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Atmospheric Fe deposition modes at Bermuda and the adjacent Sargasso Sea

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[1] The atmospheric fluxes of Fe and Al were determined from bulk (wet + dry) plus wet only deposition and aerosol samples collected at Bermuda from 1999 to 2001. During the sampling period, dry deposition accounted for majority (>70%) of total Fe deposition estimated by both modeled and measured means. This contrasts the North Pacific, where generally wet dominates dry deposition. This may be caused by the size and proximity of continental dust sources and seasonal patterns of precipitation to the North Atlantic. Comparing time series measurement of wet soluble Fe deposition indicates that the North Atlantic Oscillation (NAO) may impact the deposition mode of Fe in these areas. The dry deposition velocities of Fe-bearing aerosols (DDV_{Fe}) were estimated from paired averages of measured dry deposition (bulk-wet fluxes) divided by corresponding aerosol concentrations, and a seasonal trend was observed. During the summer months of 1999, the DDV_{Fe} ranged from 0.1 to 0.3 $cm\ s^{-1}$; yet, during the fall and winter months, the DDV_{Fe} ranged from 2.0 to 6.0 $cm\ s^{-1}$. It is hypothesized that with increased humidity and wind speeds at Bermuda during fall and winter, mineral aerosol particles could be internally mixed with sea salt and gaseous S and N pollutants to increase effective particle sizes. Such marine particle aggregation is reported elsewhere and could seasonally increase the apparent dry deposition velocity of mineral aerosols.

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Index Terms: 0312 Atmospheric Composition and Structure: Air/sea constituent fluxes (3339, 4504); 1616 Global Change: Climate variability (1635, 3305, 3309, 4215, 4513); 4906 Paleoclimatology: Aerosols (0305, 4801).

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1. Introduction

[2] Atmospheric transport is the main pathway for the introduction of many trace elements to the surface waters of the open ocean [Prospero, 1996; Martin *et al.*, 1989; Jickells, 1999; Arimoto, 2001]. The atmospheric input of lithogenic elements, especially Fe, has important implication for the nutrient demands in marine surface ecosystems [Duce *et al.*, 1991; Duce and Tindale, 1991; Jickells, 1995; Jickells *et al.*, 2005]. Bermuda is located in the northwest quadrant of the Sargasso Sea and provides a good locale for atmospheric deposition of both anthropogenic and natural aerosols to the North Atlantic ocean [Chen and Duce, 1983; Arimoto *et al.*, 1992, 1995; Sedwick *et al.*, 2005].

[3] Lithogenic aerosols transported to Bermuda and deposited into surface waters originate from the surrounding continental region after transport by seasonal wind systems [Arimoto *et al.*, 1992, 1995; Tindale and Pease, 1999]. The types and quantities of aerosols transported are strongly dependent on dust sources, transport pathways and meteorological conditions of the seasons [Chen and Duce, 1983; Arimoto *et al.*, 2003]. Generally, the seasons at Bermuda are defined as follows: March–May as Spring, June–August as Summer, September–November as Fall and December–February as Winter [Huang *et al.*, 1999]. Significant events of dust at Bermuda originate from North Africa after clockwise transport on trade winds from SE to SW around the subtropical gyre [Chen and Duce, 1983; Arimoto *et al.*, 1992, 1995]. These dust episodes usually occur between May and September because of the northward shift of the Intertropical Convergence Zone (ITCZ) and intensification of Bermuda-Azores high-pressure systems [Arimoto *et al.*, 1995; Jickells, 1999; Church *et al.*, 2000]. From late fall to early spring months, the air parcel trajectories to the Bermuda region occur most frequently from regions of North America [Chen and Duce, 1983; Arimoto *et al.*, 1995; Huang *et al.*, 1999; Arimoto, 2001]. The Saharan dust transport then becomes relatively minor as the Bermuda-Azores high-pressure system begins to migrate southeastward from late fall through winter [Moulin *et al.*, 1997]. Also, during the winter months, stronger surface winds can raise sea salt concentrations in Bermuda aerosol samples [Woodcock, 1953; Arimoto *et al.*, 2003].

[4] Aerosol metals including Fe can reach the sea surface following two major pathways: (1) dry

deposition (settling or impact of atmospheric aerosols or aggregates) and (2) wet deposition (scavenging involving an atmospheric aqueous phase) [Zhuang *et al.*, 1992; Jickells and Spokes, 2001; Gao *et al.*, 2003]. Only certain inorganic and organic forms of dissolved atmospheric Fe can be assimilated by phytoplankton [Hutchins *et al.*, 1999; Achilles *et al.*, 2003]. Important is the solubility of dry deposited lithogenic Fe or resulting from wet scavenging that is subsequently delivered to seawater as nutrient for oceanic biogeochemistry [Duce and Tindale, 1991; Jickells and Spokes, 2001; Hand *et al.*, 2004; Luo *et al.*, 2005]. In this respect, processes associated with the wet and dry aerosol deposition mode could be biogeochemically distinct [DiTullio *et al.*, 1993; Chester *et al.*, 1997; Wu *et al.*, 2001; Chen and Siefert, 2004]. Because of relatively low atmospheric pH and organic constituents, trace metals in rainwater may be more labile when deposited to the sea surface [Church *et al.*, 1992; Spokes *et al.*, 1994; Desboeufs *et al.*, 1999, 2005].

[5] Previous studies indicate that dry Fe deposition accounted for the majority (>70%) of total atmospheric deposition of Fe at Bermuda, based on comparison between dry Fe deposition estimated from measured aerosol concentration and wet Fe deposition derived from analysis of rainwater samples [Duce *et al.*, 1991; Jickells *et al.*, 1998]. However, more recent measurements of lithogenic elements in both wet and bulk (wet + dry) deposition samples, collected at Bermuda from 1996 to 1998, suggested that wet Fe deposition could account for around half of total Fe deposition [Kim and Church, 2001]. These differences may be due to a change in the primary mode of Fe deposition (wet versus dry) and indicate the need for further time series investigations of modes and seasonal sources of mineral aerosol deposition at Bermuda between different years.

[6] Continental dust particles enter the marine atmosphere and, in combination with sea salt, can undergo significant modification by pollutant N and S species before and during their dry and wet removal [Zhang and Iwasaka, 2004]. Here, they may rapidly become mixtures of mineral, sea salt, sulfate and/or nitrate [Andreae *et al.*, 1986; Okada *et al.*, 1990; Fan *et al.*, 1996; Zhou *et al.*, 1996; Niimura *et al.*, 1998; Zhang *et al.*, 2003]. These processes can cause changes in composition and particle size affecting their dry deposition velocities used to calculate atmospheric dry fluxes.

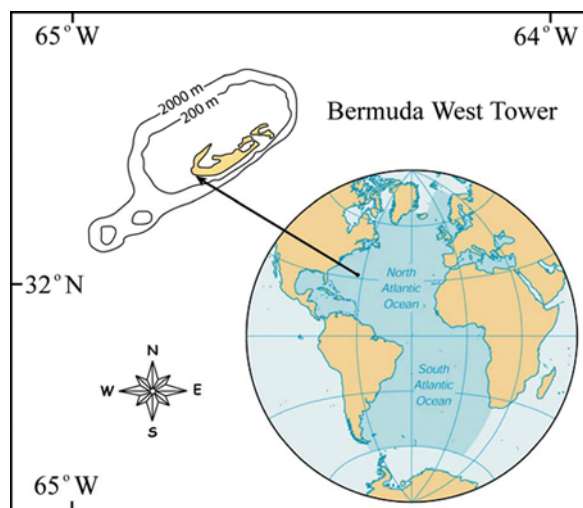


Figure 1. Location is shown for the atmospheric sampling of aerosol, wet, and bulk deposition. The Bermuda West Tower was erected as part of the AEROCE program in 1988–1997, located at Tudor Hill (32.27°N, 64.87°W) on the southwest coast of Bermuda.

[7] In this paper, aerosol, wet deposition and bulk deposition samples were collected synchronously over seasonal time periods during 1999–2001 and used to characterize the transport and deposition modes of Fe at Bermuda. It is proposed that the dry deposition velocity of Fe-bearing aerosols can be evaluated by using such atmospheric data and used to reevaluate dry Fe deposition at Bermuda.

2. Sample Collection and Analytical Methods

2.1. Sampling

[8] Thirty nine aerosol and fifteen atmospheric deposition (wet and bulk) samples were synchronously collected on the Bermuda West Tower (BWT) at Tudor Hill Bermuda (32.27°N, 64.87°W; Figure 1) from July 1999 to February 2001. The 23 m tall anodized aluminum tower was erected as part of the AEROCE program in 1988–1997 and stands 50 m above sea level. The wet and bulk deposition samples were collected in duplicate using 2 L wide mouth fluorinated polyethylene bottles that also served for both stored acidification and digestion vessel [Kim *et al.*, 1999].

[9] The aerosol sample collections were controlled by an automated system, which permitted collection according to onshore wind direction and speed, to avoid contamination from local sources [Arimoto *et al.*, 1992]. Aerosol samples were

collected using 47 mm acid-cleaned Millipore type HA filters mounted inverted in rain shields fashioned from the same wide-mouth 2 L bottles that may discriminate against very large (>10 micron) particles [Véron *et al.*, 1992]. The aerosol filters were unloaded in a Class 100 clean bench and stored frozen in acid-cleaned plastic petri dishes for later subsampling, processing and analysis.

[10] Deposition samples were collected over monthly intervals using both open bulk and automated wet-only collectors [Kim *et al.*, 1999]. Precipitation depth was measured by a rain gauge at the tower capped with Si oil to minimize evaporation. All samples were stored frozen in the acid-cleaned sampling bottles until processing.

2.2. Analytical Methods

[11] All the labware used in the experiments were rigorously acid cleaned in a Class-100 clean room. Aerosol samples were digested in Teflon vials using subboiled HNO₃ (70%) and HF (48%, Fisher Scientific trace metal grade) in a ratio of 3:1 by volume. Either 1/2 or 1/4 of the aerosol filter was used for acid digestion on the basis of whether the visual appearance of the aerosol was light or dark, respectively. After complete digestion, the residue was redissolved into 5% HNO₃.

[12] Both bulk and wet deposition samples were completely digested in the same wide mouth fluorinated polyethylene bottles as the collection using a mixture of distilled HNO₃-HCl-HF (3:2:1 by volume), then redissolved into 5% HNO₃. The concentrations of trace metals (Fe, Al) were analyzed by a Graphite Furnace Atomic Adsorption Spectrometer (GF-AAS, Perkin Elmer 3300) using multielement calibration standards (SCP Science) in a 5% HNO₃ matrix.

[13] Wet deposition samples were also acidified (0.4% HCl v/v) with subboiled HCl, leaching for at least one month, and then analyzed by GF-AAS in this matrix to test the leaching efficiency at pH 2. The leaching method has been widely adopted in past studies, and this portion of Fe in wet deposition is defined as total dissolvable Fe [Véron *et al.*, 1992; Kim *et al.*, 1999].

[14] Blanks were run to estimate the level of potential contamination introduced by the lab operation procedure. All lab blanks were lower than the detection limit of GF-AAS for Fe (5 pg, <5% of lowest concentrate sample) and Al (10 pg, <2% of lowest concentrate sample). Compared to the low-

Table 1. Total and Wet Fe and Al Fluxes at Bermuda Estimated From Bulk and Wet Deposition Samples ($\pm 5\%$ for Both Fe and Al)^a

Sampling Date	Fe Flux ($\mu\text{g m}^{-2} \text{d}^{-1}$)			Al Flux ($\mu\text{g m}^{-2} \text{d}^{-1}$)		
	Bulk	Wet (Total)	Wet (Dissolvable)	Bulk	Wet (Total)	Wet (Dissolvable)
5 Jul 1999 to 2 Aug 1999	63.8	55.6	19.6	124	82.2	43.6
2 Aug 1999 to 7 Sep 1999	140	118	35.9	295	197	62.8
7 Sep 1999 to 28 Sep 1999	673	82.4	21.7	1060	172	41.3
28 Sep 1999 to 25 Oct 1999	349	109	22.1	566	151	34.8
25 Oct 1999 to 24 Nov 1999	39.3	15.7	4.55	95.8	17.3	6.91
24 Nov 1999 to 20 Dec 1999	91.5	62.0	40.9	608	68.7	41.5
20 Dec 1999 to 20 Jan 2000	61.5	14.9	1.18	94.4	17.7	6.58
20 Jan 2000 to 18 Feb 2000	46.4	18.4	10.1	88.1	30.9	19.7
18 Feb 2000 to 14 Mar 2000	133	18.0	14.8	247	36.6	17.6
14 Mar 2000 to 14 Apr 2000	71.6	64.8	19.6	164	90.2	35.9
19 Sep 2000 to 12 Oct 2000	541	204	16.8	474	362	44.7
12 Oct 2000 to 10 Nov 2000	163	44.7	10.4	250	57.7	22.0
10 Nov 2000 to 8 Dec 2000	102	13.7	3.58	235	93.4	29.5
8 Dec 2000 to 18 Jan 2001	43.4	78.2	5.79	74.7	46.7	12.4
25 Jan 2001 to 15 Feb 2001	61.1	9.9	3.55	255	21.0	11.2

^aTotal means measured Fe concentration in wet deposition samples after total digestion. Dissolvable means measured Fe concentration in wet deposition samples after leaching with 0.4% HCl for 1 year.

est loading of any aerosol sample, the field blanks were always less than 10% for both elements. A lab standard (SCP Science) was used to check the accuracy of the analyses. The external reproducibility, including both chemical preparation and GF-AAS measurements, was performed on replicate samples resulting in precisions of less than 5%.

3. Results

[15] Total and wet Fe and Al fluxes were estimated by analyzing bulk and wet deposition samples (Table 1). The measured dry Fe and Al fluxes were derived from the difference between total and wet Fe and Al fluxes respectively, while the calculated dry Fe fluxes were estimated from measured Fe relative mass concentrations in the aerosol samples times a model-derived dry deposition velocity [Jickells *et al.*, 1987; Duce *et al.*, 1991] (Table 2). This study used a mean deposition velocity of 1.0 cm s^{-1} suggested by comparing dust deposition estimates to sediment trap records [Jickells, 1999].

[16] The leached recovery using 0.4% HCl (total dissolvable Fe) for wet deposition samples (Table 1) were below 30%, and only three (winter/early spring 1999) showed major leaching ($>50\%$). This is in variance with the major recovery of Fe ($>70\%$), also using 0.4% HCl in a previous annual set of wet deposition samples during 1996–1997 [Kim *et al.*, 1999]. This could be due either to the less efficient processing of some unleached par-

ticles, or seasonal variance in the composition of materials deposited by the wet deposition from year to year. As will be discussed, it appears that in those years when the North Atlantic Oscillation relaxes (negative NAO), more North African dust is wet scavenged during the summer months.

[17] Overall, the calculated dry Fe fluxes during our sampling period were comparable to previous results at the BWT and the Bermuda Testbed Mooring (BTM) in the nearby Sargasso Sea, which averaged on the order of $100 \mu\text{g m}^{-2} \text{d}^{-1}$ during the summer/fall months over the past several decades [Duce *et al.*, 1976; Arimoto *et al.*, 2003; Sholkovitz and Sedwick, 2006]. Previous studies of wet Fe deposition at Bermuda also using 0.4% HCl, showed the wet Fe fluxes (total dissolvable) at Bermuda and the nearby Sargasso Sea in the past decades were around $50 \mu\text{g m}^{-2} \text{d}^{-1}$ [Véron *et al.*, 1992; Church and Véron, 1993; Kim *et al.*, 1999; A. Véron, unpublished data, 1982–1998]. Thus over the long term of the past three decades, Fe deposition to the Bermuda region of the Sargasso Sea appears to be on average a third wet dissolvable and two thirds dry.

[18] The Fe fluxes (calculated dry, total digested wet and bulk) were the highest during the summer/fall months as reported elsewhere over this sampling period, although total dissolvable Fe in wet deposition did not show any seasonal trend (Figure 2). The calculated dry Fe deposition showed several dust periods during the summer/fall months, which

Table 2. Calculated Dry Fe and Al Fluxes at Bermuda^a

Sampling Date	Fe Flux ($\mu\text{g m}^{-2} \text{d}^{-1}$)	Al Flux ($\mu\text{g m}^{-2} \text{d}^{-1}$)	Air Volume (m^3)
29 Jun 1999 to 5 Jul 1999	1710	2780	36
5 Jul 1999 to 12 Jul 1999	260	273	25
12 Jul 1999 to 20 Jul 1999	24.9	30.3	121
20 Jul 1999 to 27 Jul 1999	109	165	272
27 Jul 1999 to 2 Aug 1999	111	179	255
2 Aug 1999 to 16 Aug 1999	154	250	212
16 Aug 1999 to 23 Aug 1999	32.6	52.3	235
23 Aug 1999 to 31 Aug 1999	21.6	3.12	145
31 Aug 1999 to 7 Sep 1999	13.9	22.8	207
13 Sep 1999 to 22 Sep 1999	186	335	68
22 Sep 1999 to 28 Sep 1999	33.1	68.9	75
28 Sep 1999 to 4 Oct 1999	3.09	5.66	92
14 Oct 1999 to 11 Oct 1999	6.06	5.05	47
11 Oct 1999 to 18 Oct 1999	8.18	10.3	293
18 Oct 1999 to 29 Oct 1999	117	185	174
1 Nov 1999 to 10 Nov 1999	16.4	25.6	116
10 Nov 1999 to 15 Nov 1999	11.8	15.2	227
15 Nov 1999 to 24 Nov 1999	9.70	14.5	104
24 Nov 1999 to 29 Nov 1999	11.6	46.8	16
29 Nov 1999 to 6 Dec 1999	12.7	20.4	171
6 Mar 2000 to 14 Mar 2000	108	107	10
24 Mar 2000 to 6 Apr 2000	13.1	23.7	2
6 Apr 2000 to 14 Apr 2000	106	85.1	3
28 Apr 2000 to 5 May 2000	31.1	39.6	22
19 Sep 2000 to 26 Sep 2000	520	956	182
26 Sep 2000 to 4 Oct 2000	36.6	74.7	182
4 Oct 2000 to 10 Oct 2000	18.3	27.0	80
19 Oct 2000 to 25 Oct 2000	8.19	28.6	10
25 Oct 2000 to 1 Nov 2000	103	32.5	200
1 Nov 2000 to 10 Nov 2000	6.20	8.99	276
10 Nov 2000 to 17 Nov 2000	5.51	6.38	210
17 Nov 2000 to 24 Nov 2000	13.5	18.4	236
24 Nov 2000 to 29 Nov 2000	39.0	12.4	139
29 Nov 2000 to 8 Dec 2000	7.19	7.71	229
8 Dec 2000 to 14 Dec 2000	10.9	15.9	76
14 Dec 2000 to 19 Dec 2000	17.2	20.4	57
19 Dec 2000 to 30 Dec 2000	5.86	8.06	219
30 Dec 2000 to 3 Jan 2001	4.03	5.15	185
3 Jan 2001 to 18 Jan 2001	8.94	11.4	345

^a See text for details. The total external reproducibility was less than 5% for both Fe and Al.

accounted for a majority (~60%) of the calculated dry Fe deposition at Bermuda (1999–2001).

[19] To identify the sources of Fe to Bermuda, Al was used as a reference for crustal derived elements [Taylor and McLennan, 1985]. Most Fe/Al ratios in aerosol samples were close to the reported average Fe/Al ratio (0.63) in Saharan dust, especially during the high dust season [Chen and Duce, 1983; Arimoto *et al.*, 1992, 1995; Guieu *et al.*, 2002; Prospero, 2002] (Figure 3). This indicates that most of the Fe and Al deposited at Bermuda come from similar lithogenic sources in North Africa and America. However, there were several weekly episodes that exceed this crustal ratio by a factor 2–6, suggesting possible noncrustal sources

of background atmospheric contaminants [Sedwick *et al.*, 2007]. There are also a few episodes of Fe/Al ratios significantly below the crustal average that (in the absence of tower contamination) could be related to other fractionated sources, including surface microlayer resuspension [Arimoto *et al.*, 1997]. These possible noncrustal sources and their implication are discussed next.

4. Discussion

4.1. Atmospheric Deposition at Bermuda

[20] It has been well documented that mineral dust deposition at Bermuda generally shows a maximum during summer months and minimum during

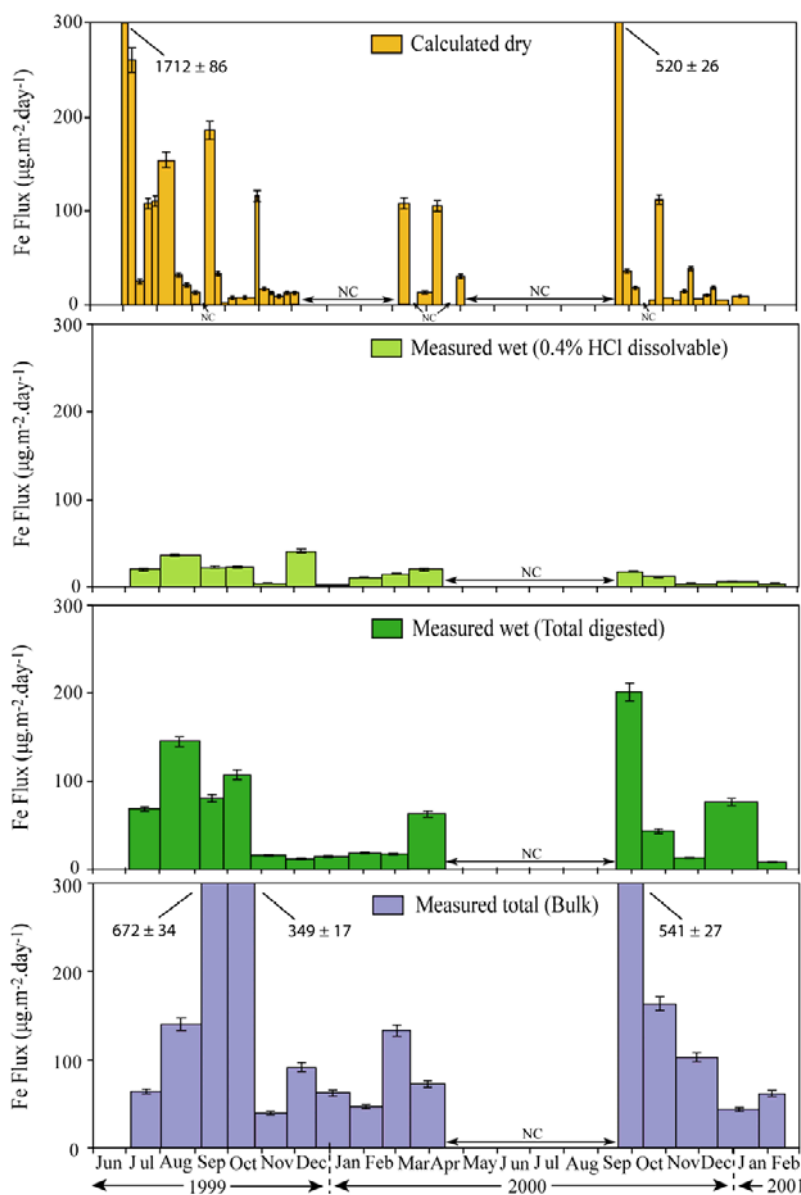


Figure 2. The temporal variation of Fe fluxes reflected by (a) aerosol samples, (b) wet deposition in 0.4% HCl leached (total dissolvable), (c) wet deposition (total digested), and (d) bulk (measured total) deposition collected at the Bermuda West Tower from July 1999 to January 2001. NC means no samples were collected during those periods.

winter/spring months [Chen and Duce, 1983; Arimoto *et al.*, 1995; Jickells, 1999; Arimoto, 2001]. However, dust transport at Bermuda actually is more complicated because of transitional trade winds, especially during the spring/fall period [Anderson *et al.*, 1996; Huang *et al.*, 1999]. Lithogenic elements deposited on the trades are typically from North Africa, but North America nearby can also provide aerosols frequently mixed with anthropogenic particles, often containing soluble Fe, Mn and V from industries using fuel oil in the N.E. United States [Rahn, 1981; Desboeufs *et al.*, 2005].

[21] As noted in Figures 3a and 4, the weekly Fe/Al weight ratios in aerosols showed relatively high Fe/Al weight ratios particularly during low Fe loaded samples, mainly during the spring and fall. Such low iron loaded aerosols can originate from North American air masses typically associated with elevated Fe/Al mass ratios indicative of anthropogenic fuel combustion products [Chen and Duce, 1983; Arimoto *et al.*, 1995]. Such mineral aerosols potentially have high solubility that could conceivably account for a significant fraction of the aeolian flux of “soluble iron” to open ocean

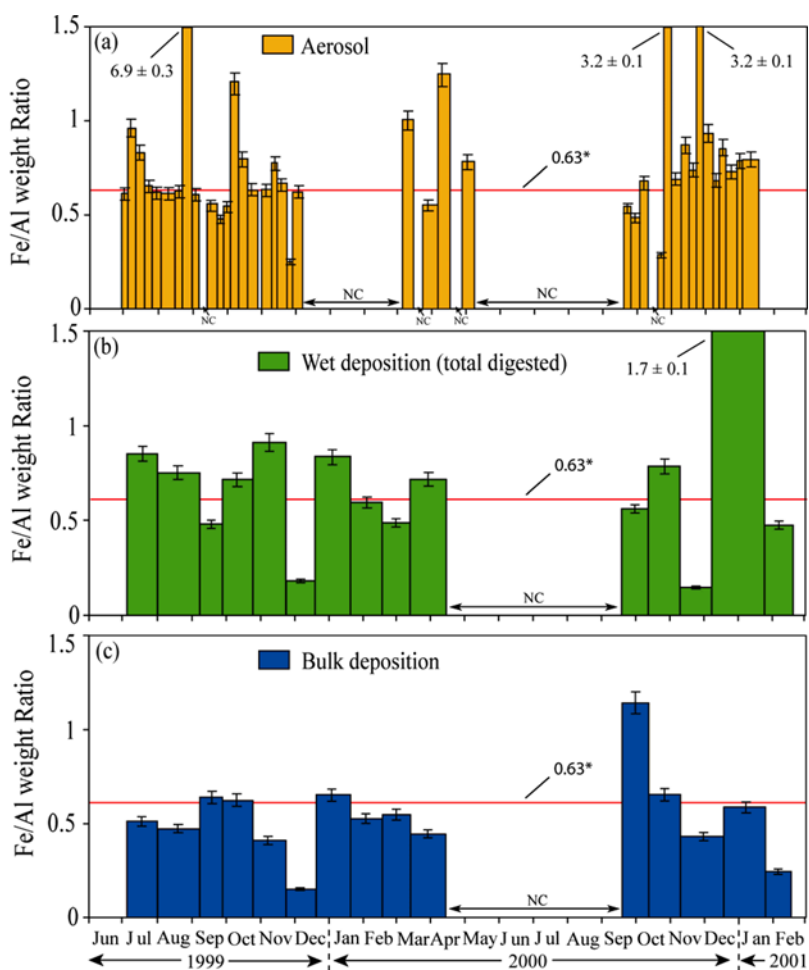


Figure 3. The Fe/Al weight ratios in (a) aerosols, (b) wet deposition (total digested with HNO_3 , HCl , and HF), and (c) bulk deposition samples, collected at the Bermuda Sampling Tower from July 1999 to February 2001. NC denotes when no samples were collected, and the line marked at 0.63* indicates the average Fe/Al ratio in Saharan dust [Arimoto *et al.*, 1995; Guieu *et al.*, 2002].

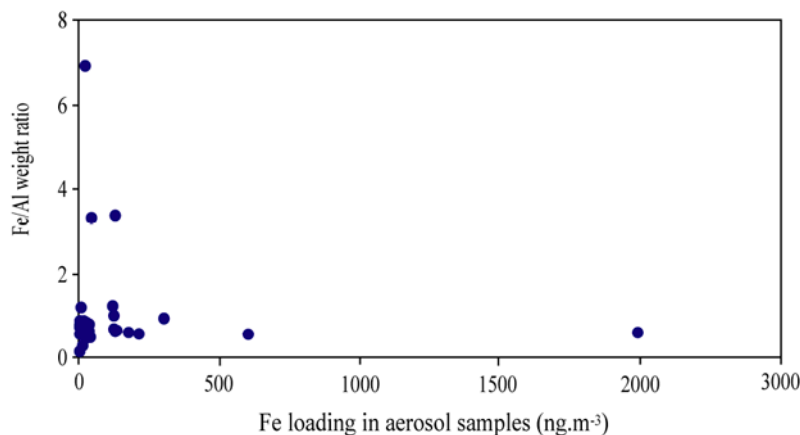


Figure 4. The Fe/Al weight ratios versus Fe loading (ng m^{-3}) in aerosol samples collected at the Bermuda West Tower from July 1999 to February 2001.

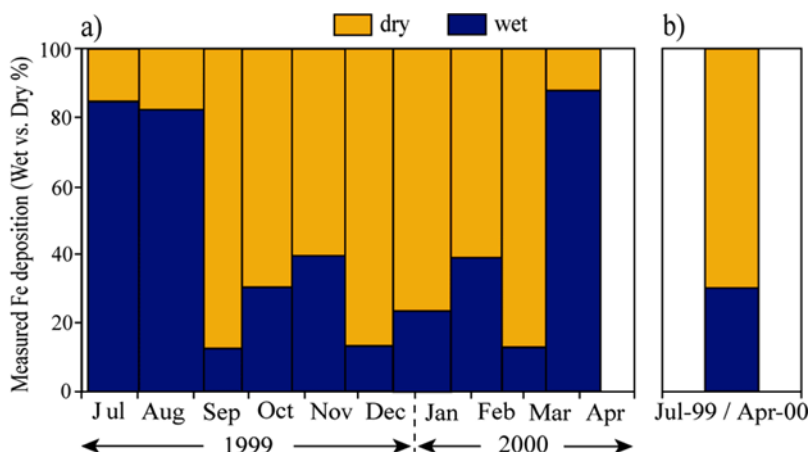


Figure 5. The Fe deposition mode (wet versus dry) at Bermuda for the sampling period from July 1999 to April 2000 (a) for individual sampling periods and (b) integrated over the entire sampling period.

regions, contributing potentially to the production of oceanic biota and subsequent carbon cycling [Sedwick *et al.*, 2007].

4.1.1. Wet Deposition

[22] The magnitude of wet-deposition fluxes is highly uncertain, owing to a lack of extensive field data and the patchy or episodic nature of rainfall over the open ocean [Jickells and Spokes, 2001; Jickells *et al.*, 2005; Mahowald *et al.*, 2005; Sedwick *et al.*, 2005]. In this study, total dissolvable Fe (TDFe, soluble at pH 2 (0.4% HCl) in the wet deposition samples was compared to the total wet deposition and shown on average lower than 30% (Figure 2b). This infers that a significant portion of total Fe in the rain is likely bound in aluminosilicate structures and not readily available for biological utilization. However, there were three sample periods in late fall and winter 1999, when the fractions of TDFe were as high as 80% of total wet Fe deposition, as was the case in a previous period at Bermuda, 1996–1997 [Kim and Church, 2001]. As discussed, some aerosols characterized as a mixture of soil dust and anthropogenic aerosols have relatively higher fractional solubility, although not necessarily when scavenged by rainwater [Sedwick *et al.*, 2007]. Nevertheless, when these anthropogenic materials are scavenged by the rain, they may provide some of the observed surface TDFe to surface seawater as biologically available Fe.

4.1.2. Dry Deposition

[23] The empirical dry Fe deposition in this study was estimated from the measured difference between the total bulk Fe deposition and total wet Fe

deposition. Overall, the measured dry Fe deposition accounted for the majority (~70%) of total Fe atmospheric deposition to Bermuda for most of the annual period from July 1999 to April 2000 (Figure 5). This agrees with published data for 1988–1994 [Arimoto *et al.*, 1995; Church *et al.*, 2006]. However, wet deposition accounted for almost half of total Fe deposition based on similar bulk and wet collections during the 1996–1997 period at Bermuda [Kim and Church, 2001]. This might suggest a significant change in deposition mode of Fe, especially during summer when relatively high rainfall and dust events are coincident.

4.2. Climatological Controls on Deposition by the North Atlantic Oscillation

[24] It has been suggested that precipitation amount and types of aerosols can significantly affect Fe deposition mode (wet versus dry) at Bermuda [Kim and Church, 2001; Sedwick *et al.*, 2007]. The North Atlantic Oscillation index (NAO) has often been evoked to constrain dust loading and deposition over the subtropical North Atlantic, as well as in model climate simulations [Moulin *et al.*, 1997; Chiapello and Moulin, 2002; Ginoux *et al.*, 2004; Scaife *et al.*, 2005]. The NAO index is suggested to have such a significant impact on atmospheric transport since it reflects the dynamics of wind regimes [Rogers, 1984; Hurrell, 1995; Hurrell and Van Loon, 1997]. By comparing the 15 year interannual variability of dust distribution and emission with the NAO index, it has been shown that the winter surface dust concentration is correlated with the NAO index over much of the North Atlantic [Ginoux *et al.*, 2004]. It is observed that the high NAO indices are clearly correlated to significant dust outbreaks and export from Africa.

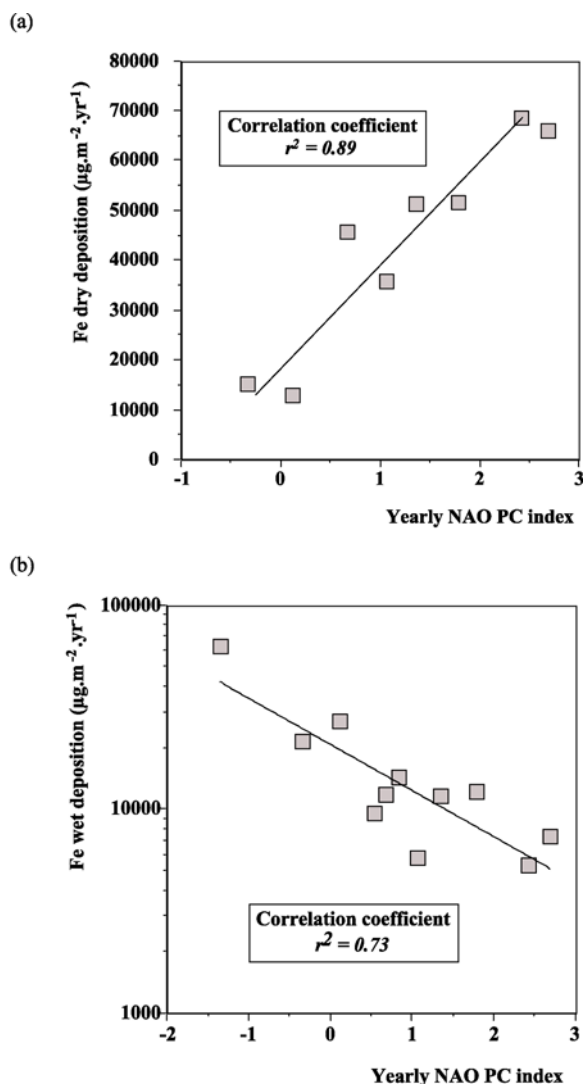


Figure 6. Relation between the long-term (a) dry Fe deposition and (b) wet deposition to Bermuda versus the annual NAO-PC index between 1988 and 1998 [Hurrell, 1995; Hurrell et al., 2003].

Furthermore, the 15-year simulation of global dust distribution in the North Atlantic atmosphere (GOCART model) and the NAO index significantly covary [Ginoux et al., 2004]. However, when considering the multidecadal records of dust loading at Barbados [Prospero, 1999], the NAO does not appear to control dust input [Ginoux et al., 2004]. This might reflect more direct downwind transport pathways in the subtropical and tropical North Atlantic regions.

[25] The unique long-term record complete for both dry and wet deposition to Bermuda collected during WATOX (1982–1985), AEROCE (1989–1994) and other (1996–1998) programs can be used to investigate the role of this meteorological

index on Fe deposition [Church and Véron, 1993; Kim et al., 1999]. The annual NAO PC (Principal Component) index is calculated from PC time series of the leading EOF (Empirical Orthogonal Function) for annual sea level pressure anomalies over the Atlantic sector (20–80°N, 90°W–40°E). This annual index appears to better represent the spatial patterns associated with wind regimes and pressure systems over the North Atlantic [Hurrell, 1995; Hurrell et al., 2003]. The Fe dry deposition to Bermuda is inferred from aerosol data and wet/bulk sampling [Arimoto et al., 1992, 2003; Kim et al., 1999; Kim and Church, 2001] for the period 1988 to 1998. These long-term dry fluxes are closely correlated to the NAO PC index ($r^2 = 0.89$, $p < 0.0001$), with dry deposition being larger during high NAO periods (Figure 6a) as could be expected from previous models [Moulin et al., 1997; Ginoux et al., 2004].

[26] The current and earlier findings show that wet deposition could account for significant Fe input to Bermuda, suggesting a need to investigate the long-term record for Fe wet deposition. A significant yet negative relationship ($r^2 = 0.73$, $p = 0.0001$) is found between long-term annual Fe wet deposition and NAO PC (Figure 6b) with higher Fe wet fluxes during low NAO index periods. These findings could simply reflect larger rain amounts deposited to Bermuda associated with the origin of North American frontal systems during low NAO modes. Indeed, rain amounts during the WATOX and AEROCE programs are partly controlled by variation of the annual NAO PC ($r^2 = 0.35$). However, this low correlation coefficient can only explain a small fraction of Fe wet deposition variability. Further it suggests that the negative effect of NAO on wet deposition at Bermuda is primarily climatologic and secondarily wet scavenging dependent.

[27] Recent findings in northern Caribbean corals are consistent with the correlation found between the NAO index and Fe wet deposition on Bermuda [Desenfant et al., 2006]. A significant correlation is found between lead isotope imprints in these annually banded corals and NAO indices for the period 1965–1997 when trade easterly flows mostly occur during low NAO episodes. These results substantiate the intricacy in modeling wet deposition and strengthen the need for long-term, station based collection of coupled rain and dry deposition. This is particularly important when assessing the input of nutrient limiting elements and their bioavailability in sea surface waters.

[28] It is hypothesized that significant Fe wet deposition during low NAO episodes could reflect the combination of more effective (if less intense) dust transport to Bermuda associated with more wet summer scavenging. Indeed, during high NAO episodes, strong easterly flows are obstructed southward, while low NAO allows the northward transport of less intense dust plumes more efficiently scavenged under the prevalent moisture evident in westerly frontal systems. Therefore, while low NAO might not allow the transport of strong dust plumes to Bermuda, it permits more Fe wet deposition that could act as a more effective summer source of bioavailable Fe to marine surface organisms. As likely, the NAO change could simply change the amount and nature of summer rain on Bermuda. Basically, more rain would mean more wet deposition of iron in a dust limiting system.

[29] Equally important is the relative interannual wet deposition in delivering nutrient Fe scavenged from mineral aerosol, hence the need for assessing both wet and dry trace element deposition. Such evidence is needed for a complete investigation of both transport pathways (the strength of dust outbreak and easterly flows) and scavenging processes that control wet and dry deposition to explain trace element input to the North Atlantic.

[30] The limited data in space and time indicate that on a global basis wet deposition of mineral aerosol Fe may dominate over dry deposition [Duce *et al.*, 1991]. In the Sea-Air Exchange Program (SEAREX) Pacific stations, the total wet deposition of mineral matter apparently accounted for 80% of total deposition [Uematsu *et al.*, 1985]. On the basis of model simulation, wet deposition of mineral dust may exceed dry deposition by up to a factor of 10 over the transpacific transport path [Zhao *et al.*, 2003]. Here total wet deposition is a function of precipitation and the major process of Asian soil dust removal from the atmosphere to the North Pacific Ocean. However, at Bermuda and the nearby Sargasso Sea, dry deposition appears to dominate much of the time, possibly because of the relatively higher concentrations of coarse mode aerosol and their proximate continental source [Duce *et al.*, 1991; Jickells *et al.*, 1998]. For example, recent measurements during the PRIDE experiment show supermicron (typically 3.5 μm) particles for dust that has traveled from North Africa over the Caribbean [Reid *et al.*, 2003; Grini and Zender, 2004]. This could indicate effective trade easterly advection of course mode mineral

aerosol, or particle growth in the moist marine environment as elaborated in the following section.

4.3. Dry Deposition Velocity

[31] Following is the approach of Duce *et al.* [1991], based on a report prepared by the *Group of Experts on the Scientific Aspects of Marine Pollution* [1989]. Here, aerosol dry deposition velocity (DDV) has been estimated according to the sizes of the major aerosol types: $V_d = 0.1 \text{ cm s}^{-1}$ (\pm a factor of 3) for submicrometer particles, $V_d = 1.0 \text{ cm s}^{-1}$ (\pm a factor of 3) for mineral dust and other supermicrometer particles not associated with sea salt, and $V_d = 3.0 \text{ cm s}^{-1}$ (\pm a factor of 2) for large sea-salt-associated particles.

[32] More refined methods for calculating dry deposition would also be problematic in the absence of supporting meteorological data. Usually for mineral aerosols, dry deposition velocity is assumed to be between 0.3 and 3 cm s^{-1} [Arimoto and Duce, 1986; Prospero, 1996]. Thus, there are large uncertainties when using a single value of 1.0 cm s^{-1} dry deposition velocity times the measured concentrations in aerosol samples to calculate the dry flux of lithogenic elements (e.g., Fe and Al). Because of the difficulty in directly measuring aerosol dry deposition or its velocity, this model derived DDV is still used extensively in estimating dry deposition [Arimoto *et al.*, 1992, 1995].

[33] Dry deposition velocity for Fe-bearing aerosols were estimated using data from the eight integral periods when bulk and wet deposition plus aerosol sampling approximately matched over several weeks (Figure 7). The DDV of Fe-bearing aerosols (DDV_{Fe}) can be estimated from the equation as follows:

$$\text{DDV}_{\text{Fe}} = \text{DF}/\text{MC}_{\text{Fe}} \quad (1)$$

where DF is the dry Fe flux ($\mu\text{g m}^{-2} \text{ d}^{-1}$), calculated from the difference between the measured total Fe flux and wet Fe flux, and MC_{Fe} is the measured Fe concentration in aerosol samples (ng m^{-3}).

[34] A seasonal variation of DDV_{Fe} is suggested at Bermuda when the DDV_{Fe} from the integral summer months in 1999 were <1 (0.1 and 0.3 cm s^{-1} , respectively), but ranging 2 to 6 cm s^{-1} during fall and winter months (Figure 7). Thus using a common value of 1.0 cm s^{-1} to approximate the dry deposition velocity and calculate dry Fe flux could result in underestimation during the fall

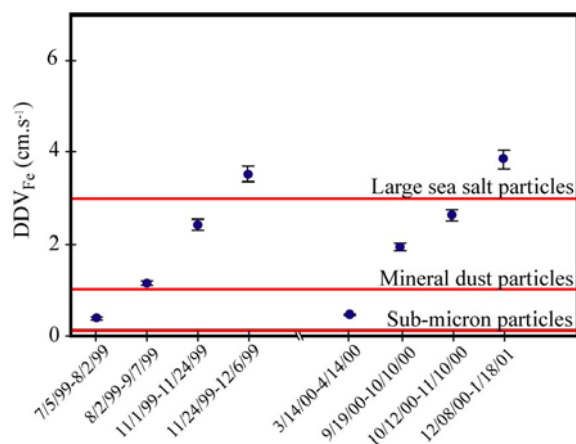


Figure 7. The dry deposition velocities of Fe-bearing aerosols (DDV_{Fe}) estimated by atmospheric deposition and aerosol samples collected at the Bermuda West Tower from July 1999 to February 2001. Only the samples (bulk deposition, wet deposition and mineral aerosol) collected during approximately the same sampling period were used to estimate the dry deposition velocity. Submicron particles have an average modeled dry deposition velocity of 0.1 cm s^{-1} (\pm a factor of 3). Mineral dust particles include micrometer mineral dust (and other supermicrometer particles not associated with sea salt); these particles have an average modeled dry deposition velocity of 1.0 cm s^{-1} (\pm a factor of 3). Large sea-salt particles have an average modeled dry deposition velocity of 3.0 cm s^{-1} (\pm a factor of 2).

and winter months, and in an overestimation during the summer months by many factors (Figure 7). Nevertheless, the mean DDV_{Fe} over the eight integral periods of 1999–2001 is 1.7 cm s^{-1} , close the model-derived dry deposition velocity of 1.0 cm s^{-1} . Using the same approach, the dry deposition velocity of Al (DDV_{Al}) is generally comparable to DDV_{Fe} during the summer (0.3 to 1.3 cm s^{-1}) but averaged 2.4 cm s^{-1} during the fall/winter integral periods.

[35] Wind speed, humidity, kinematic viscosity of air, and particle sizes are direct factors that control the dry deposition velocity of aerosol particles [Slinn and Slinn, 1980; Wesely and Hicks, 2000]. Generally, the dry deposition velocities of sea salt are higher than those of mineral aerosols [Duce et al., 1991; Arimoto et al., 2003]. Thus understanding dust and sea salt interactions should help refine global model estimates of lithogenic trace element fluxes to the ocean [Zhang and Iwasaka, 2004; Jickells et al., 2005].

[36] It is suggested by larger DDV_{Fe} and DDV_{Al} estimates in this paper that mineral aerosol and sea

salt may internally mix to form aerosol aggregates, particularly during the fall/winter months when stronger wind systems prevail at Bermuda and the nearby Sargasso Sea [Michaels and Knap, 1996; Huang et al., 1999]. While recycled Fe from sea salt contributes less than 4% of total Fe deposition at Bermuda for any given month, during the winter months, the percentage of noncrustal Al can sometimes exceed 30% [Arimoto et al., 2003]. This could be why the estimated DDV_{Al} are observed here to be generally higher than DDV_{Fe} during the fall/winter months. If so, it could implicate the role of sea salt cycling of lithogenic materials deposited to and trapped in the surface microlayer [Arimoto et al., 1997].

[37] Using Cascade impactor sampling at Bermuda, the mass median diameter of aerodynamic equivalent (AE) aerosol particles have been measured to be around $2 \mu\text{m}$ during the spring/summer months, corresponding to modeled dry deposition velocities (DDV) between 0.3 and 0.6 cm s^{-1} [Duce et al., 1976; Arimoto et al., 1997]. This agrees with the DDV_{Fe} and DDV_{Al} estimates from the summer-spring deposition data in this study. Unfortunately during fall/winter months there are no equivalent particle size measurements at Bermuda to corroborate the larger DDV estimated here. Generally larger particle sizes have higher dry deposition velocities; using a resistance method for the mineral dust, $2\text{--}4 \text{ cm s}^{-1}$ of DDV corresponds to particle mean diameters about $6\text{--}16 \mu\text{m}$ [Lee et al., 2005].

[38] In this study, the increasing trends of DDV_{Fe} and DDV_{Al} during the fall/winter months are consistent with marine particle growth during increased humidity and sea salt aerosol production. Over the Pacific, dust particles in the marine boundary layer are commonly observed to form larger aggregates through the collisions with sea-salt particles and coagulations with N or S pollutants [Andreae et al., 1986; Zhang et al., 2005; Zhang and Iwasaka, 2006]. Atmospheric factors could include increased turbulence, humidity and sea salt particle burdens as observed at Puerto Rico in the summer that may similarly explain the increasing trend of DDV_{Fe} and DDV_{Al} during the later (fall/winter) seasons at Bermuda as well [Maring et al., 2003].

5. Conclusion

[39] Atmospheric data in this study demonstrate that atmospheric Fe flux at Bermuda is highly variable on

timescales ranging from weeks to months, and annually for flux type as a function of the transport pathway and synoptic meteorology. Long-term meteorology as defined by the North Atlantic Oscillation (NAO)–PC Index appears to influence the relative dry and wet deposition at Bermuda and perhaps other oceanic areas. As reported earlier at Bermuda, the nature of atmospheric dust from regional sources and transport systems leads to dominant dry Fe deposition (~70% of total) inputs in 1999–2001.

[40] Dry deposition velocities of Fe-bearing aerosols (DDV_{Fe}) have been estimated from the atmospheric deposition and aerosol concentrations measurements. They show a consistent seasonal variation with relatively low (0.3 cm s^{-1}) to high (6 cm s^{-1}) iron dry deposition velocities (DDV_{Fe}) during the spring/summer and fall/winter, respectively. The annually averaged DDV_{Fe} is estimated at 1.7 cm s^{-1} , close to the commonly used 1.0 cm s^{-1} , yet seasonal use could lead to under or over estimation of dry fluxes at Bermuda of many factors, and potentially other marine sites.

[41] It is suggested that increased collisions and coagulations might increase dust particle size by internally mixing mineral dust, sea salt and other continental emissions in the atmospheric marine boundary at Bermuda during fall/winter. The effect would be to increase the apparent settling velocity during these periods. This indicates that seasonal meteorology should be considered when using deposition models to estimate the magnitude and mode of deposition of mineral dust at Bermuda, and perhaps generally over those ocean areas impacted by continental aerosol.

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References

Achilles, K. M., T. M. Church, S. W. Wilhelm, G. W. Luther III, and D. A. Hutchins (2003), Bioavailability of iron to

- Trichodesmium colonies in the western subtropical Atlantic Ocean, *Limnol. Oceanogr.*, *48*, 2250–2255.
- Anderson, J. R., P. R. Buseck, T. L. Patterson, and R. Arimoto (1996), Characterization of the Bermuda tropospheric aerosol by combined individual-particle and bulk-aerosol analysis, *Atmos. Environ.*, *30*(2), 319–338, doi:10.1016/1352-2310(95)00170-4.
- Andreae, M. O., R. J. Charlson, R. Bruynseels, H. Storms, R. V. Grieken, and W. Maenhaut (1986), Internal mixture of sea salt, silicates, and excess sulfate in marine aerosols, *Science*, *232*, 1620–1623, doi:10.1126/science.232.4758.1620.
- Arimoto, R. (2001), Aeolian dust and climate: Relationships to sources, tropospheric chemistry, transport and deposition, *Earth Sci. Rev.*, *54*, 29–42, doi:10.1016/S0012-8252(01)00040-X.
- Arimoto, R., and R. A. Duce (1986), Dry deposition models and the air/sea exchange of trace elements, *J. Geophys. Res.*, *91*, 2787–2792, doi:10.1029/JD091iD02p02787.
- Arimoto, R., R. A. Duce, D. L. Savoie, and J. M. Prospero (1992), Trace elements in aerosol particles from Bermuda and Barbados: Concentrations, sources and relationships to aerosol sulfate, *J. Atmos. Chem.*, *14*, 439–457, doi:10.1007/BF00115250.
- Arimoto, R., R. A. Duce, B. J. Ray, W. G. Ellis Jr., J. D. Cullen, and J. T. Merrill (1995), Trace elements in the atmosphere over the North Atlantic, *J. Geophys. Res.*, *100*, 1199–1213, doi:10.1029/94JD02618.
- Arimoto, R., B. J. Ray, N. F. Lewis, and U. Tomza (1997), Mass-particle size distributions of atmospheric dust and the dry deposition of dust to the remote ocean, *J. Geophys. Res.*, *102*, 15,867–15,874, doi:10.1029/97JD00796.
- Arimoto, R., R. A. Duce, B. J. Ray, and U. Tomza (2003), Dry deposition of trace elements to the western North Atlantic, *Global Biogeochem. Cycles*, *17*(1), 1010, doi:10.1029/2001GB001406.
- Chen, L. Q., and R. A. Duce (1983), The source of sulfate, Vanadium and mineral matter in aerosol particles over Bermuda, *Atmos. Environ.*, *17*, 2055–2064, doi:10.1016/0004-6981(83)90362-1.
- Chen, Y., and R. L. Siefert (2004), Seasonal and spatial distributions and dry deposition fluxes of atmospheric total and labile iron over the tropical and subtropical North Atlantic Ocean, *J. Geophys. Res.*, *109*, D09305, doi:10.1029/2003JD003958.
- Chester, R., M. Nimmo, and P. Corcoran (1997), Rain water-aerosol trace metal relationships at Cap Ferrat: A coastal site in the western Mediterranean, *Mar. Chem.*, *58*, 293–312, doi:10.1016/S0304-4203(97)00056-X.
- Chiapello, I., and C. Moulin (2002), TOMS and METEOSAT satellite records of the variability of Saharan dust transport over the Atlantic during the last two decades (1979–1997), *Geophys. Res. Lett.*, *29*(8), 1176, doi:10.1029/2001GL013767.
- Church, T. M., and A. J. Véron (1993), The wet deposition of trace metals from the North Atlantic troposphere: Continental sources and spatial/temporal trends, *Eos Trans AGU*, *74*(43), Fall Meet. Suppl., 146.
- Church, T., A. Véron, C. Patterson, and D. Settle (1992), Trace metal scavenging from the north Atlantic troposphere, in *Precipitation Scavenging and Atmosphere-Surface Exchange*, vol. 1, *The Georgii Volume: Precipitation Scavenging Processes*, pp. 433–446, Hemisphere, Washington, D. C.
- Church, T. M., L. Alleman, A. Véron, and G. Kim (2000), Long term and seasonal deposition of crustal dust elements to the Sargasso Sea, *Eos Trans. AGU*, *81*(48), Fall Meet. Suppl., Abstract A61D-09.

- Church, T. M., A. J. Véron, Z. Tian, E. R. Sholkovitz, and P. N. Sedwick (2006), Atmospheric input of lithogenic elements to the Sargasso Sea: The long and short of Bermuda observations, *Eos Trans. AGU*, 87(52), Fall Meet. Suppl., Abstract A42C-07.
- Desboeufs, K. V., R. Losno, F. Vimeux, and S. Cholbi (1999), The pH-dependent dissolution of wind-transported Saharan dust, *J. Geophys. Res.*, 104(D17), 21,287–21,300, doi:10.1029/1999JD900236.
- Desboeufs, K. V., A. Sofikitis, R. Losno, J. L. Colin, and P. Ausset (2005), Dissolution and solubility of trace metals from natural and anthropogenic aerosol particulate matter, *Chemosphere*, 58, 195–203, doi:10.1016/j.chemosphere.2004.02.025.
- Desenfant, F., A. J. Veron, G. F. Camoin, and J. Nyberg (2006), Reconstruction of pollutant lead invasion into the tropical North Atlantic during the twentieth century, *Coral Reefs*, 25, 473–484.
- DiTullio, G. R., D. A. Hutchins, and K. W. Bruland (1993), Interaction of iron and major nutrients controls phytoplankton growth and species composition in the tropical North Pacific Ocean, *Limnol. Oceanogr.*, 38, 495–508.
- Duce, R. A., and N. W. Tindale (1991), Atmospheric transport of iron and its deposition in the ocean, *Limnol. Oceanogr.*, 36, 1715–1726.
- Duce, R. A., G. L. Hoffman, B. J. Ray, I. S. Fletcher, P. R. Walsh, J. L. Fasching, S. R. Piotrowicz, E. J. Hoffman, J. M. Miller, and J. L. Heffter (1976), Trace metals in the marine atmosphere: Sources and fluxes, in *Marine Pollutant Transfer*, edited by H. Windom and R. Duce, pp. 77–119, D.C. Heath and Co., Lexington, Mass.
- Duce, R. A., et al. (1991), The atmospheric input of trace species to the world ocean, *Global Biogeochem. Cycles*, 5, 193–259, doi:10.1029/91GB01778.
- Fan, X., K. Okada, N. Niimura, K. Kai, K. Arao, S. Guang-Yu, Q. Yu, and Y. Mitsuta (1996), Mineral particles collected in China and Japan during the same Asian dust-storm event, *Atmos. Environ.*, 30, 347–351, doi:10.1016/1352-2310(95)00271-Y.
- Gao, Y., S.-M. Fan, and J. L. Sarmiento (2003), Aeolian iron input to the ocean through precipitation scavenging: A modeling perspective and its implication for natural iron fertilization in the ocean, *J. Geophys. Res.*, 108(D7), 4221, doi:10.1029/2002JD002420.
- Ginoux, P., J. M. Prospero, O. Torres, and M. Chin (2004), Long-term simulation of global dust distribution with the GOCART model: Correlation with North Atlantic Oscillation, *Environ. Model. Softw.*, 19(2), 113–128, doi:10.1016/S1364-8152(03)00114-2.
- Grini, A., and C. S. Zender (2004), Roles of saltation, sandblasting, and wind speed variability on mineral dust aerosol size distribution during the Puerto Rican Dust Experiment (PRIDE), *J. Geophys. Res.*, 109, D07202, doi:10.1029/2003JD004233.
- Group of Experts on the Scientific Aspects of Marine Pollution (1989), *The atmospheric input of trace species to the world ocean*, Rep. Stud., 38, 111 pp., London.
- Guieu, C., M.-D. Loye-Pilot, C. Ridame, and C. Thomas (2002), Chemical characterization of the Saharan dust end-member: Some biogeochemical implications for the western Mediterranean Sea, *J. Geophys. Res.*, 107(D15), 4258, doi:10.1029/2001JD000582.
- Hand, J. L., N. M. Mahowald, Y. Chen, R. L. Siefert, C. Luo, A. Subramaniam, and I. Fung (2004), Estimates of atmospheric-processed soluble iron from observations and a global mineral aerosol model: Biogeochemical implications, *J. Geophys. Res.*, 109, D17205, doi:10.1029/2004JD004574.
- Huang, S. L., K. A. Rahn, R. Arimoto, W. C. Graustein, and K. K. Turekian (1999), Semiannual cycles of pollution at Bermuda, *J. Geophys. Res.*, 104, 30,309–30,317, doi:10.1029/1999JD900801.
- Hurrell, J. W. (1995), Decadal trends in the North Atlantic Oscillation: Regional temperatures and precipitation, *Science*, 269, 676–679, doi:10.1126/science.269.5224.676.
- Hurrell, J. W., and H. van Loon (1997), Decadal variations associated with the North Atlantic Oscillation, *Clim. Change*, 36, 301–326, doi:10.1023/A:1005314315270.
- Hurrell, J. W., Y. Kushnir, M. Visbeck, and G. Ottersen (2003), An overview of the North Atlantic Oscillation, in *The North Atlantic Oscillation: Climate Significance and Environmental Impact*, *Geophysical Monogr. Ser.*, vol. 134, edited by J. W. Hurrell et al., pp. 1–35, AGU, Washington, D. C.
- Hutchins, D. A., A. E. Witter, A. Butler, and G. W. Luther III (1999), Competition among marine phytoplankton for different chelated iron species, *Nature*, 400, 858–861, doi:10.1038/23680.
- Jickells, T. D. (1995), Atmospheric inputs of metals and nutrients to the oceans: Their magnitude and effects, *Mar. Chem.*, 48, 199–214, doi:10.1016/0304-4203(95)92784-P.
- Jickells, T. D. (1999), The inputs of dust derived elements to the Sargasso Sea: A synthesis, *Mar. Chem.*, 68, 5–14, doi:10.1016/S0304-4203(99)00061-4.
- Jickells, T. D., and L. J. Spokes (2001), Atmospheric Iron Inputs to the Oceans, in *The Biogeochemistry of Iron in Seawater*, edited by D. R. Turner and K. Hunter, pp. 85–121, John Wiley, Hoboken, N. J.
- Jickells, T. D., T. M. Church, and W. G. Deuser (1987), A comparison of atmospheric inputs and deep-ocean particle fluxes for the Sargasso Sea, *Global Biogeochem. Cycles*, 1, 117–130, doi:10.1029/GB001i002p00117.
- Jickells, T. D., S. Dorling, W. G. Deuser, T. M. Church, R. A. Arimoto, and J. M. Prospero (1998), Air-borne dust fluxes to a deep water sediment trap in the Sargasso Sea, *Global Biogeochem. Cycles*, 12, 311–320, doi:10.1029/97GB03368.
- Jickells, T. D., et al. (2005), Global iron connections between desert dust, ocean biogeochemistry, and climate, *Science*, 308, 67–71, doi:10.1126/science.1105959.
- Kim, G., and T. M. Church (2001), Seasonal biogeochemical fluxes of ²³⁴Th, ²¹⁰Po in the open Sargasso Sea: Influence by atmospheric iron deposition, *Global Biogeochem. Cycles*, 15(3), 651–661, doi:10.1029/2000GB001313.
- Kim, G., L. Alleman, and T. M. Church (1999), Atmospheric deposition fluxes of trace elements, ⁷Be, and ²¹⁰Pb to the Sargasso Sea, *Global Biogeochem. Cycles*, 13, 1183–1192, doi:10.1029/1999GB900071.
- Lee, S., Y. Chun, and J. Nam (2005), Estimation of dry deposition during Asian dust events in spring of 2002, *J. Meteorol. Soc. Jpn.*, 83A, 241–254, doi:10.2151/jmsj.83A.241.
- Luo, C., N. M. Mahowald, N. Meskhidze, Y. Chen, R. L. Siefert, A. R. Baker, and A. M. Johansen (2005), Estimation of iron solubility from observations and a global aerosol model, *J. Geophys. Res.*, 110, D23307, doi:10.1029/2005JD006059.
- Mahowald, N. M., A. R. Baker, G. Bergametti, N. Brooks, R. A. Duce, T. D. Jickells, N. Kubilay, J. M. Prospero, and I. Tegen (2005), Atmospheric global dust cycle and iron inputs to the ocean, *Global Biogeochem. Cycles*, 19, GB4025, doi:10.1029/2004GB002402.
- Maring, H., D. L. Savoie, M. A. Izaguirre, L. Custals, and J. S. Reid (2003), Mineral dust aerosol size distribution change

- during atmospheric transport, *J. Geophys. Res.*, *108*(D19), 8592, doi:10.1029/2002JD002536.
- Martin, J. M., F. Elbaz-Poulichet, C. Guieu, M. D. Loÿe-Pilot, and G. Han (1989), River versus atmospheric input of material to the Mediterranean Sea: An overview, *Mar. Chem.*, *28*, 159–182, doi:10.1016/0304-4203(89)90193-X.
- Michaels, A. F., and H. Knap (1996), Overview of the U. S. JGOFS Bermuda Atlantic Time-series Study and the Hydrostation S program, *Deep Sea Res., Part II*, *43*(2–3), 157–198, doi:10.1016/0967-0645(96)00004-5.
- Moulin, C., C. E. Lambert, F. Dulac, and U. Dayan (1997), Control of atmospheric export of dust from North Africa by the North Atlantic oscillation, *Nature*, *387*, 691–694, doi:10.1038/42679.
- Niimura, N., K. Okada, X. Fan, K. Kenji, A. Kimio, G. Shi, and S. Takahashi (1998), Formation of Asian dust-storm particles mixed internally with sea salt in the atmosphere, *J. Meteorol. Soc. Jpn.*, *76*, 275–288.
- Okada, K., et al. (1990), X-ray spectrometry of individual Asian dust-storm particles over the Japanese Islands and the North Pacific Ocean, *Atmos. Environ., Part A*, *24*, 1369–1378.
- Prospero, J. M. (1996), Saharan dust transport over the North Atlantic Ocean and Mediterranean: An overview, in *The Impact of Desert Dust Across the Mediterranean*, edited by S. Guerzoni and R. Chester, pp. 133–151, Kluwer Acad., Dordrecht, Netherlands.
- Prospero, J. M. (1999), Long-term measurements of the transport of African mineral dust to the southeastern United States: Implications for regional air quality, *J. Geophys. Res.*, *104*(D13), 15,917–15,927, doi:10.1029/1999JD900072.
- Prospero, J. M. (2002), The chemical and physical properties of marine aerosols: An introduction, in *Chemistry of Marine Water and Sediments*, edited by A. Gianguzza, E. Pellizzetti, and S. Sammarano, pp. 35–82, Springer, Berlin.
- Rahn, K. A. (1981), The Mn/V ratio as a tracer of large-scale sources of pollution aerosols for the Arctic, *Atmos. Environ.*, *15*, 1457–1464, doi:10.1016/0004-6981(81)90352-8.
- Reid, J. S., et al. (2003), Comparison of size and morphological measurements of coarse mode dust particles from Africa, *J. Geophys. Res.*, *108*(D19), 8593, doi:10.1029/2002JD002485.
- Rogers, J. C. (1984), The association between the North Atlantic Oscillation and the Southern Oscillation in the Northern Hemisphere, *Mon. Weather Rev.*, *112*, 1999–2015, doi:10.1175/1520-0493(1984)112<1999:TABTNA>2.0.CO;2.
- Scaife, A. A., J. R. Knight, G. K. Vallis, and C. K. Folland (2005), A stratospheric influence on the winter NAO and North Atlantic surface climate, *Geophys. Res. Lett.*, *32*, L18715, doi:10.1029/2005GL023226.
- Sedwick, P. N., T. M. Church, A. R. Bowie, C. M. Marsay, S. J. Ussher, K. M. Achilles, P. J. Lethaby, R. J. Johnson, M. M. Sarin, and D. J. McGillicuddy (2005), Iron in the Sargasso Sea (Bermuda Atlantic Time-series Study region) during summer: Eolian imprint, spatiotemporal variability, and ecological implications, *Global Biogeochem. Cycles*, *19*, GB4006, doi:10.1029/2004GB002445.
- Sedwick, P. N., E. R. Sholkovitz, and T. M. Church (2007), Impact of anthropogenic combustion emissions on the fractional solubility of aerosol iron: Evidence from the Sargasso Sea, *Geochem. Geophys. Geosyst.*, *8*, Q10Q06, doi:10.1029/2007GC001586.
- Sholkovitz, E. R., and P. N. Sedwick (2006), Open-ocean deployment of a buoy-mounted aerosol sampler on the Bermuda Testbed Mooring: Aerosol iron and sea salt over the Sargasso Sea, *Deep Sea Res., Part I*, *53*(3), 547–560, doi:10.1016/j.dsr.2005.12.002.
- Slinn, S. A., and W. G. N. Slinn (1980), Predictions for particle deposition on natural waters, *Atmos. Environ.*, *14*(9), 1013–1016, doi:10.1016/0004-6981(80)90032-3.
- Spokes, L., T. Jickells, and B. Lim (1994), Solubilisation of aerosol trace metals by cloud processing: A laboratory study, *Geochim. Cosmochim. Acta*, *58*, 3281–3287, doi:10.1016/0016-7037(94)90056-6.
- Taylor, S. R., and S. M. McLennan (1985), *The Continental Crust: Its Composition and Evolution*, Blackwell, Oxford, U. K.
- Tindale, N. W., and P. P. Pease (1999), Aerosols over the Arabian Sea: Atmospheric transport pathways and concentrations of dust and sea salt, *Deep Sea Res., Part II*, *46*, 1577–1595, doi:10.1016/S0967-0645(99)00036-3.
- Uematsu, M., R. A. Duce, and J. M. Prospero (1985), Deposition of atmospheric mineral particles in the North Pacific Ocean, *J. Atmos. Chem.*, *3*(1), 123–138, doi:10.1007/BF00049372.
- Véron, A., T. M. Church, C. C. Patterson, Y. Erel, and J. T. Merrill (1992), Continental origin and industrial sources of trace metals in the northwest Atlantic troposphere, *J. Atmos. Chem.*, *14*, 339–351, doi:10.1007/BF00115243.
- Wesely, M. L., and B. B. Hicks (2000), A review of the current status of knowledge on dry deposition, *Atmos. Environ.*, *34*, 2261–2282, doi:10.1016/S1352-2310(99)00467-7.
- Woodcock, A. H. (1953), Salt nuclei in marine air as a function of altitude and wind force, *J. Meteorol.*, *10*, 362–371.
- Wu, J., E. A. Boyle, W. Sunda, and L. S. Wen (2001), Soluble and colloidal iron in oligotrophic North Atlantic and North Pacific oceans, *Science*, *293*, 847–849, doi:10.1126/science.1059251.
- Zhang, D. Z., and Y. Iwasaka (2004), Size change of Asian dust particles caused by sea salt interaction: Measurements in southwestern Japan, *Geophys. Res. Lett.*, *31*, L15102, doi:10.1029/2004GL020087.
- Zhang, D. Z., and Y. Iwasaka (2006), Comparison of size changes of Asian dust particles caused by sea salt and sulfate, *J. Meteorol. Soc. Jpn.*, *84*(5), 939–947, doi:10.2151/jmsj.84.939.
- Zhang, D. Z., Y. Iwasaka, G. Y. Shi, J. Y. Zang, A. Matsuki, and D. Trochkin (2003), Mixture state and size of Asian dust particles collected at southwestern Japan in spring 2000, *J. Geophys. Res.*, *108*(D24), 4760, doi:10.1029/2003JD003869.
- Zhang, D. Z., Y. Iwasaka, and G. Y. Shi (2005), Sea salt shifts the range sizes of Asian dust, *Eos Trans. AGU*, *86*(50), 523, doi:10.1029/2005EO500003.
- Zhao, T. L., S. L. Gong, X. Y. Zhang, and I. G. McKendry (2003), Modeled size-segregated wet and dry deposition budgets of soil dust aerosol during ACE-Asia, Implications for trans-Pacific transport, *J. Geophys. Res.*, *108*(D23), 8665, doi:10.1029/2002JD003363.
- Zhou, M., K. Okada, F. Qian, P. Wu, L. Su, B. E. Casareto, and T. Shimohara (1996), Characteristics of dust-storm particles and their long-range transport from China to Japan—Case studies in April 1993, *Atmos. Res.*, *40*, 19–31, doi:10.1016/0169-8095(95)00023-2.
- Zhuang, G., Z. Yi, R. A. Duce, and P. R. Brown (1992), Link between iron and sulphur cycles suggested by detection of Fe(II) in remote marine aerosols, *Nature*, *355*, 537–539, doi:10.1038/355537a0.