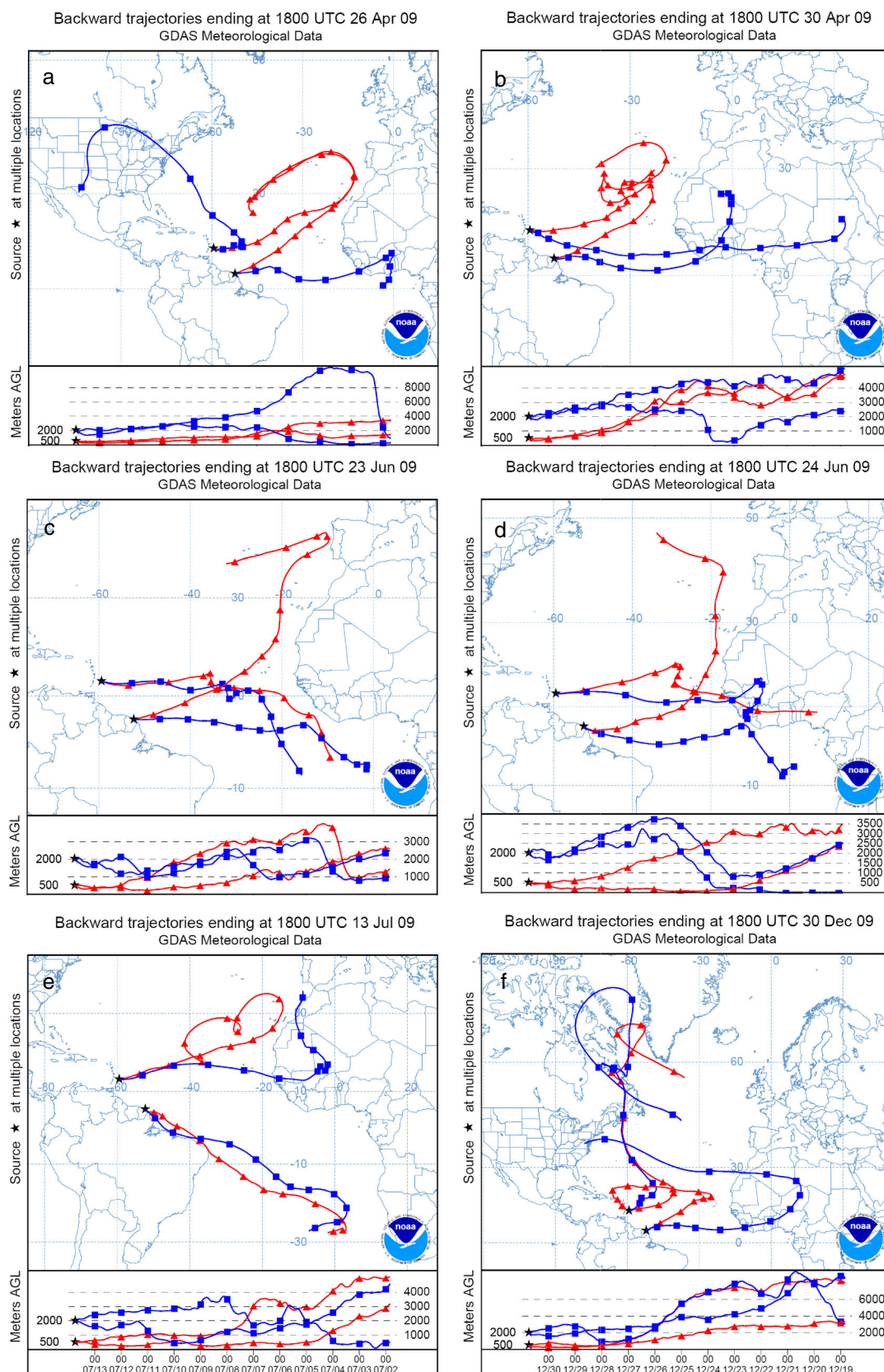
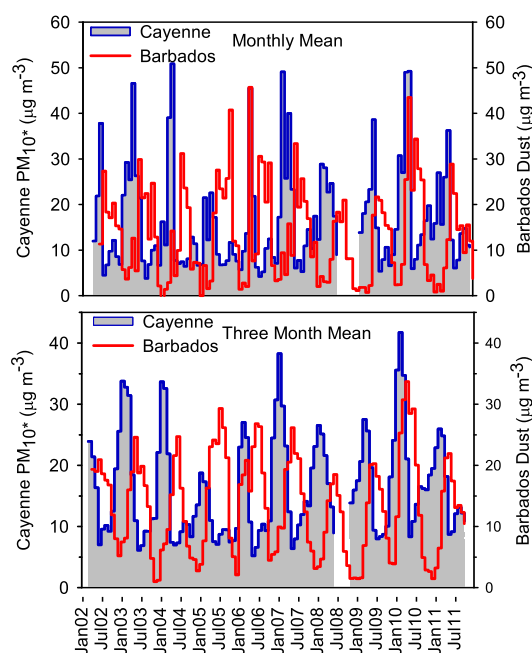


Figure 6



**Figure 7.** Cayenne  $PM_{10}^*$  and Barbados dust mean concentrations, 2002–2011. (top) Monthly means and (bottom) 3 month moving averages.

along or over the northeast coast of Brazil (Figure 6e, 13 July). Finally in late November and into December, we see evidence of the start of next dust transport season. In Figure 6f, 30 December, Cayenne 2000 m BTs once again track back to the Sahel and the Sahara while the Barbados BTs go to the North Atlantic. Note that in all examples in Figure 6, the transit time from the coast of Africa to Cayenne and Barbados is 5–6 days, times that are typical based on the visual tracking of dust outbreaks in satellite products.

For purposes of clarity, in Figure 6 we only plotted one daily BT pair for each site, choosing 1800 UT to approximate midday transport to Barbados and Cayenne. As one might expect, BTs calculated over such a long time period and over a data-sparse environment can be subject to considerable variability. To characterize this variability we present in the supporting information (Figure S3) HYSPLIT ensemble BTs for these same 6 days. For the most part, especially those BTs going back to Africa, the BTs in each ensemble track rather closely across the tropical Atlantic but they generally lose coherence over the continent. Using 24 June 2009 as an example, the BT bundles from Barbados and Cayenne are tightly grouped on two distinctly different paths over the ocean but over West Africa, the BTs in each bundle disperse widely and the two bundles overlap considerably. A similar example is 30 April 2009. Thus, it is unrealistic to use BTs in an attempt to link specific aerosol samples to specific source regions in Africa. An additional major source of variability in the ensemble BTs is found in the altitude distribution. As seen in Figure S3, it is not possible to reliably link a dust sample to a specific altitude or to a specific source region with any degree of certainty.

### 3.4. Longer-Term Variability of Dust Transport to the Receptor Region

To examine the longer-term time variability, we show in Figure 7a Cayenne and Barbados monthly means. In most years, there is little overlap in the respective dust transport seasons. Major exceptions occurred in 2009 and 2010, as discussed above. Another was in March 2006. In general (with the notable exception of 2005 and 2006), the seasonal peak  $PM_{10}^*$  concentrations at Cayenne are greater than those of Barbados dust; in most years,  $PM_{10}^*$  at Cayenne serves as a fairly good predictor of Barbados dust concentrations. Dust seasonality is more clearly seen in Figure 7b which shows the same data as 3 month moving means. There

most of our case studies of dust events in this work and in our past work. There is considerable debate about the source of the dust in the MBL over the western North Atlantic: is it injected directly into low-level air near the coast of Africa or is it transported primarily in the SAL and subsequently mixed downward into the MBL during transit [Reid *et al.*, 2003; Carlson and Prospero, 1972]? There is evidence that both modes may play a role and that the relative importance changes from event-to-event and with the season.

Toward the end of June, dust transport to Cayenne and Barbados ends abruptly. An example of this transition is shown with BTs on 23 June 2009 (Figure 6c), when PM was low at both sites. In this example and others over this interim period, Cayenne BTs generally track to the Gulf of Guinea and the Gulf coastal states. One day later, 24 June 2009 (Figure 6d), dust increases sharply at Barbados and 2000 m BTs lead to the Sahel region.

In mid-July at Cayenne (Figure 5, marked by an arrow), PM transport from Brazil begins [Prospero *et al.*, 1981]. At this time, BTs from Cayenne show a strong southeasterly tendency with many moving

**Figure 6.** NOAA HYSPLIT back trajectories [Draxler and Rolph, 2003] from Barbados and Cayenne selected for transitional dust periods in 2009, marked by arrows in Figure 4. The 500 m trajectories start in the marine boundary layer over both sites; the 2000 m level is located in the nominal Saharan Air Layer. The symbols on the altitude and trajectory paths mark the 24 hour points. HYSPLIT ensemble back trajectories for these same days are presented in the supporting information Figure S3.

can be considerable variability in the breadth of the dust season from year-to-year and, in any 1 year, large differences between the two sites. Nonetheless, the general coherence of the dust records from these two sites suggests that the factors affecting African dust transport are broadly based and persistent over the course of a year. However, the year-to-year relative variability in the breadth of the dust seasons at Cayenne and Barbados as seen in Figure 7b suggests that other factors can strongly modulate dust activity and transport. In this context, 2005 and 2006 stand out as anomalies. In 2005, the Barbados dust season was especially strong and long in duration while that at Cayenne was unusually weak and short. In 2006, the Barbados season was also strong and long and there was a long overlap with the Cayenne season which, in contrast, was markedly weaker. It is notable that the monthly mean  $\text{PM}_{10}$ \* Cayenne concentrations measured in this study are comparable to those from Cayenne in 1978–1979 [Prospero *et al.*, 1981]. In contrast to 2005 and 2006, dust transport to Cayenne was unusually strong in 2010 not only during the nominal dusty season but also through the summer and fall, eventually merging with the 2011 dust season.

### 3.5. Linking Dust Transport to African Sources

The strong contrast between the seasonal cycles at Cayenne and those at Guadeloupe-Barbados and the rapid switching in dust seasonal transport paths implies that the African dust plume maintains a relatively uniform latitudinally constrained distribution over the western Atlantic during much of the year.

The seasonal shift in the plume is linked in part to changes in dust source regions [Prospero *et al.*, 2002; Formenti *et al.*, 2011; Ben-Ami *et al.*, 2012] coupled with changes in the large-scale wind fields [Engelstaedter *et al.*, 2006; Engelstaedter and Washington, 2007] and in part to the seasonal progression of the Intertropical Convergence Zone (ITCZ) and the West African Monsoon [Williams, 2008]. Early in the year Sahel sources are most active, especially the Bodélé Depression in northern Chad, one of the most persistently active and intense dust sources in the world [Koren *et al.*, 2006; Washington *et al.*, 2009]. In spring satellites show huge dust pulses emerging from the Bodélé and moving to the West on an almost daily basis [Knippertz and Todd, 2012; Koren *et al.*, 2006]. Ben-Ami *et al.* [2012] used satellite AOD to map out dust transport paths over the tropical Atlantic during 2000–2009, a period that overlaps much of our record. They found that during the Cayenne dust season, the AOD over the eastern tropical Atlantic is highly correlated with dust emissions from the Bodélé and thus suggests that the Bodélé is the main source of dust at Cayenne.

Meteorological processes associated with the monsoon front over West Africa at this time of year are thought to play an important role in dust mobilization and transport [Williams, 2008; Knippertz and Todd, 2012; Schepanski *et al.*, 2009]. The CALIOP lidar satellite shows intense dust activity along the northern edge of the front, the activity migrating with the seasonal movement of the front [Adams *et al.*, 2012]. Doherty *et al.* [2012] analyze composites of wind and precipitation during the winter season; they find that the multidecade interannual variability of Barbados winter dust loads (and by extrapolation, Cayenne dust loads) is related to changes in near-surface northeasterly winds over the Sahel - Bodélé region coincident with the movement of the ITCZ. Wind variability plays a large role in driving changes in dust emissions in the region [Ridley *et al.*, 2013].

These studies suggest that emissions from the Bodélé are likely a major, perhaps dominant, contributor to PM collected at Cayenne. As mentioned above, African emissions in general and Bodélé emissions in particular are believed to have a significant impact on a variety of processes over the Amazon basin and to play a role in soil formation. During an extended measurement campaign in Amazonia at a site 1200 km southwest of Cayenne [Rizzo *et al.*, 2013], episodes of increased concentrations of Al, Si, Ti, and Fe were observed and attributed to African dust. Four periods in early 2008 were specifically identified as African events. These periods precisely match the timing of dust peaks in Cayenne although concentrations were much lower than at Cayenne, probably because of removal by rain during the transit from the coast.

Ideally we like to be able to link our aerosol samples to specific sources in Africa by relating the composition of our dust samples to those of suspected African sources. Comprehensive studies by Scheuven *et al.* [2013] and Moreno *et al.* [2006] find substantial differences in the mineralogical and elemental composition of dust from active source regions that could conceivably serve as the basis for identification. However, these data are largely based on soil samples or deposited dust samples. Because of the loss of large dust particles during transit, the composition of the long-range dust could differ substantially from that of the source material. Indeed, large differences are seen in size distribution of dust over or near African sources but these largely disappear after relatively short transport times [Stuut *et al.*, 2005; Mahowald *et al.*, 2013]. At Barbados, after a

week in transit, the mineral composition is relatively unchanging from event-to-event [Glaccum and Prospero, 1980] and, as previously stated, the elemental composition does not show evidence of any substantial variability over the course of a year [Trapp *et al.*, 2010]. However, recent work at several Caribbean sites suggests that source signatures might be retrieved from isotopic composition (e.g., Sr-Nd-Hf and Pb) and rare earth element anomalies [Pourmand *et al.*, 2014; Kumar *et al.*, 2014; Aboucamy *et al.*, 2013].

The presence of African dust and biomass burning products over the Amazon was also detected in lidar aerosol profiles [Ansmann *et al.*, 2009; Baars *et al.*, 2011] although smoke was often the dominant component in the aerosol profile. Extensive Polarization-Raman-lidar observations made in early 2008 [Baars *et al.*, 2011] at a site (2.5°N, 60.0°W) 60 km north of Manaus show dust as a prominent component in AOD with profiles typically extending to altitudes of 2–3 km. The strong events cited in Baars *et al.* follow within a day or two the appearance of strong PM<sub>10</sub>\* events in Cayenne where values mostly peaked in the range of 40–60  $\mu\text{g m}^{-3}$ . A large Amazon event on 08 May was part of a huge dust outbreak that yielded PM<sub>10</sub>\* around 30  $\mu\text{g m}^{-3}$  at Cayenne, dust concentrations of about 80  $\mu\text{g m}^{-3}$  on Barbados, and PM<sub>10</sub>\* values of about 100  $\mu\text{g m}^{-3}$  on Guadeloupe. Thus, the plume associated with this one dust event simultaneously impacted a latitudinal span of at least 2000 km.

As the season shifts to summer in Africa, sources farther to the north (e.g., in northern Chad, Mali, Mauritania, and southern Algeria) become more active, linked to the development and movement of African easterly waves in concert with extratropical disturbances [Knippertz and Todd, 2010]. Thus, changes in the terrain properties in these source regions (e.g., rainfall, vegetation cover, and land use [Ginoux *et al.*, 2012]), and the meteorological processes that affect them will act to modulate transport to receptor sites in the western Atlantic. In this context, the exceptional years noted in Figure 7, 2005 and 2006 (especially 2005), warrant study to better understand the cause of this anomaly—was it due to changes in source area activity or to transport or to both? Year-to-year changes in dust transport to Cayenne relative to Guadeloupe and Barbados may be a sensitive indicator of climate variability over West Africa that impacts on dust emissions. Anomalies such as these will serve as a good test of the ability of models to forecast dust emissions and long-range transport.

## 4. Impacts of Dust Transport on the Receptor Regions

### 4.1. Deposition to Soils

Dust is known to play a role in a wide range of atmospheric, ocean, and land surface processes. These have been addressed in recent reviews [e.g., Shao *et al.*, 2011; Jickells *et al.*, 2005; Maher *et al.*, 2010; Muhs, 2012; Schulz *et al.*, 2012; Mahowald *et al.*, 2011]. We touch briefly on some aspects of these processes as they relate to our regional interests.

African dust has been shown to be a major contributor to soils on many islands in the Caribbean, Florida, and the Bahamas [Muhs *et al.*, 2007] and Bermuda [Muhs *et al.*, 2012]. Our present study suggests that dust could play a similar role in northern South America. In particular, as previously discussed, Sahel emissions are estimated to be an important source of nutrients deposited to the Amazon Basin. Phosphorous is of particular interest because of its role as a major nutrient in both ocean and terrestrial systems [Okin *et al.*, 2011a; Mahowald *et al.*, 2008, 2009]. Measurements at Barbados and Miami show that aerosol-P concentrations are highly correlated with the concentration of African dust [Zamora *et al.*, 2013] and that P accounts for 880 ppm of the dust mass, slightly higher than the average crustal abundance, 665 ppm [Wedepohl, 1995]. A large fraction of the P (8–18%) is readily soluble in water and thus would quickly be released to soils. Given the large soluble fraction of P and the high deposition rate of dust during the dust season [Prospero *et al.*, 2010], there will be a rapid release of P to soils, thus providing a burst of “fertilizer” to plants during the major growth season.

Recent studies show that African dust can be a significant source of P to regional soils. Das *et al.* [2013] show that dust-P deposition is important on the Yucatan Peninsula [Das *et al.*, 2013]. Glaser *et al.* [2013] in a study of peat cores in the Florida Everglades show that dust-P has apparently played a large role in South Florida over their record which extends to 4600 years before present. It is notable that dust deposition rates were apparently much higher over the early record, 4600–2800 years ago, which suggests that either dust emissions were much greater at that time or that transport and deposition patterns have subsequently

shifted. West Africa experienced an extended wet phase over the Holocene 6000–9000 years before present during which time a large part of North Africa, including the Sahel, experienced a “greening” [Lenton, 2013]. The greater dust transport-deposition rates in the early Everglades record might be a consequence of the drying-out of large water bodies, a transition that would expose rich new dust sources [Prospero et al., 2002; Bullard et al., 2011].

Nutrients associated with African dust deposition have also been linked to toxic algal blooms in the Gulf of Mexico and the coastal environment of South Florida [Walsh et al., 2006; Lenos et al., 2012]. Dust deposition is associated with blooms of *Trichodesmium*. The subsequent enhanced production of nitrogen nutrients appears to stimulate blooms of *Karenia brevis* which result in the appearance of “red tides” which are typically followed by reports of respiratory difficulties in coastal populations along the west coast of Florida.

#### 4.2. Deposition to the Tropical Atlantic Ocean

The transport of dust to our receptor region should be viewed in the larger context of transport to the Atlantic. Dust is a major supplier of nitrogen species, phosphorous, and Fe all of which can play an important role in ocean productivity [Okin et al., 2011a]. Of these Fe is particularly important because in many ocean regions, primary productivity appears to be Fe limited. The Atlantic has been a major focus of study in an effort to understand the relationship between African dust transport and ocean biogeochemistry. To this end many oceanographic cruises have transected the region in an effort to characterize the temporal-spatial changes in aerosol properties and those of the underlying water column. Compilations of ship of aerosol measurements on ship transects yield distributions of dust-Fe [Buck et al., 2010; Baker et al., 2013] and nitrogen and phosphorus species [Baker et al., 2010] that are clearly linked to transport from Africa. The importance of this transport is reflected in the distribution of dissolved Fe which peaks in the tropical low latitudes. The gradient of dissolved Fe is particularly steep in equatorial waters [Ussher et al., 2013]. Dissolved Fe concentrations peak along the latitude of the winter-spring aerosol plume that impacts on Cayenne and along the northern boundary of the ITCZ which acts as an efficient removal mechanism. Thus, the ITCZ acts as a very effective barrier to the transport of dust to the South Atlantic [Adams et al., 2012; Huang et al., 2009, 2010]; as we might expect, Fe water concentrations decline sharply to the south.

Schlosser et al. [2013] present field evidence for the existence of distinct biogeochemical provinces of the North and South tropical Atlantic Ocean. They link seasonal shifts in these provinces to the seasonal shift in the dust plumes and removal in the ITCZ, processes that effectively modulate the distribution of diazotrophs and the associated nitrogen fixation. Thus, changes in dust transport and deposition could result in large changes in these biogeochemical provinces. Long-term measurements in Cayenne can provide a continuity that links the sporadic ship measurements made over the Atlantic and to field campaigns in Amazonia. Such studies could yield insights on the climate-related causes of transport variability, how it might have changed in the past, and how it might be linked to shifts in productivity in the equatorial region.

#### 4.3. African Dust and Air Quality

The link between air quality and health is largely based on studies in urbanized regions which often show a strong statistical relationship between various health metrics and the atmospheric concentration of species linked to human activities [Anderson, 2009]. Airborne PM has been specifically identified as a health factor, in particular, PM<sub>10</sub> which because of its size can efficiently penetrate the human respiratory system [Heal et al., 2012]. Soil dust could be a contributing factor to PM-related health issues [Morman and Plumlee, 2013; Plumlee et al., 2006; Goudie, 2014]. In recognition of the global scope of pollutants, the World Health Organization [2006] has established air quality guidelines (AQG) for ambient PM. However, these guidelines give little consideration to natural sources of PM and their health impacts. Consequently, monitoring efforts have focused on perceived problem regions proximate to major population centers and anthropogenic sources. To our knowledge, our study is the first to unambiguously document the persistent and systematic impact of long-range dust transport on air quality using standard instrumentation.

The annual rates of PM<sub>10</sub> exceedences at Cayenne and Guadeloupe (Table 1) are greater than those measured at many air quality sites in Europe [Putaud et al., 2010], most notably, sites in southern Europe and the Mediterranean Basin which, because of their proximity to North Africa, are frequently impacted by African dust events [Perez et al., 2008; Pey et al., 2013; Viana et al., 2014]. Studies of Asian dust (e.g., in Taipei [Chan and Ng, 2011] and western Japan [Kashima et al., 2012]) and African dust (e.g., in Madrid [Jiménez

*et al.*, 2010]) suggest that long-range dust has a quantifiable effect on health. However, the interpretation of these and similar studies is open to question because they are carried out in environments where, among other confounding factors, dust is mixed with high levels of pollutant species [Pey *et al.*, 2013; Viana *et al.*, 2014] which are themselves known to impact health [Karanasiou *et al.*, 2012]. Indeed, in general, it has not been possible to associate specific aerosol components, natural or anthropogenic, to specific health outcomes [Stanek *et al.*, 2011].

Thus, it is not possible to extrapolate the results of existing aerosol-health studies to remote pristine sites such as ours because of the absence of a coordinated PM monitoring program. However, individual studies have documented substantial African dust impacts in other areas of the Caribbean [Reid *et al.*, 2003; Prospero and Mayol-Bracero, 2013] and the southern United States [Perry *et al.*, 1997; Prospero, 1999] in some cases using PM<sub>10</sub> instrumentation [Prospero *et al.*, 2001; Bozlaker *et al.*, 2013]. Bozlaker *et al.* is of particular interest because it documents the PM<sub>10</sub> impact of a 3 km layer of African dust that in late July 2008 extended from Central America, across the Gulf of Mexico and Texas, and into northern Oklahoma and Arkansas.

There have been attempts to model the health impact of fine particles [Liu *et al.*, 2009; Evans *et al.*, 2013; Giannadaki *et al.*, 2013]. These suggest that dust is a prominent component in many areas and that dust could have a significant impact on mortality. However, such studies suffer from a number of problems. First, dust models are in an early stage of development; there are considerable differences among model products, most notably in emission estimates [Huneus *et al.*, 2011] and transport efficiency [Evan *et al.*, 2014; Kim *et al.*, 2014]. Thus, impact assessments will be very much model dependent. Second, while dust models include dust size distributions in their products [Mahowald *et al.*, 2013], there is very little data that can be used to test these products against long-range dust measurements [Schepanski *et al.*, 2009]. Third, as previously stated, PM health impacts are based largely on epidemiological studies carried out in urban-regional complexes. Dose-response relationships in such settings are probably not valid for assessing the possible health impacts of dust. To our knowledge, there are no data sets comparable to ours that shows the impact of long-range dust on air quality metrics in remote regions. While there have been many studies of dust impacts on health for specific environments and threats [e.g., Morman and Plumlee, 2013; Plumlee *et al.*, 2006], it is difficult to extrapolate these to global scales. Thus, there is a clear need for coordinated studies of long-range dust events in which the health-related aerosol properties are more carefully measured and documented.

## 5. Understanding the Dust Cycle and the Role of Modeling

We will only be able to assess global scale impacts of dust when we have models that can reliably describe the entire dust cycle starting with sources and ending at sinks. However, at this stage of development, models cannot consistently accomplish this goal. Given the prominence of African dust sources and the extensive data set from the Caribbean, these data have often been used to test dust models. In recent examples [Huneus *et al.*, 2011; Kim *et al.*, 2014; Evan *et al.*, 2014], global dust models were compared against various data sets including dust measurements from Barbados and Miami. Most models [Huneus *et al.*, 2011] captured the general seasonality of surface-level dust concentrations at these two sites although the predicted concentrations spanned a wide range. Removal processes, which must be crudely parameterized, also present a challenge to models [Kim *et al.*, 2014]. The most extensive dust deposition study was carried out in Florida [Prospero *et al.*, 2010] where 3 years of measurements were made at nine stations distributed over the length of the State. A comparison of nine models included in Huneus *et al.* [2011] showed that while most captured the broad features of the seasonal cycle, they produced a wide range of deposition estimates. This discrepancy is partly due to the problems in predicting concentrations and also the paths that individual dust events followed across the Atlantic. On a larger scale, models in general had difficulties in duplicating the strong seasonal shift in the African dust plume; in particular, the spring time shift to the low latitudes which we show here to be very strong. Efforts to reconcile various dust model products with satellite measurements [Yu *et al.*, 2013; Generoso *et al.*, 2008; Ridley *et al.*, 2012; Kim *et al.*, 2014] show broad agreement but also significant differences especially with regard to spring transport.

Model development will require a more complete understanding of the dust processes starting with those at the sources. There is a need for a better understanding of source terrains and the impact of various processes including land use and the effects of climate change [Ginoux *et al.*, 2012; Mahowald *et al.*, 2010]. Small-scale terrain features can strongly impact on the modeled emissions and their distributions [Lawrence and Neff, 2009;

*Bullard et al.*, 2011; *Heinold et al.*, 2011; *Okin et al.*, 2011b]. Even changes in model grid scale can result in substantial changes in model output [*Ridley et al.*, 2013, 2014]. As noted earlier in this section, the role of removal during transport is poorly understood, and there are few measurements in our receptor region that could quantify this process. The modeling of dust removal presents a number of challenges. Primary among them is the complex interaction that takes place between the African air outbreaks and the dynamic meteorological environment over the tropical Atlantic. Early studies based on Barbados [*Carlson and Benjamin*, 1980; *Carlson and Prospero*, 1972] suggested that African dust could play a strong role in these processes because of its impact on radiation and its consequent effect on the vertical distribution of atmospheric heating and the reduction of sea surface insolation [*Avellaneda et al.*, 2010]. Subsequent studies showed that dust effects could act through a complex series of atmosphere-ocean processes [*Evan et al.*, 2011, 2012; *Heinold et al.*, 2011]. Dust can also play a role in cloud microphysical processes where it can act as both condensation nuclei [*Twohy et al.*, 2009] and freezing nuclei [*Cziczo et al.*, 2013].

The complexity of these various processes is illustrated in a study of the interaction of the SAL with African Easterly Waves which are commonly associated with dust outbreaks in summer [*Zipser et al.*, 2009]. Because the main axis of the SAL lies on the northern edge of the ITCZ in all seasons, one would expect interactions with the ITCZ and the associated rainfall as noted above. There is evidence that dust in the SAL region can influence rainfall rates in the ITCZ [*Huang et al.*, 2009, 2010; *Wilcox et al.*, 2010]. Of particular interest is the possibility that African dust events can modulate tropical cyclone activity [*Dunion and Velden*, 2004; *Evan et al.*, 2011]. The interactions of the SAL with the ITCZ and tropical cyclones will also affect the deposition of dust and associated micronutrients to these tropical waters with consequent effects on marine biogeochemistry as noted above.

## 6. Conclusions

Our study shows that African dust is transported across the tropical North Atlantic in great quantities almost all year long and that this transport follows a well-defined seasonal pattern that can be largely linked to shifts in source activity in Africa. The dust carried in this transport is the major driver of PM<sub>10</sub> concentrations at our monitoring sites during their respective dust seasons when PM<sub>10</sub> often exceeds the WHO air quality guidelines. The impact of one specific source—the Bodélé Depression in Chad—is remarkable in that it demonstrates how one source could substantially affect air quality over a distance of 9000 km. Although these PM<sub>10</sub> concentrations are cause for concern, there is no evidence that they have actually had an impact on human health. In order to assess dust impacts, we will need a fuller understanding of the relationship between dust properties, concentrations, and health. The Caribbean might be a good place to study such relationships because air quality is relatively high as measured in terms of local and regional anthropogenic emissions as compared to continental sites in Europe and the United States. In the absence of many of the confounding factors found in urban environments, it might be possible to establish dose-response relationships for dust exposure.

On a global scale, dust could be a major factor in respiratory health. Over a third of the Earth's land surface is classified as semiarid to hyperarid [*United Nations Environment Programme*, 1997], large areas of which are major dust sources. Particularly notable is the band of aridity that extends from the west coast of North Africa, through the Middle East and Central Asia, and deep into China. Despite the widespread occurrence of dust throughout this "dust belt" and the large populations that are affected, there have been few studies of air quality metrics that focus on dust. Most relevant to our results are those of *Marticorena et al.* [2010] who measured daily PM<sub>10</sub> at three sites in West Africa located along 14°N (in Niger, Mali, and Senegal), a region which serves as a major source of dust that impacts our receptor sites. Measurements were made from January 2006 and December 2008, a period that overlaps our record. During the dusty season at the African sites (roughly November to June), daily PM<sub>10</sub> concentrations ranged almost constantly between 100 and 1000  $\mu\text{g m}^{-3}$ . Thus, over an 8 month period, the population is constantly exposed to PM<sub>10</sub> concentrations that greatly exceed the World Health Organization AQG. The African PM<sub>10</sub> concentrations are roughly a factor of 10 greater than those measured in Cayenne during its dust season. Despite such evidence, there have been few studies of health impacts in dust-rich natural environments—on a global scale, only a total of 50 papers [*de Longueville et al.*, 2013]. Of these, only 11 dealt with African dust, and almost all concerned with the impact on Europe. There is a clear need for focused studies in these regions.

Finally, over the longer term, there is concern about the possible impact of climate change on emissions, including dust [Monks *et al.*, 2009]. At Barbados, where the dust record begins in 1965 [Prospero and Lamb, 2003], concentrations increased greatly in the 1970s and early 1980s, linked to intense drought in Africa. Over the past two decades, African dust transport to the Atlantic has declined from the peak in the 1980s, a change that is further documented by satellite measurements since the late 1990s [Hsu *et al.*, 2012; Chin *et al.*, 2014]. However, climate modeling carried out in conjunction with the recent Intergovernmental Panel for Climate Change assessment [Seneviratne *et al.*, 2012] projects that large areas of North Africa along the northern and northwestern margins will become more arid in coming decades, a change that would likely lead to increased dust transport to the western Atlantic. But models could not agree on future trends in large areas of North Africa that are known to be home to some of the most active present-day dust sources, most notably the Sahel and particularly the Bodélé Depression. Thus, it is not possible to anticipate how climate change will affect PM levels over the Western Atlantic and what the consequent impacts on health or the environment might be. Studies have shown that North Africa is particularly responsive to changes in climate, serving as a “tipping point” indicator for such transitions [Lenton, 2013] with the Bodélé possibly playing an important role in the present-day context [Washington *et al.*, 2009]. In order to better anticipate the impacts of climate change in African dust emissions, we need an improved understanding of the entire dust cycle. Indeed, the magnitude and persistence of African dust emissions and the subsequent transport to the Caribbean Basin and South America argues that this is the ideal natural laboratory for the study of long-range dust source-receptor relationships.

## Author's Contributions

F-X. Collard and J. Molinie directed the programs that collected the data at Cayenne and Guadeloupe, assisted by A. Jeannot; all played a role in the analysis and interpretation of the data in the local and regional context. J.M. Prospero was responsible for the program in Barbados; he, in close collaboration with the coauthors, led in the interpretation of these data in the broader geographical and climate context.

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