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Instruments and Methods

Open-ocean deployment of a buoy-mounted aerosol sampler on the Bermuda Testbed Mooring: Aerosol iron and sea salt over the Sargasso Sea

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Abstract

We report results from the first deployment of a buoy-mounted aerosol sampler on the Bermuda Testbed Mooring (BTM) in the Sargasso Sea, in which a time-series of 21 aerosol samples were collected over the period May 5–September 29, 2004. These aerosol samples were analyzed for iron and soluble sodium (as a proxy for sea salt). Also analyzed was a time-series of 22 aerosol samples collected over the same period at the Tudor Hill atmospheric sampling tower on Bermuda. The buoy sampler worked as intended and successfully collected a time-series of aerosol samples, thus demonstrating that moored buoys can be used as oceanic observatories to provide information on the temporal (weekly, monthly and seasonal) variability in the concentration of aerosol iron (and other trace elements) over the surface ocean. The magnitude and time variation of aerosol Fe concentrations calculated from the BTM buoy samples are in close agreement with the corresponding aerosol Fe record from the Tudor Hill tower, which is located approximately 80 km northwest of the mooring site. Both the BTM and Tudor Hill samples record periods of high aerosol iron loadings in late June and late July 2004, reflecting the transport of soil dust from North Africa, with the highest concentration of aerosol iron at the BTM site ($0.83\,\mu g\,m^{-3}$) measured in late June. Concentrations of sea-salt aerosol calculated from the BTM samples are comparable to values measured over the Sargasso Sea and for samples collected at the Tudor Hill tower. Sea-salt aerosols do not appear to impede the collection of mineral aerosols by the buoy-mounted sampler.

Keywords: Iron; Sea water; Aerosols; Buoy systems; In situ instrumentation; Air–water exchange; Sargasso Sea; Western North Atlantic; Bermuda

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1. Introduction

John Martin's "Iron Hypothesis" has carried the element iron (Fe) into the mainstream of oceanography (Martin and Fitzwater, 1988; Martin, 1990; Martin et al., 1991), and research focusing on iron as a limiting nutrient is now an important component of chemical and biological oceanography (Falkowski et al., 1998; Johnson et al., 2002). Iron deficiency has been shown to limit the growth of phytoplankton in the high-nutrient low-chlorophyll regions of the Equatorial Pacific, the Southern

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Ocean, and the subarctic North Pacific (e.g., Coale et al., 1996, 2004; Boyd et al., 2000, 2004; De Baar et al., 2005). In addition, iron is potentially a limiting nutrient for nitrogen-fixing autotrophs such as *Trichodesmium,* which represent an important component of primary production in the subtropical and tropical oceans (e.g., Capone et al., 1997, 2005; Karl, 2000; Deutsch et al., 2001; Lenes et al., 2001). Consequently, a quantitative knowledge of the sources and sinks of iron in the surface ocean is required for the development of global and regional models of carbon cycling, the marine ecosystem, and climate change (Lefevre and Watson, 1999; Archer and Johnson, 2000; Arimoto, 2001; Moore et al., 2002, 2004; Ridgwell, 2002; Parekh et al., 2004; Jickells et al., 2005).

A major source of iron to the open ocean is the atmospheric transport and deposition of iron-bearing soil dust (mineral aerosol) from the arid regions of North Africa, central Asia and Australia (Duce and Tindale, 1991; Duce et al., 1991; Gao et al., 2001; Jickells and Spokes, 2001; Fung et al., 2000; Jickells et al., 2005). Hence, atmospheric deposition of dust-borne iron to the ocean and the subsequent release of this Fe to sea water are processes of critical importance to marine primary production. The chemical and biological response of the upper ocean to natural dust events has thus received increased study in recent years. A number of studies have confirmed that the deposition of mineral dust carried from Asia and Africa leads to elevated concentrations of dissolved Fe in surface waters of the Pacific and Atlantic Oceans (e.g., Wu and Boyle, 2002; Johnson et al., 2003; Sarthou et al., 2003; Boyle et al., 2005; Sedwick et al., 2005). However, it has proven more difficult to establish a direct link between the deposition of mineral dust and increased phytoplankton growth, given the highly episodic nature of dust deposition and the complex biogeochemistry of Fe. Gabric et al. (2002), Bishop et al. (2002), Erickson et al. (2003) and Cropp et al. (2005) have presented circumstantial evidence of this link, while the sediment trap studies of Jickells et al. (1998) and Neuer et al. (2004) suggest that deposition of African dust is not accompanied by elevated levels of primary production or export production off Bermuda and the Canary Islands, respectively.

Given the relevance of atmospheric iron deposition to the ecology and biogeochemistry of the oceans, technological advances in the measurement of mineral aerosol deposition to the ocean are an important priority for future research on this topic (Johnson et al., 2002). This need is particularly acute for the remote open ocean, for which there have been few direct measurements of aerosol concentration and composition over extended time periods. Toward this goal, we present the first results from a buoy-mounted aerosol sampler that was deployed over the period early May to late September 2004 on the Bermuda Testbed Mooring (BTM). This instrument worked as intended and successfully collected a time-series of aerosol samples, thus demonstrating that moored buoys can be used as oceanic observatories to provide information on the temporal (weekly, monthly and seasonal) variability in the concentration of aerosol iron (and other trace elements) over the surface ocean. These concentration data can then be used to estimate dry deposition fluxes of iron to the ocean.

This paper focuses specifically on the time-series records of aerosol iron and sea-salt concentrations obtained from this maiden open-ocean deployment of the buoy-mounted aerosol sampler. We compare these aerosol records with those based on samples collected concurrently at the Tudor Hill tower on the island of Bermuda.

2. Background: quantifying atmospheric iron over the open ocean

The deposition of soil dust to the ocean is extremely episodic, with large variations in mineral aerosol concentrations occurring over periods ranging from days to seasons to decades (Duce and Tindale, 1991; Duce et al., 1991; Prospero, 1996a,b; Arimoto et al., 1992, 1995; Gao et al., 2001; Prospero and Lamb, 2003; Sedwick et al., 2005). This high level of temporal and spatial variability represents a sampling challenge. Three types of "observation platforms" have previously been used to determine the atmospheric concentrations of aerosols over the open ocean, from which aerosol dry deposition fluxes may be estimated: (1) research ships; (2) island-based atmospheric sampling towers; and (3) earth-orbiting satellites. This paper introduces a fourth platform, the buoy-mounted aerosol sampler. The idea of using buoys as platforms to sample mineral aerosols sprang from the observation of reddish brown dust adhering to the meteorological sensors of a buoy moored in the ocean off northwest Africa (Sholkovitz et al., 1998). Dust recovered from this buoy was used to characterize the organic and inorganic composition of aerosols over the eastern subtropical Atlantic Ocean (Eglinton et al., 2002).

With regard to quantifying aerosols over the ocean, there are inherent advantages and disadvantages in each of the above-mentioned observation platforms. Ships are poorly suited to obtain time-series records of the aerosol concentrations over the oceans, since aerosol sampling from a ship requires heading into the wind for prolonged periods—a situation that is rarely practical. Aerosol sampling from island-based towers, on the other hand, is well established through past research programs such as SEAREX and AEROCE. Sampling from towers on Bermuda, Barbados, Enewetak and Midway, for example, has produced detailed records of mineral aerosol loadings over time scales of days to decades (e.g., Uematsu et al., 1983; Arimoto et al., 1992, 2003; Prospero, 1996a,b; Prospero et al., 2003). However, islands are not always located in suitable positions with respect to specific research questions. In addition, there is the potential for artifacts associated with sampling marine aerosols from island-based towers, most notably the potential for contamination from local (island) and nearshore sources. This typically necessitates collection of aerosols under conditions of restricted wind speed and direction, which in turn restricts—and potentially biases—the samples collected at island locations.

Data from satellites have provided tremendous insights into the temporal and spatial evolution of dust storms and the subsequent transport of soil dust over the oceans (e.g., Husar et al., 1997, 2001; Cakmur et al., 2001; Prospero et al., 2002; Moulin and Chiapello, 2004; Mahowald and Dufresne, 2004; Cropp et al., 2005; Kaufman et al., 2005). Satellite-derived images of aerosol distributions over the world ocean, updated daily, can now be accessed on internet sites maintained by NASA and the US Naval Research Laboratory (see http://toms.gsfc.nasa.gov/aereosol/aerosols_v8.html and http://nrlmry.navy.mil/aerosol). However, measurements of aerosol optical depth do not provide accurate information on the concentration and dry deposition of mineral dust at the air–sea boundary, hence it is difficult to relate satellite data to aerosol deposition fluxes. The last decade has also seen significant advances in the development of numerical models that simulate the transport and deposition of soil dust to the ocean (e.g., Ginoux et al., 2001; Colarco et al., 2003; Mahowald et al., 2003; Luo et al., 2003; Boyle et al., 2005). However, these complex models use a number of poorly constrained parameters and assumptions to estimate mineral aerosol and iron deposition fluxes to the oceans, which leads to considerable uncertainties in the model results.

As an aerosol observation platform, an autonomous sampler mounted on a buoy has several significant advantages over ships, island-based towers and satellites. On a buoy, aerosol samples can be collected close (2–3 m above) to the air–sea boundary, where deposition is actually occurring. Unlike ships, buoys can stay on station for months to years, providing a cost-effective ocean platform for time-series studies of aerosols at sea. And, unlike islands, which fall under various government jurisdictions, scientists have free access to buoys in the open ocean. There are also a number of challenges and disadvantages associated with buoy-mounted equipment. The relentless pounding of the ocean means that buoys are highly unstable platforms on which to deploy samplers and instruments. Electrical power is limited, and there is no technical support once the instrument is deployed. And finally, high wind speeds lead to high concentrations of sea spray aerosols from whitecaps and braking waves, which can potentially corrode and clog an aerosol sampling device.

In this paper, we focus on aerosol concentration data, which can be used to estimate the dry deposition of aerosol iron (F_d) using a variety of deposition models (Lewis and Schwartz, 2005). The simplest of these models, which has been used in previous aerosol studies dealing with the dust deposition to the ocean (e.g., Arimoto et al., 2003), calculates F_d according to:

$$F_d = C_a \times V_d,$$

where C_a is the aerosol Fe concentration and V_d is the so-called dry deposition velocity of the Fe-bearing aerosol particles. As noted by Arimoto et al. (2003), there is a significant uncertainty in this estimate, since V_d increases by 30-fold going from sub-micron particles ($V_d \sim 0.1\,\mathrm{cm\,s^{-1}}$) to mineral dust ($V_d \sim 1\,\mathrm{cm\,s^{-1}}$) to large sea-salt particles ($V_d \sim 3\,\mathrm{cm\,s^{-1}}$), with appropriate values of V_d for each of these particle size ranges only known to within a factor of 2–3 (± 3 for Fe-bearing mineral dust particles). Hence, dry deposition fluxes estimated from aerosol iron concentrations using this simple model are uncertain by at least a factor of 3. It should also be noted that wet deposition is an important carrier of aerosol iron to the oceans. As with dry deposition, there are likely to be large temporal and spatial variations in the wet deposition of Fe to the ocean. Estimates of wet deposition of iron require a knowledge of both precipitation flux and the concentration of Fe in rainwater over extended time periods; consequently, there are few robust estimates of the wet deposition of Fe to the ocean, with published estimates spanning a range of ~30–95% (Jickells and Spokes, 2001; Hand et al., 2004; Jickells et al., 2005). We recognize that wet deposition may account for a significant fraction of the atmospheric Fe flux to the Sargasso Sea, although further consideration of wet deposition is beyond the scope of this paper.

3. Study sites

The Woods Hole Oceanographic Institution (WHOI) buoy-mounted aerosol sampler had its maiden deployment in the open ocean from early May to late September 2004 on the BTM, which is located in the Sargasso Sea approximately 80 km southeast of Bermuda (31°41.8′N, 64°10.5′W). The BTM is located near the sites of two major oceanographic time-series programs, the Bermuda Atlantic Time-series Study (BATS; Steinberg et al., 2001) and the Oceanic Flux Program (OFP; Conte et al., 2001). Previous studies at this location have shown that the deposition of mineral aerosols originating in North Africa leads to elevated concentrations of dissolved and particulate Fe in surface water and deep-water sediment traps (Jickells et al., 1990, 1998; Sedwick et al., 2005). The BTM is a heavily utilized platform for a variety of research in the Sargasso Sea (e.g., Dickey et al., 1996, 1997; Gilboy et al., 2000; Bell et al., 2002; Conte et al., 2003; Babin et al., 2004). Deployment and recovery of the BTM buoy is carried out on the R/V *Weatherbird* by the WHOI buoy group (John Kemp and colleagues) and the University of California Santa Barbara buoy group (Tommy Dickey and colleagues). The BTM buoy is taken out of the water in April and October, when it is returned to the Bermuda Biological Station for Research (BBSR) for servicing. The tower frame on the buoy was modified to hold the WHOI aerosol sampler during the April 2004 service period.

During deployment of the buoy-mounted aerosol sampler, aerosol filter samples were also collected at the Tudor Hill atmospheric sampling tower on Bermuda. This tower is located on the southwest coast of Bermuda at 32°16′N, 64°52′W. The Tudor Hill aerosol sampler is located atop a 23-m high aluminum tower, which is situated on the side of a hill approximately 28 m above sea level. Hence, the aerosol samples at Tudor Hill were collected approximately 50 m above sea level. The sampling tower was erected in 1988 for the AEROCE project, for which daily sampling carried out until August 1998 (e.g., Arimoto et al., 1992, 2003; Prospero, 1996a,b). In early 2003, the tower was refitted with new meteorological sensors and sampling controller system. This system allows aerosol samples to be collected under specified conditions of wind speed and direction, in order to avoid contamination from local aerosol sources.

4. Sampling methods

The basic information for the aerosol samples collected at the BTM buoy and Tudor Hill tower can be found in the web site named in Section 4.1. As discussed in this section, there were a number of significant differences between the aerosol sampling protocols used at the BTM and the Tudor Hill tower. The buoy-mounted aerosol sampler uses a low-flowrate air intake system, whereas the Tudor Hill tower sampler uses a high-flowrate system. There are also significant differences in the configurations of the aerosol sampling inlets and filter supports, and in the material and nominal porosities of the aerosol filters used for the BTM and Tudor Hill sites.

4.1. Bermuda Testbed Mooring buoy

Fig. 1 is a photograph of the WHOI buoy-mounted aerosol sampler deployed on the BTM in May 2004. Three main components are indicated:

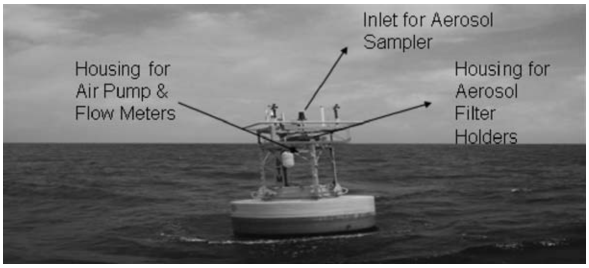

Fig. 1. Photograph of the WHOI buoy-mounted aerosol sampler on the Bermuda Testbed Mooring in May 2004. Three main components of the aerosol sampler are shown: (1) the filter-holder housing (see Fig. 2 for more details), (2) the aerosol inlet and motorized closure, and (3) the air pump and flow meter housing. For scale, the buoy float has a diameter of 2.5 m.

the aerosol inlet on top of the frame, the housing for the filter holders, and the housing for the air pump and two mass flow meters. A full description of this instrument can be found in Sholkovitz et al. (2001), and at www.whoi.edu/science/MCG/dept/facilities/sea_aer/index.html. The heart of this sampler is a 24-place filter-holder carousel that rotates successive sample filters under the aerosol inlet. Fig. 2 shows a photograph of the carousel with all 24 filter holders loaded, as well as three "field filter blanks" that are positioned in the center on the carousel and which remain in the sampler during the entire deployment period. These blank filters were analyzed along with the aerosol-laden sample filters. The sampler has two motors: one rotates the filter holders into position under the aerosol inlet for sequential sampling, while the other moves the active filter holder vertically so that it forms a seal with the aerosol inlet.

The main features of the buoy-mounted aerosol sampler include:

(1) Up to 24 sequential aerosol samples are collected on 25-mm diameter filters.
(2) Sample and blank filters are stored for return to the laboratory for analysis.
(3) Microprocessors and software are used to run the sampler, set filtering time parameters, and record the volume of air passed through each sample filter.
(4) Key meteorological parameters are measured and recorded during deployment.
(5) Autonomous remote operation is powered by batteries.
(6) All parts contacting with air and aerosols are fabricated from titanium or plastic.
(7) Construction is rugged and watertight.
(8) A motorized valve on the aerosol inlet protects samples and sampler from rain/spray.

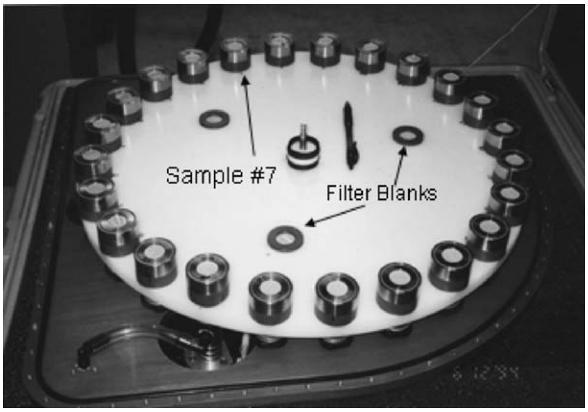

Fig. 2. Photograph showing inside of the buoy-sampler filter housing after the 5 month deployment. Shown are the 24 filter holders for time-series aerosol samples and the three field filter blanks around the center of the carousel. The tubing connecting the filters holder to the pump can be seen in the lower left corner. The brown colored filter sample (filter #7) corresponds to a period of elevated mineral aerosol loading in late June. For scale, the outer diameter of the Ti filter holders is 4.2 cm.

The sampling protocol for the buoy-mounted aerosol sampler is as follows. Isopore polycarbonate membrane filters (Millipore Corporation), 25-mm diameter and 1.2-μm pore size, were used to collect aerosols. This pore size was chosen as a compromise between a reasonable flowrate (10–16 L min^{-1}) and a pore size small enough to retain the majority of dust and sea-salt aerosols in the Bermuda region, where these particles typically fall in the 1-10 μm size range (Arimoto et al., 1997; Maring et al., 2003). While sampling was not restricted for wind direction, pumping was paused and the inlet valve was shut when it was raining and/or when the wind speed exceeded 10 m s^{-1} for more than 5 min. The latter sampling restriction was intended to limit the amount of sea spray entering the filtering system.

To measure the volume of air passed through each sample filter, two mass-flow meters were placed in series. This provided a redundant system, in case one flow meter failed, as well as two separate flowrate measurements for each sample. The averaged flowrate from the two meters was used to calculate the aerosol concentrations presented here. The flow meters were not calibrated during their 5 month deployment on the buoy.

The aerosol sampler was programmed to start collecting the first sample (filter #0) on April 15, 2004; however, inclement weather delayed deployment of the buoy until May 4, 2004, at which time sampling commenced when the sampler was powered up. Since it was not possible to reprogram the sampler to collect an initial sample on May 4, the actual time-series sampling commenced with the third filter in the carousel (filter #2). In addition, the buoy was recovered a week earlier than planned, which meant that the time-series sampling ended before the last filter in the carousel (filter #23) was used. Hence, the time-series of aerosol samples collected on the BTM contains 21 out of a possible maximum of 24 filter samples.

Prior to deployment, the sampler was programmed so that aerosols were filtered for a maximum of 3 days within an allotted "window" of sampling time. For example, BTM filter #7, which captured the largest dust loading in late June (Fig. 3), collected aerosols for 3 days out of a 7-day sampling window. If sampling was not paused due to rain or high wind speed, then aerosol sampling was completed during the first 3 days of the allotted sampling-time window. These pre-set time windows ranged from 10 days for filters #0–6 (collected mid-April to late May), to 7 days for filters #7–11 (collected late May to late July), to 5 days for filters #12–18 (collected late July to late August), then back to 7 days for filters #19–23 (collected September). The shorter sampling-time windows were intended to coincide with higher atmospheric dust loadings, which were anticipated during late July and August in our study region (e.g., Prospero, 1996a,b; Arimoto et al., 2003). A total time limit of 3 days of filtration was imposed for each sample, in order to conserve battery power for the air pump.

Over the 5 month deployment, most of the samples represent filtration for more than 60% of the allotted 3-day maximum filtration time. Of the 21 samples collected, 10 completed the maximum 3 days of filtering. Two of the samples (filters #3 and #22) filtered for only small percentages (10% and 7%, respectively) of the maximum filtration time, due to the rain control. Over the entire deployment, sampling was paused for only 2% of the time due to the high wind speed ($>10\,\mathrm{m\,s^{-1}}$) control; this was only significant for filter #13 (August 3–7), for which filtration was paused for 12% of the 5-day sampling window. The percentage of rain-control paused time over the entire deployment was 53%. This rain-control time is inconsistent with meteorological records of rainfall at Bermuda during the deployment, and we suspect that this inconsistency reflects a technical problem with the rain detector, such as sea spray or condensation providing a false "rain" signal. Despite this problem, which is currently being investigated, a complete time-series of aerosol samples was collected by the buoy-mounted sampler at the BTM.

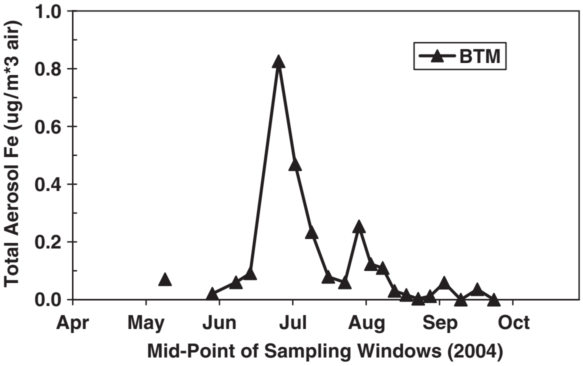

Fig. 3. Time-series record of the total iron aerosol concentrations at the BTM buoy. Total refers to the sum of two acid dissolutions (1N HCl and concentrated HNO_3–HF). The mid-point of the air filtering date is used as the time point on the x-axis.

Averaged flow rates for each filter sample ranged from 10.4 to $15.7\,\mathrm{L\,min^{-1}}$, although most of the samples were collected within a narrower range of flow rates ($11–14\,\mathrm{L\,min^{-1}}$). Flow rates did not decrease for filters that collected higher loadings of mineral aerosol and/or sea salt. Therefore, we conclude that sea salt did not significantly clog the filters, nor hamper the ability of the buoy-mounted sampler to record dust events at sea. Wind speeds were not high during the deployment of the sampler, exceeding $10\,\mathrm{m\,s^{-1}}$ for only 2% of the total sampler deployment time, and rarely exceeding $15\,\mathrm{m\,s^{-1}}$. Hence, from the results of this deployment, we cannot gauge the effectiveness of the $10\,\mathrm{m\,s^{-1}}$ wind speed control in reducing the sea-salt loading of the sample filters. The average volume of air that passed through the filters ranged from a 32 to $62\,\mathrm{m^3}$, with the exceptions of filters #3 and #22, for which the rain control resulted in extended periods of non-sampling time.

4.2. Tudor Hill atmospheric sampling tower

Aerosol samples were collected at the Tudor Hill atmospheric sampling tower over time periods of approximately 1 week. Samples were collected using high-flowrate filtration system whereby air is pulled through large ($\sim400\,\mathrm{cm^2}$) Whatman-41 filters at flowrates of around $1000\,\mathrm{L\,min^{-1}}$ (Uematsu et al., 1983; Arimoto et al., 1992). These filters are widely used by atmospheric chemists; their fibrous cellulose structure makes them effective filters for collecting micron and submicron aerosols. Sampling details for the aerosol samples collected at Tudor Hill between April 28, 2004 and October 5, 2004 can be found in the web site named in Section 4.1. The timing and sampling intervals are generally similar to those used for the BTM buoy-mounted sampler. The sampling-time windows for each of the Tudor Hill filters ranged from 5 to 9 days. The sector-control was set so that air sampling only proceeded under conditions of onshore (marine-air) wind direction (210°–315°). Filtering was also paused when wind speed was less than $1\,\mathrm{m\,s^{-1}}$, or in the presence of rain, or when humidity exceeded 95%.

Pumps were on for periods ranging from 4% to 81% of each sampling-time window, which were set by the timing of weekly service visits to the tower site. Two exceptions are filters #17 and #21, for which sampling lasted only several hundred minutes in total. The typical flow rates and filter area of the Tudor Hill sampling system are around 100 times greater than those for the BTM buoy-mounted aerosol sampler. Excluding filters #17 and #21, total air volumes of $400–8300\,\mathrm{m^3}$ were filtered for the Tudor Hill aerosol samples. In contrast, the maximum volume of air filtered by the BTM buoy-mounted sampler (for filter #6) was $62.2\,\mathrm{m^3}$.

5. Sample treatment and analytical methods

A simple, three-step dissolution method was applied to both the BTM and Tudor Hill aerosol filter samples. Analysis of solutions from this dissolution scheme yields three operationally defined pools of aerosol Fe; in this paper, we will mainly consider the total concentration of iron that was rendered soluble during the three dissolution steps. Dissolutions were carried out in series, using sealed Teflon vials. All equipment used in the dissolution procedure was acid cleaned and rinsed with Milli-Q deionized water before use. All acids used in the dissolutions were prepared from reagent grade acids by sub-boiling distillation in Teflon. The entire polycarbonate filters from the BTM buoy sampler were used in the dissolution steps, whereas only a small portion (~1/100 of the active filter area) of the Tudor Hill cellulose filters were used, for which each portion was weighed to quantify the fraction of the total filter area. Given that the aerosol samples were collected and stored for 2–8 months prior to dissolution and analysis, the leaching protocol was *not* designed to specifically address questions concerning the oxidation state and solubility of the aerosol iron.

The aerosol-laden filters were first soaked in 4 g of Milli-Q water (initial pH ~5.5) for 1 day, with the objective of dissolving the sea-salt component. Thus defined soluble sodium concentration, which provides a proxy for calculating sea-salt aerosol concentrations (Arimoto et al., 2003), was determined in a small aliquot of the Milli-Q leach solutions using atomic emission spectroscopy. The filtered material was next leached in 1 N HCl at room temperature for 1 day; the 1 N HCl solution was produced by adding concentrated HCl to the Teflon vials containing the filters in the Milli-Q leach solutions. The rationale for the choice of a 1 N HCl leach solution is that such a solution was expected to dissolve the less-refractory iron-bearing phases in the aerosol samples, such as iron oxides. After removing all of the dilute HCl leach solution for analysis, 1.8 mL of a 3:1 mixture of concentrated HNO_3 and HF was added to each filter residue in the Teflon vial. The tightly sealed vials were then placed in an oven at 50 °C for 36 h. This last dissolution step is assumed to have dissolved most of the lithogenic mineral particles; no attempt was made to dissolve any highly refractory mineral particles remaining after the HNO_3–HF treatment.

Iron was measured in each of the leach solutions using the colorimetric ferrozine method of Stookey (1970). A reductant (10% by weight hydroxylamine hydrochloride in 6 N HCl) was added to reduce all dissolved iron to Fe(II). This step typically involved adding 0.2 mL of the reductant to 2.0 mL of 10-fold dilutions of the 1 N HCl and HNO_3–HF leach solutions, followed by heating in an oven at 60 °C for 1 h to facilitate reduction of ferric iron. Hence, total dissolved iron was determined in each of the leach solutions. The ferrozine method, as employed here, had a detection limit equivalent to an aerosol iron concentration of $\sim 0.015\,\mu\mathrm{g\,m^{-3}}$. Iron was also measured in three types of blanks: (1) the acid leach matrices, (2) leaches of new Isopore and Whatman-41 filters, and (3) leaches of the Isopore and Whatman-41 "field blank" filters. All of the blank leach solutions contained negligible or low iron concentrations (equivalent to arerosol concentrations of $\sim 0.015\,\mu\mathrm{g\,m^{-3}}$) that have been subtracted from the aerosol concentrations. The dissolved Fe concentrations of the Milli-Q leach solutions were too low to be reliably quantified using the ferrozine method; these samples were subsequently analyzed using inductively coupled plasma mass spectrometry, as part of a larger analysis of the BTM aerosol samples (data not shown).

6. Results and discussion

6.1. Aerosol sample color

When the filter holder housing of the buoy-mounted sampler was opened after the 5-month deployment, the color of the filters indicated that the sampler had captured at least one period of high mineral aerosol loading. The reddish brown color of

BTM filter #7 (see http://www.whoi.edu/science/MCG/dept/facilities/sea_aer/maintextpg.html) and corresponding Tudor Hill sample #A9, collected in late June, stood out in comparison with the other filters. Aerosols of this color are typical of soil dust transported from North Africa to Bermuda in the summer (Tomza et al., 2001; Arimoto et al., 2002). As discussed below, sample filters #7 and A9 yielded the highest concentrations of aerosol iron. With the exception of samples collected during "dusty" periods in June and July, the colors of the BTM filter samples were indistinct. The aerosols collected on our Tudor Hill filter samples, which had higher mass loadings, displayed colors ranging from light gray to distinctly red-brown, consistent with the descriptions of Tomza et al. (2001). The former were mostly associated with samples collected in April and May, prior to the high aerosol loadings. Samples collected in September were very light brown to gray in color.

6.2. Aerosol iron concentrations

The time-series of aerosol iron concentrations from the BTM buoy and Tudor Hill tower are presented in Figs. 3 and 4. The concentration data are presented in units of μg of aerosol iron per m^3 of air, which has been calculated from the mass of iron on each filter divided by the total volume of air passed through that filter. Fig. 3 shows only the BTM aerosol Fe time-series, and Fig. 4 compares the time-series of aerosol Fe calculated from the BTM and Tudor Hill samples. Figs. 3 and 4 show "total" aerosol Fe concentrations, as calculated from the sum of the iron concentrations in the HCl and HNO_3–HF leach solutions. The aerosol Fe concentrations, calculated from the separate analyses of the HCl and HNO_3–HF leach solutions, can be found in the web site named in Section 4.1.

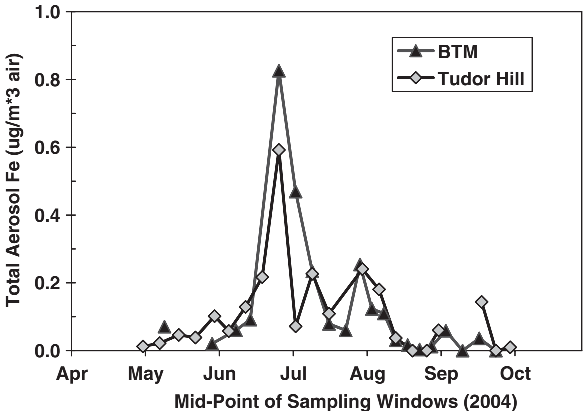

Fig. 4. Comparison of time-series records of total aerosol iron concentrations from the BTM buoy-mounted sampler and Tudor Hill tower.

These data show that the BTM buoy-mounted sampler captured two periods of elevated aerosol Fe loading (Fig. 3), with the highest concentration of total aerosol iron (~0.83 μg m^{-3}) associated with sample filter #7, collected between June 24 and June 30, 2004. A second, smaller, peak in total aerosol Fe concentration (~0.25 μg m^{-3}) is associated with filter #12, collected during the period July 29–August 2, 2004. While the two peaks in aerosol Fe concentrations are the most outstanding features of the BTM time-series, it is important to note that these two high-dust periods are defined by more than one datum. In each case, the actual periods of elevated aerosol Fe loadings span about 2 weeks, or 3–4 sample filters. The limited temporal resolution of the BTM aerosol sampler—3 days of filtering over periods of 5, 7 and 10 days—may mean that we failed to sample even higher aerosol iron loadings at this location. Indeed, large day-to-day variability in mineral dust concentration has been documented in aerosol samples collected at the Tudor Hill tower (Arimoto, 2001; Arimoto et al., 1995, 2003).

The major fraction of aerosol Fe in the BTM samples is contained in the more refractory material, that which was dissolved in the concentrated HNO_3–HF mixture. The only substantial concentration of aerosol iron in the 1N HCl leach solution (12% of the total Fe) was associated with BTM sample filter #7, which records the highest total aerosol Fe loading of the time-series (Fig. 3); this same feature is observed in the Tudor Hill aerosol Fe record (filter #A9). As already noted, Fe concentrations in the Milli-Q leach solutions were too low to be reliably quantified using the ferrozine method. Subsequent analysis by ICP-MS revealed that less than 4% of the total aerosol Fe in BTM sample filter #7 was rendered soluble in the Milli-Q deionized-water leach, a finding that is consistent with other studies of aerosol-iron solubility (e.g., Jickells and Spokes, 2001; Hand et al., 2004).

The Tudor Hill tower samples record the highest concentration of total aerosol Fe (~0.59 μg m^{-3}) between June 24 and July 1, 2004. The timing of this concentration maximum almost exactly matches the late June peak (~0.83 μg m^{-3}) recorded by the BTM buoy-mounted sampler (Fig. 4). The Tudor Hill tower samples also record a second peak in late

July–early August, with a maximum aerosol Fe concentration (~0.24 μg m^{-3}) nearly equal to that recorded by the BTM samples. In addition, both the BTM and Tudor Hill samples record small maxima in aerosol Fe loadings in early and mid-September, with higher concentrations recorded at the Tudor Hill site (Fig. 4).

When the time-series records of total aerosol Fe from BTM and Tudor Hill sites are compared, only one substantial difference is apparent. As noted above, both time-series samples record high dust loadings during the last week of June. However, the next sample collected at the BTM (July 1–7) also has a relatively high aerosol Fe loading of ~0.47 μg m^{-3}, but this is not the case for the corresponding Tudor Hill sample (July 1–8), which records a total aerosol Fe concentration of only 0.07 μg m^{-3}. Some of this discrepancy may be explained by small differences in sampling times and/or trajectories of airborne dust carried over the two sites. However, a more likely explanation is that air sampling was paused at the Tudor Hill site during much of this period. The Tudor Hill sampler filters air when wind direction is in the range 210°–315° (i.e., SSW through NW winds). During summer, however, when plumes of North African dust are carried over the Sargasso Sea, winds often blow from the southeast quadrant—"out-of-sector" for Tudor Hill. Indeed, meteorological data from the Tudor Hill tower show that the wind direction was out-of-sector for 53% of the time from July 1 to July 8. In contrast, for samples collected either side of this time period, wind direction was out-of-sector for only about 20% of the time. This comparison highlights an important advantage of the buoy-mounted aerosol sampler over an island-based tower; namely, the aerosol samples collected on a buoy are not constrained or aliased by wind direction. The inventories of total aerosol Fe integrated over the entire sampling periods for the BTM and Tudor Hill sites differ by only 22%. Most of this difference may be attributed to the lower aerosol Fe concentration in the one filter sample collected at Tudor Hill for the period July 1–8, as discussed above (Fig. 4).

Since North African mineral aerosols contain around 4% Fe by mass (Jickells and Spokes, 2001), the maximum aerosol Fe concentration at BTM in late June 2004 is equivalent to a mineral dust concentration of ~20 μg m^{-3}. This maximum dust concentration falls in the low to mid-range of maximum summer dust concentrations reported for Tudor Hill (Arimoto, 2001; Prospero, 1996a,b). A more direct comparison can be made between our BTM Fe data and the historical time-series record of total aerosol iron at the Tudor Hill tower, derived from neutron-activation analysis of aerosol samples collected during 1988–1994 as part of the AEROCE program (Arimoto, 2001; Arimoto et al., 1995, 2003). In comparison with this long-term record, the highest aerosol Fe concentration recorded by the BTM buoy sampler (0.83 μg m^{-3}, filter #7) is similar to the highest Fe concentrations recorded at Tudor Hill over the summers of 1990, 1991 and 1994. In contrast, our BTM Fe maximum is only ~20–30% of the average maximum aerosol Fe concentration for the summers of 1988, 1989, 1992 and 1993. Hence, the 2004 BTM aerosol Fe record represents a "low-dust" year in the Sargasso Sea. We can also compare our 2004 aerosol Fe data from the BTM sampler with the results of Sedwick et al. (2005), who collected aerosols in the BATS region aboard R/V *Weatherbird II* in July–August 2003. These authors report total aerosol iron concentrations averaging 0.56 μg m^{-3}—quite similar to our maximum BTM value—for the period July 24–August 5, 2003, with a maximum daily concentration of 1.55 μg m^{-3}.

6.3. Sea-salt aerosol concentrations

The time-series records of sea-salt aerosol concentrations from the BTM buoy sampler and the Tudor Hill tower are shown together in Fig. 5,

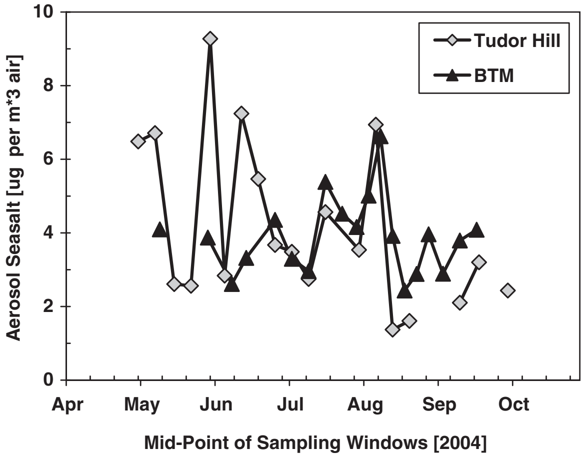

Fig. 5. Time-series records of the sea-salt aerosol concentrations from the BTM buoy sampler and the Tudor Hill tower. Sea-salt concentrations were derived from the measurement of Na in Milli-Q water leach solutions.

presented as μg sea salt per m^3 of air. These values are calculated from the Na concentrations in the Milli-Q leach solutions and the Na/salinity ratio (0.306) of seawater. The full concentration range of the BTM samples is small (2–6 $\mu g\,m^{-3}$), with the majority of samples falling in the range of 3–5 $\mu g\,m^{-3}$. The time variations in sea-salt aerosol concentrations display no obvious cyclicity nor any relationship with aerosol Fe loadings. The calculated sea-salt content of the Tudor Hill samples suggests a wider range in sea-salt aerosol concentrations than do the BTM samples; most of the Tudor Hill samples indicate aerosol concentrations within the range of the BTM samples, although five of the Tudor Hill samples record significantly higher sea-salt aerosol concentrations of 7–9 $\mu g\,m^{-3}$ (Fig. 5). This difference may be due to the wind speed restriction of the BTM sampler, which paused sampling when wind speed exceeded 10 $m\,s^{-1}$. However, given that sampling was paused for only 2% of the entire BTM deployment time due to high wind speed (see Section 4.1), the higher sea-salt loadings at Tudor Hill may also reflect (1) spatial differences between the two sampling sites (particularly elevation above the sea surface), (2) aerosols produced by surf on the barrier reef that is upwind of the Tudor Hill site, and (3) the use of different types of filters for the two sites. The range in sea-salt aerosol loadings estimated from our BTM and Tudor Hill samples are similar to the historical sea-salt aerosol data from Tudor Hill in late spring and summer (Arimoto et al., 1992, 1995). Our BTM sea-salt aerosol concentrations are also similar to both measured and modeled values for the Sargasso Sea (e.g., Erickson et al., 1986; Gong et al., 1997).

An important conclusion that emerges from a comparison of the BTM and Tudor Hill records is that sea salt does not hamper the ability of the buoy-mounted sampler to provide a time-series record of mineral aerosol concentrations over the open ocean, at least for wind speeds below 10 $m\,s^{-1}$. There are two pieces of evidence to support this assertion. First, the flow rates for the BTM sample filters remained within a narrow range (12–14 $L\,min^{-1}$) and showed no significant decrease during the periods of elevated dust or salt loadings. Second, the aerosol Fe records from the BTM and Tudor Hill sites are similar, even though the former uses a low-flowrate system and the latter uses a high-flowrate system, with different inlet configurations and filter types used at each site. The results of previous studies indicate that the large filters used for the high-flowrate Tudor Hill sampling system were not appreciably clogged by sea salt (e.g., Savoie et al., 2002).

6.4. Buoys vs. islands as aerosol sampling platforms

One fundamental question that our observations can address concerns the lateral scale over which buoy- and island-based measurements can be extrapolated to estimate aerosol concentrations over the adjacent ocean. The results shown in Fig. 4 indicate that both the magnitude and time variation of mineral aerosol concentrations were very similar for the BTM and Tudor Hill sites during the buoy-sampler deployment period. These sites are separated by approximately 80 km of open ocean, and use aerosol sampling systems which have several significant differences. Although this agreement may be fortuitous, our results suggest that both sampling systems were equally efficient in sampling mineral aerosols from laterally homogeneous air masses. However, this conclusion applies to average aerosol concentrations integrated over periods of several days or more; we cannot resolve short-term variations over time scales of a day or less. Our results are consistent with the numerical simulations of dust transport described by Mahowald et al. (2003), for which there is a strong correlation between mineral aerosol concentrations for open-ocean locations separated by less than 100 km, as well as satellite observations of aerosols over the North Atlantic (Chiapello et al., 1999; Kaufman et al., 2005).

Our results suggest that mineral aerosol concentrations estimated from samples collected at the Tudor Hill tower provide a robust indication of aerosol loadings over the BTM and the BATS region, ~80 km to the southeast of Bermuda. This implies that aerosol samples collected at other island-tower sites are likely to be representative of the adjacent ocean region over distances of ~100 km; for example, observations made at the Bellows Beach tower on O'ahu are likely to be representative of conditions at ocean station ALOHA (Hawaii Ocean Time-series site), some 100 km to the north. This does not mean, however, that buoy-mounted samplers are redundant at mooring sites such as the BTM or station ALOHA; here the buoy sampler has an advantage over island-based towers in that it can collect samples in any wind direction, whereas sampling of uncontaminated marine air from island-based towers is limited according to wind direction.

7. Conclusions

The WHOI buoy-mounted aerosol sampler successfully collected a time-series of 21 aerosol samples during its a 5-month (May–September 2004) deployment on the Bermuda Testbed Mooring. Measurements of iron concentrations show that the buoy sampler recorded the passage of two plumes of mineral aerosol from North Africa: a large dust event in late June, and a smaller dust event in late July. The buoy-mounted aerosol sampler is now ready to enhance ocean–atmosphere research, through its ability to function as an autonomous, sea-surface aerosol observatory over periods of many months. This instrument is well suited for the sampling of aerosols in remote areas of the open ocean, since island-based aerosol measurements are unlikely to reflect aerosol loadings over scales of more than ~100 km (Mahowald et al., 2003). In combination with modern oceanographic sampling and observation methods, the buoy-mounted aerosol sampler offers to advance our understanding of the effects of atmospheric deposition on the concentration and speciation of trace elements in the upper ocean, and on biological processes in ocean surface waters.

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References

Archer, D.E., Johnson, K., 2000. A model of the iron cycle in the ocean. Global Biogeochemical Cycles 14, 269–279.

Arimoto, R., 2001. Eolian dust and climate: relationships to sources, tropospheric chemistry, transport and deposition. Earth–Science Review 54, 29–42.

Arimoto, R., Duce, R.A., Savoie, D.L., Prospero, J.M., 1992. Trace elements in aerosol particles from Bermuda and Barbados: concentrations, sources and relationships to aerosol sulfate. Journal of Atmospheric Chemistry 14, 439–457.

Arimoto, R., Duce, R.A., Ray, B.J., Ellis Jr., W.G., Cullen, J.D., Merrill, J.T., 1995. Trace elements in the atmosphere over the North Atlantic. Journal of Geophysical Research 100, 1199–1213.

Arimoto, R., Ray, B.J., Lewis, N.F., Tomza, U., 1997. Mass-particle size distributions of atmospheric dust and the dry deposition of dust to the remote ocean. Journal of Geophysical Research 102, 15,867–15,874.

Arimoto, R., Balsam, W., Schloesslin, C., 2002. Visible spectroscopy of aerosol particles collected on filters: iron oxide minerals. Atmospheric Environment 36, 89–96.

Arimoto, R., Duce, R.A., Ray, B.J., Tomza, U., 2003. Dry deposition of trace elements to the western North Atlantic. Global Biogeochemical Cycles 17 (1), 1010.

Babin, S.M., Carton, J.A., Dickey, T.D., Wiggert, J.D., 2004. Satellite evidence of hurricane-induced phytoplankton blooms in an ocean desert. Journal of Geophysical Research 109, C03043.

Bell, J., Betts, J., Boyle, E., 2002. MITESS: a moored in situ trace element serial sampler for deep-sea mooring. Deep-Sea Research Part I 49, 2103–2118.

Bishop, J.K.B., Davis, R.E., Sherman, J.T., 2002. Robotic observations of dust storm enhancement of carbon biomass in the North Pacific. Science 298, 817–821.

Boyd, P.W., Watson, A., Law, C.S., Abraham, E., Trull, T., Murdoch, R., Bakker, D.C.E., Bowie, A.R., Buesseler, K.O., Chang, H., Charette, M.A., Croot, P., Downing, K., Frew, R., Gall, M., Hadfield, M., Hall, J., Harvey, M., Jameson, G., La Roche, J., Liddicoat, M., Ling, R., Maldonado, M., McKay, R.M., Nodder, S., Pickmere, S., Pridmore, R., Rintoul, S., Safi, K., Sutton, P., Strzepek, R., Tannenberger, K., Turner, S., Waite, A., Zeldis, J., 2000. A mesoscale phytoplankton bloom in the polar Southern Ocean stimulated by iron fertilization. Nature 407, 695–702.

Boyd, P.W., et al., 2004. The decline and fate of an iron-induced subarctic phytoplankton bloom. Nature 428, 549–553.

Boyle, E.A., Bergquist, B.B., Kayser, R., Mahowald, N., 2005. Iron, manganese, and lead at Hawaii Ocean Time-series Station ALOHA: temporal variability and an intermediate water hydrothermal plume. Geochimica et Cosmochimica Acta 69, 933–952.

Cakmur, R.V., Miller, R.L., Tegen, I., 2001. A comparison of seasonal and interannual variability of soil dust aerosols over the Atlantic Ocean as inferred by TOMS AI and AVHRR AOT retrievals. Journal of Geophysical Research 106 (D16), 18,287–18,303.

Capone, D.G., Zehr, J.P., Paerl, H.W., Bergman, B., Carpenter, E.J., 1997. *Trichodesmium*, a globally significant marine cyanobacterium. Science 276, 1221–1229.

Capone, D.G., Burns, J.A., Montoya, J.P., Subramaniam, A., Mahaffey, C., Gunderson, T., Michaels, A.F., Carpenter, E.J., 2005. Nitrogen fixation by *Trichodesmium* spp.: an important source of new nitrogen to the tropical and subtropical North Atlantic Ocean. Global Biogeochemical Cycles 19, GB2024.

Chiapello, I., Prospero, J.M., Herman, J.R., Hsu, N.C., 1999. Detection of mineral dust over the North Atlantic Ocean and Africa with the Nimbus 7 TOMS. Journal of Geophysical Research 104 (D8), 9277–9292.

Coale, K.H., et al., 1996. A massive phytoplankton bloom induced by ecosystem-scale iron fertilization experiment in the equatorial Pacific Ocean. Nature 383, 495–501.

Coale, K.H., et al., 2004. Southern ocean iron enrichment experiment: carbon cycling in high- and low-Si waters. Science 304, 408–414.

Colarco, P.R., Toon, O.B., Holben, B.N., 2003. Saharan dust transport to the Caribbean during PRIDE 1. Influence of dust sources and removal mechanisms on the timing and magnitude of downwind aerosol optical depth events from simulations of in situ and remote sensing observations. Journal of Geophysical Research 108 (D19), 8589.

Conte, M.H., Ralph, N., Ross, E.H., 2001. Seasonal and interannual variability in deep ocean particle fluxes at the Oceanic Flux Program (OFP)/Bermuda Atlantic Time Series (BATS) site in the western Sargasso Sea near Bermuda. Deep-Sea Research II 48, 1471–1505.

Conte, M.H., Dickey, T.D., Weber, J.C., Johnson, R.J., Knap, A.H., 2003. Transient physical forcing of pulsed export of bioreactive material to the deep Sargasso Sea. Deep-Sea Research I 50, 1157–1187.

Cropp, R.A., Gabric, A.J., McTainsh, G.H., Braddock, R.D., Tindale, N., 2005. Coupling between ocean biota and atmospheric aerosols: dust, dimethylsulphide, or artifact? Global Biogeochemical Cycles 19, GB4002.

De Barr, H.J.W., Boyd, P.W., Coale, K.H., Landry, M.R., Tsuda, A., Assmy, P., Bakker, C.F., Bpzec, M., Barber, R.T., Brzezinski, M.A., Buesseler, K.O., Boye, M., Croot, PL., Gervais, F., Gorbunov, M.Y., Harrison, P.J., Hiscock, W.T., Laan, P., Lancelot, C., Law, C.S., Levasseur, M., Marchetti, A., Millero, F.J., Nishioka, J., Nojiri, Y., van Oijen, T., Riebesell, U., Rijkenberg, M.J.A., Saito, H., Takeda, S., Timmermans, K.R., Veldhuis, M.J.W., Waite, A.M., Wong, C.-S., 2005. Synthesis of iron fertilization experiments: from the Iron Age to the Age of Enlightenment. Journal of Geophysical Research 110, C09S16.

Deutsch, C., Gruber, N., Key, R.M., Sarmiento, J.L., Ganachaud, A., 2001. Denitrification and N_2 fixation in the Pacific Ocean. Global Biogeochemical Cycles 15, 483–506.

Dickey, T.D., Fyre, M., Stramska, H., Jannasch, H.W., Boyle, E., Manov, D., Sigurdson, D., Michaels, A., 1996. The Bermuda Testbed Mooring program. Eos 76, 78.

Dickey, T.D., Frye, D., Jannasch., H.W., Boyle, E., Knap, A.H., 1997. Bermuda sensor system testbed. Sea Technology 81, 81–86.

Duce, R.A., Tindale, N.W., 1991. Atmospheric transport of iron and its deposition in the ocean. Limnology and Oceanography 36, 1715–1726.

Duce, R.A., Liss, P.S., Merrill, J.T., Atlas, E.L., Buat-Menard, P., Hicks, B.B., Miller, J.M., Prospero, J.M., Arimoto, R., Church, T.M., Ellis, W., Galloway, J.M., Hansen, L., Jickells, T.D., Knap, A.H., Reinhardt, K.H., Schneider, B., Soudine, A., Toko, J.J., Tsunogai, S., Wollast, R., Zhou, M., 1991. The atmospheric input of trace species to the world ocean. Global Biogeochemical Cycles 5, 193–259.

Eglinton, T.I., Eglinton, G., Dupont, L., Sholkovitz, E.R., Montlucon, D., Reddy, C.M., 2002. Composition, age and provenance of organic matter in N.W. African dust over the Atlantic Ocean. Geochemistry, Geophysics, Geosystems, doi:10.1029/2001GC000269.

Erickson, D.J., Merrill, J.T., Duce, R.A., 1986. Seasonal estimates of global atmospheric sea-salt distributions. Journal of Geophysical Research 91, 1067–1072.

Erickson III, D.J., Hernandez, J.L., Ginoux, P., Gregg, W.W., McClain, C., Christian, J., 2003. Atmospheric iron delivery and surface ocean biological activity in the Southern Ocean and Patagonian region. Geophysical Research Letters 30 (12), 1609.

Falkowski, P.G., Barber, R.T., Smetacek, V., 1998. Biogeochemical controls and feedbacks on ocean primary production. Science 281, 200–206.

Fung, I.Y., Meyn, S.K., Tegen, I., Doney, S.C., John, J.G., Bishop, J.K.B., 2000. Iron supply and demand in the upper oceans. Global Biogeochemical Cycles 14, 281–295.

Gabric, A.J., Cropp, R., Ayers, G.P., McTainsh, G., Braddock, R., 2002. Coupling between cycles of phytoplankton biomass and aerosol optical depth as derived from SeaWIFS time series in the Subantarctic Southern Ocean. Geophysical Research Letters 29 (7).

Gao, Y., Kaufman, Y.J., Tanre, D., Kolber, D., Falkowski, P.G., 2001. Seasonal distribution of aeolian iron fluxes to the global ocean. Geophysical Research Letters 28, 29–32.

Gilboy, T.P., Dickey, T.D., Sigurdson, D.E., Yu, X., Manov, D., 2000. An intercomparison of current measurements using a vector measuring current meter, an acoustic Doppler current profiler, and a recently developed acoustic current meter. Journal of Atmospheric and Ocean Technology 17, 561–574.

Ginoux, P., Chin, M., Tegen, I., Prospero, J.M., Holben, B., Dubovik, O., Lin, S.-J., 2001. Sources and distributions of dust aerosols simulated with the GOCART model. Journal of Geophysical Research 106 (D17), 20,255–20,273.

Gong, S.L., Barrie, L.A., Prospero, J.M., Savoie, D.L., Ayers, G.P., Blanchet, J.-P., Spacek, L., 1997. Modeling sea-salt aerosols in the atmosphere 2. Atmospheric concentrations and fluxes. Journal of Geophysical Research 102, 3819–3830.

Husar, R.B., Prospero, J.M., Stowe, L.L., 1997. Characterization of trophospheric aerosols over the oceans with the NOAA advanced very high resolution radiometer optical thickness operational product. Journal of Geophysical Research 102, 16,889–16,909.

Husar, R.B., Tratt, D.M., Schichtel, B.A., Falke, S.R., Li, F., Jaffe, D., Gasso, S., Gill, T., Laulainen, N.S., Lu, F., Reheis, M.C., Chun, Y., Westphal, D., Holben, B.N., Gueymard, C., McKendry, I., Kuring, N., Feldman, G.C., McClain, C., Frouin, R.J., Merrill, J., DuBois, D., Vignola, F., Murayama, T., Nickovic, S., Wilson, W.E., Sassen, K., Sugimoto, N., Malm, W.C., 2001. Asia dust events of April 1998. Journal of Geophysical Research 106 (D16), 18,317–18,330.

Jickells, T.D., Spokes, L.J., 2001. Atmospheric iron inputs to the oceans. In: Turner, D.R., Hunter, K.A. (Eds.), The Biogeochemistry of Iron in Seawater. Wiley, New York, pp. 85–121.

Jickells, T.D., Deuser, W.G., Belastock, R.A., 1990. Temporal variations in the concentrations of some particulate elements in the surface waters of the Sargasso Sea and their relationship to deep-sea fluxes. Marine Chemistry 29, 203–219.

Jickells, T.D., Dorling, S., Deuser, W.G., Church, T.M., Arimoto, R., Prospero, J.M., 1998. Air-borne dust fluxes to the deep water sediment trap in the Sargasso Sea. Global Biogeochemical Cycles 12, 311–320.

Jickells, T.D., An, Z.S., Andersen, K.K., Baker, A.R., Bergametti, G., Brooks, N., Cao, J.J., Boyd, P.W., Duce, R.A., Hunter, K.A., Kawahata, H., Kubilay, N., LaRoche, J., Liss, P.S., Mahowald, N., Prospero, J.M., Ridgwell, A.J., Tgen, I., Torres, R., 2005. Global iron connections between desert dust, ocean biogeochemistry, and climate. Science 308, 67–71.

Johnson, K.S., Moore, J.K., Smith, W.O., 2002. Workshop highlights iron dynamics in ocean carbon cycle. Eos 83, 482–484.

Johnson, K.S., Elrod, V.A., Fitzwater, S.E., Plant, J.N., Chavez, F.P., Tanner, S.J., Gordon, R.M., Westphal, D.L., Perry, K.D., Wu, J., Karl, D.M., 2003. Surface ocean-lower atmosphere interactions in the Northeast Pacific Ocean gyre: aerosols, iron and the ecosystem response. Global Biogeochemical Cycles 17 (2).

Karl, D.M., 2000. A new source of 'new' nitrogen in the sea. Trends in Microbiology 8, 301.

Kaufman, Y.J., Koren, I., Remer, L.A., Tanre, D., Ginoux, P., Fan, S., 2005. Dust transport and deposition observed from the Terra-Moderate Resolution Imaging Spectroradiameter (MODIS) spacecraft over the Atlantic Ocean. Journal of Geophysical Research 110, D10S12.

Lefevre, N., Watson, A.J., 1999. Modeling the geochemical cycle of iron in the oceans and its impact on atmospheric CO_2 concentrations. Global Biogeochemical Cycles 13, 727–736.

Lenes, J.M., Darrow, B.P., Cattrall, C., Heil, C., Vargo, G.A., Callanhan, M., Byrne, R.H., Prospero, J.M., Bates, D.E., Fanning, K.A., Walsh, J.J., 2001. Iron fertilization and the Trichodesmium response on the West Florida shelf. Limnology and Oceanography 46, 1261–1277.

Lewis, E.R., Schwartz, S.E., 2005. Sea salt aerosol production: mechanisms, methods, measurements, and models—a critical review. Geophysical Monograph Series, vol. 152. American Geophysical Union, Washington, DC, 413pp.

Luo, C., Mahowald, N.M., del Corral, J., 2003. Sensitivity study of meterological parameters on mineral aerosol mobilization, transport and distribution. Journal of Geophysical Research 108 (D15), 4447.

Mahowald, N., Luo, C., del Corral, J., Zender, C.S., 2003. Interannual variability in atmospheric mineral aerosols from a 22-year model simulation and observational data. Journal of Geophysical Research 108 (D12).

Mahowald, N.M., Dufresne, J.-L., 2004. Sensitivity of TOMS aerosol index to boundary layer height: implications for detection of mineral aerosol sources. Geophysical Research Letters 31, L033103.

Maring, H., Savoie, D.L., Izaguirre, M.A., Custals, L., Reid, J.S., 2003. Mineral dust aerosol size distribution change during atmospheric transport. Journal of Geophysical Research 108, 8592.

Martin, J.H., 1990. Glacial–interglacial CO_2 changes: the Iron Hypothesis. Paleoceanography 5, 1–13.

Martin, J.H., Fitzwater, S.E., 1988. Iron deficiency limits phytoplankton growth in the north-east Pacific subarctic. Nature 331, 341–343.

Martin, J.H., Gordon, R.M., Fitzwater, S.E., 1991. The case for iron. Limnology and Oceanography 36, 1793–1802.

Moore, J.K., Doney, S.C., Glover, D.M., Fung, I.Y., 2002. Iron cycling and nutrient limitation patterns in the surface waters of the world ocean. Deep-Sea Research II 49, 463–507.

Moore, J.K., Doney, S.C., Lindsay, K., 2004. Upper ocean ecoysystem dynamics and iron cycling in a global 3-D model. Global Biogeochemical Cycles 18, GB4028.

Moulin, C., Chiapello, I., 2004. Evidence of the control of summer transport of African dust over the Atlantic by Sahel sources from TOMS satellites (1979–2000). Geophysical Research Letters 31, L02107.

Neuer, S., Torres-Padron, M.E., Gelado-Caballero, M.D., Rueda, M.J., Hernandez-Brito, J., Davenport, R., Wefer, G., 2004. Dust deposition pulses to the eastern subtropical North Atlantic gyre: does ocean's biogeochemistry respond? Global Biogeochemical Cycles 18, GB4020.

Parekh, P., Fellows, M., Boyle, E.A., 2004. Modeling the global iron cycle. Global Biogeochemical Cycles 18, BG1002.

Prospero, J.M., 1996a. The atmospheric transport of particles to the oceans. In: Ittekkot, V., Schafer, P., Honjo, S., Depetris, P.J. (Eds.), Particle Flux in the Oceans. Wiley, New York, pp. 19–52.

Prospero, J.M., 1996b. Saharan dust transport over the North Atlantic Ocean and Mediterranean Sea: an overview. In: Guerzoni, S., Chester, R. (Eds.), The Impact of Desert Dust Across the Mediteranean. Kluwer Academic Publisher, Dordrecht, pp. 133–151.

Prospero, J.M., Lamb, P.J., 2003. African droughts and dust transport to the Caribbean: climate change implications. Science 302, 1024–1027.

Prospero, J.M., Ginoux, P., Torres, O., Nicholson, S.E., Gill, T.E., 2002. Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 total ozone mapping spectrometer (TOMS) absorbing aerosol product. Reviews of Geophysics 40 (1), 2-1–2-31.

Prospero, J.M., Savoie, D.L., Arimoto, R., 2003. Long-term record of nss-sulfate and nitrate aerosols on Midway Island, 1981–2000: evidence of increased (and now decreasing ?) anthropogenic emissions from Asia. Journal of Geophysical Research 108 (D1), 4019.

Ridgwell, A.J., 2002. Dust in the Earth system: the biogeochemical linking of land, air and sea. Philosophical Transactions of the Royal Society of London A 360, 2905–2924.

Sarthou, G., Baker, A.R., Blain, S., Achterberg, E.P., Boye, M., Bowie, A.R., Croot, P., Laan, P., de Baar, H.J.W., Jickells, T.D., Worsfold, P.J., 2003. Atmospheric iron deposition and sea-surface dissolved iorn concentrations in the eastern Atlantic Ocean. Deep-Sea Research I 50, 1339–1352.

Savoie, D.L., Arimoto, R., Keene, W.C., Prospero, J.M., Duce, R.A., Galloway, J.N., 2002. Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean. Journal of Geophysical Research 107 (D18), 4356.

Sedwick, P.N., Church, T.M., Bowie, A.R., Marsay, C.M., Ussher, S.J., Achilles, K.M., Lethaby, P.J., Johnson, R.J., Sarin, M.M., McGillicuddy, D.J., 2005. Iron in the Sargasso Sea (BATS region) during summer: Eolian imprint, spatiotemporal variability, and ecological implications. Global Biogeochemical Cycles 19.

Sholkovitz, E., Allsup, G., Arthur, R., Hosom, D., 1998. Aerosol sampling from ocean buoys shows promise. Eos 79, 29 and 37.

Sholkovitz, E., Allsup, G., Hosom, D., Purcell, M., 2001. An autonomous aerosol sampler and elemental analyzer for ocean buoys and remote land sites. Atmospheric Environment 35, 2969–2975.

Steinberg, D.K., Carlson, C.A., Bates, R.R., Johnson, R.J., Michaels, A.F., Knap, A.H., 2001. Overview of the US JGOFS Bermuda Atlantic Time-series Study (BATS): a decade-scale look at ocean biology and biogeochemistry. Deep-Sea Research II 48, 1405–1447.

Stookey, L.L., 1970. Ferrozine—a new spectrometric reagent for iron. Analytical Chemistry 42, 779–781.

Tomza, U., Arimoto, R., Ray, B.J., 2001. Color-related differences in the chemical composition of aerosol-laden filters. Atmospheric Environment 35, 1703–1709.

Uematsu, M., Duce, R.A., Prospero, J.M., Chen, L., Merrrill, J.T., McDonald, R.L., 1983. Transport of mineral aerosol from Asia over the North Pacific Ocean. Journal of Geophysical Research 88, 5343–5352.

Wu, J., Boyle, E., 2002. Iron in the Sargasso Sea: implications for the processes controlling the dissolved Fe distribution in the ocean. Global Biogeochemical Cycles 16.