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.

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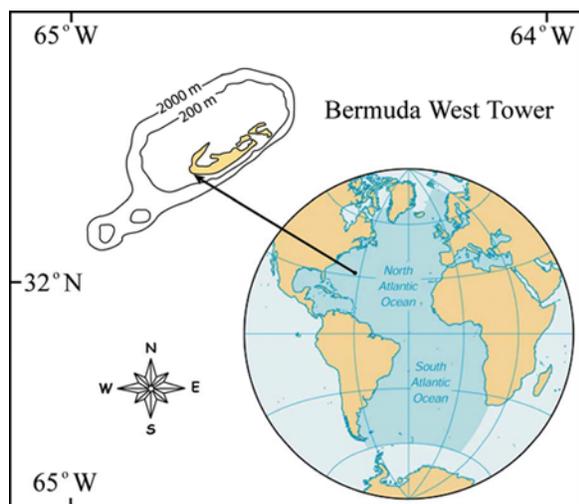
Figure 1. Location is shown for the atmospheric sampling of aerosol, wet, and bulk deposition. The Bermuda West Tower was erected as part of the AEROCE program in 1988–1997, located at Tudor Hill (32.27°N, 64.87°W) on the southwest coast of Bermuda.

[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 HNO_3 (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% HNO_3.

[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 HNO_3-HCl-HF (3:2:1 by volume), then redissolved into 5% HNO_3. 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% HNO_3 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 (±5% for Both Fe and Al)[a]

Sampling Date	Fe Flux (μg m^{-2} d^{-1})			Al Flux (μg m^{-2} 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

[a] Total 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 μg m^{-2} 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 μg m^{-2} 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 (μg m^{-2} d^{-1})	Al Flux (μg m^{-2} 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 ($\sim$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
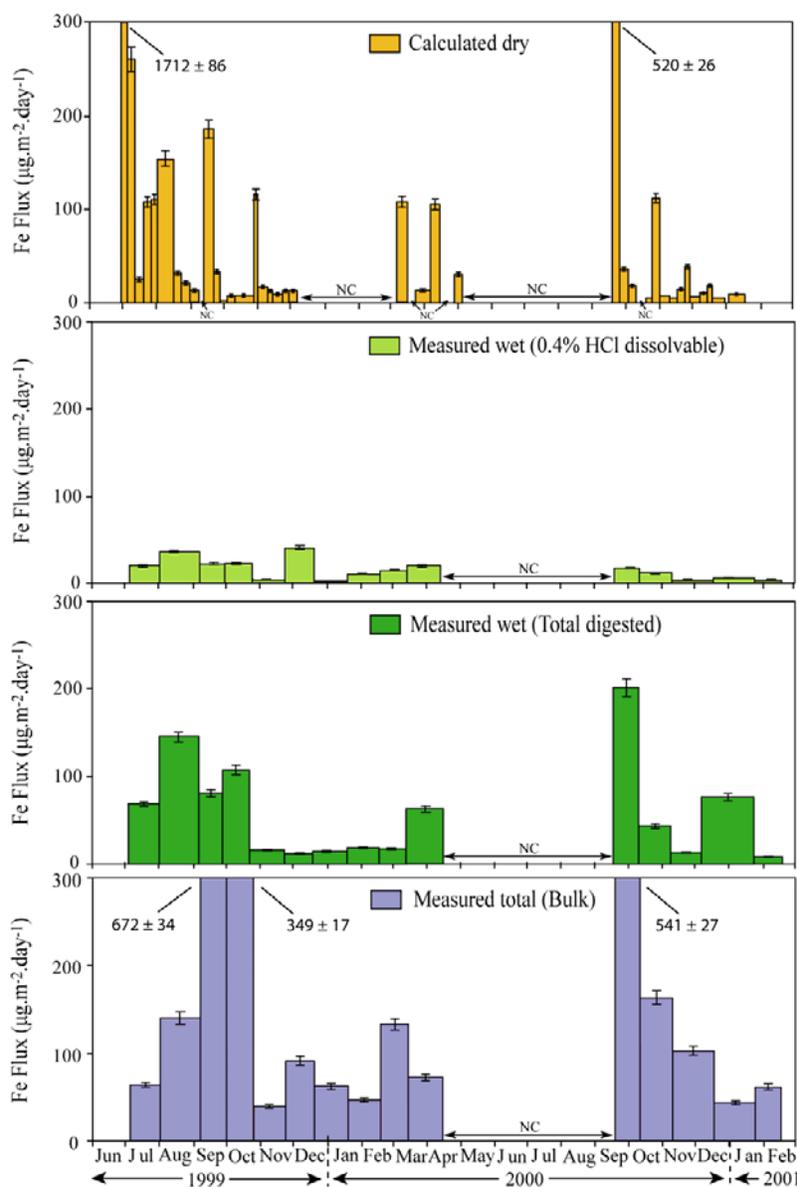
Figure 2. The temporal variation of Fe fluxes reflected by (a) aerosol samples, (b) wet deposition in 0.4% HCl leached (total dissolvable), (c) wet deposition (total digested), and (d) bulk (measured total) deposition collected at the Bermuda West Tower from July 1999 to January 2001. NC means no samples 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
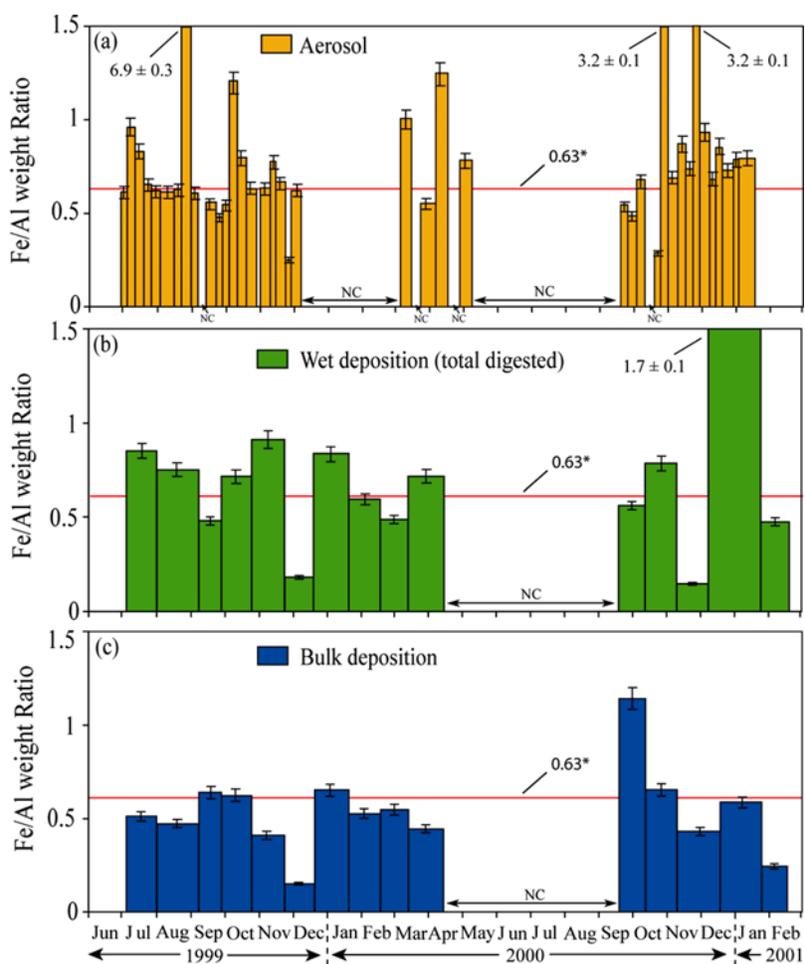
Figure 3. The Fe/Al weight ratios in (a) aerosols, (b) wet deposition (total digested with HNO_3, HCl, and HF), and (c) bulk deposition samples, collected at the Bermuda Sampling Tower from July 1999 to February 2001. NC denotes when no samples were collected, and the line marked at 0.63* indicates the average Fe/Al ratio in Saharan dust [*Arimoto et al.*, 1995; *Guieu et al.*, 2002].
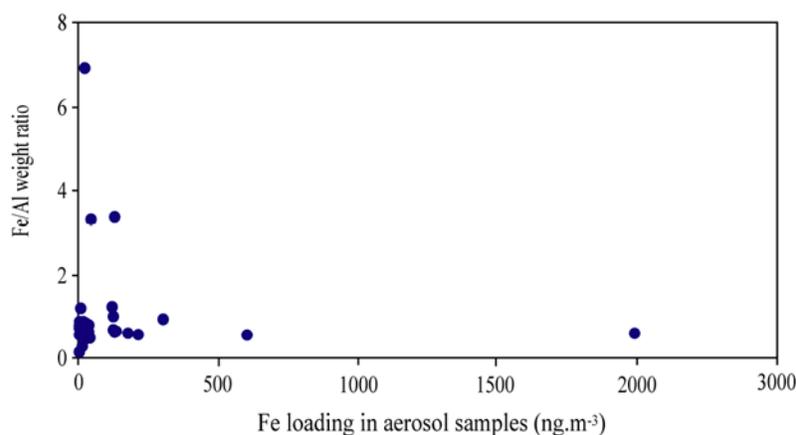
Figure 4. The Fe/Al weight ratios versus Fe loading (ng m^{-3}) in aerosol samples collected at the Bermuda West Tower from July 1999 to February 2001.
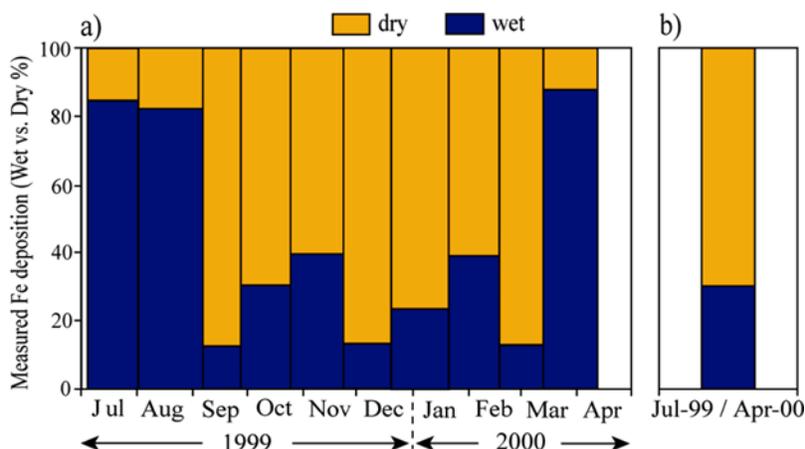
Figure 5. The Fe deposition mode (wet versus dry) at Bermuda for the sampling period from July 1999 to April 2000 (a) for individual sampling periods and (b) integrated over the entire sampling period.

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.

(a)

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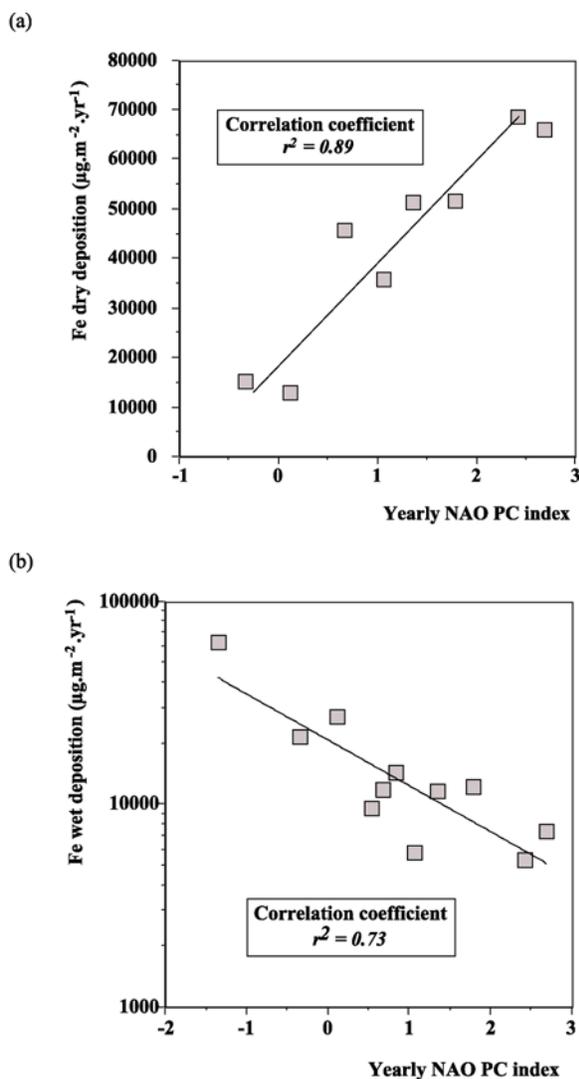
Figure 6. Relation between the long-term (a) dry Fe deposition and (b) wet deposition to Bermuda versus the annual NAO-PC index between 1988 and 1998 [*Hurrell*, 1995; *Hurrell et al.*, 2003].

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: $V_d = 0.1$ cm s^{-1} ($\pm$ a factor of 3) for submicrometer particles, $V_d = 1.0$ cm s^{-1} ($\pm$ a factor of 3) for mineral dust and other supermicrometer particles not associated with sea salt, and $V_d = 3.0$ 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:

$$DDV_{Fe} = DF/MC_{Fe} \qquad (1)$$

where DF is the dry Fe flux (μg m^{-2} 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
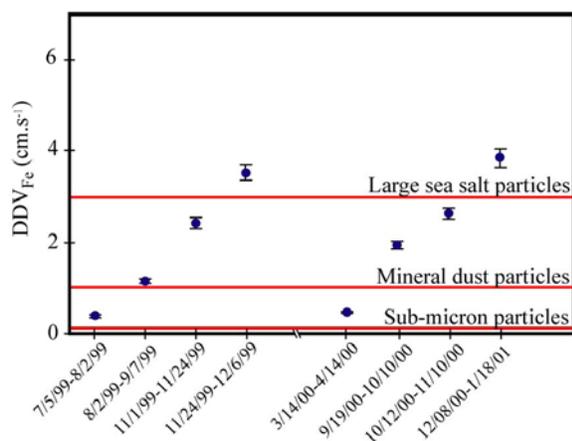
Figure 7. The dry deposition velocities of Fe-bearing aerosols (DDV$_{Fe}$) estimated by atmospheric deposition and aerosol samples collected at the Bermuda West Tower from July 1999 to February 2001. Only the samples (bulk deposition, wet deposition and mineral aerosol) collected during approximately 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 μm 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, 2–4 cm s^{-1} of DDV corresponds to particle mean diameters about 6–16 μm [*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.

Acknowledgments

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