RESEARCH ARTICLE

Key Points:
- Atmospheric input into ocean constitutes budgetary component of chemical species
- Assessment of the atmospheric input is difficult

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A novel tracer technique to quantify the atmospheric flux of trace elements to remote ocean regions

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Abstract Atmospheric input into the global ocean constitutes an important budgetary component of numerous chemical species and plays a key role in controlling biogeochemical processes in the ocean. Assessment of this input is difficult, however, because measurements of deposition rates to the ocean, particularly in remote areas, are rare and susceptible to problems of temporal and spatial variability. While the collection and analysis of aerosol samples is somewhat routine, the chemical concentration data collected from ship board or land-based aerosol samplers in and of themselves cannot yield the deposition flux of trace elements; a method is required to transform concentration measurements into flux. The ability to derive the atmospheric flux of ^{7}Be from its ocean inventory provides a key linkage between the atmospheric concentration of chemical species and their deposition to the ocean. We have demonstrated that estimates of the atmospheric flux of trace elements (TEs) can be made by multiplying the ocean inventory of ^{7}Be x [TE/^{7}Be] ratio in bulk aerosols. Flux estimates for trace elements made by the ^{7}Be ocean inventory method were comparable to fluxes derived from rain samples collected on the island of Bermuda. The situation at Bermuda allows such testing to be made, where ocean-based methods can be calibrated by convenient land locations. Our results suggest that this method would be useful for remote areas where fixed sampling stations do not exist; that is, the majority of the global ocean.

1. Introduction

Atmospheric input into the global ocean constitutes an important budgetary component of numerous chemical species [e.g., *Duce et al*., 1991; *Prospero*, 1996, 2002] and plays a key role in controlling biogeochemical processes in the ocean [e.g., *Martin et al*., 1990, *Falkowski*, 1997; *Coale et al*., 1996, *Morel et al*., 2003; *Morel and Price*, 2003; *Krishnamurthy et al*., 2009]. Assessment of this input is difficult, however, because measurements of deposition rates to the ocean are rare and susceptible to problems of temporal and spatial variability. Particularly in remote ocean areas (most of the ocean), one simply cannot "be there" to collect samples on a routine basis.

Given the dearth of direct measurements, the ocean community has relied on atmospheric transport and deposition models [e.g., *Gao et al*., 2003; *Hand et al*., 2004; *Jickells et al*., 2005; *Mahowald et al*., 2005, 2009]. However, these models themselves suffer from uncertainties in the amounts of precipitation delivered to the ocean and in the parameterization of aerosol removal processes. Thus, indirect methods are often used to estimate atmospheric inputs. One such approach involves the use of natural radionuclides which are delivered to the ocean from the atmosphere. These provide useful tracers for the input of atmospherically derived chemical species, as the source terms are definable and measurements are not readily contaminated within normal environmental conditions [*Turekian et al*., 1983; *Kadko and Prospero*, 2011]. The information obtained from the use of such tracers can be used to characterize the deposition of trace metals. For example, the global distribution of the cosmogenic isotope ^{7}Be in the atmosphere and its deposition to the ocean has been modeled [*Brost et al*., 1991; *Koch et al*., 1996, 2006], and parameters developed in such models can be extended to the modeling of other species including those from anthropogenic sources [*Kadko and Prospero*, 2011; *Landing et al*., 2012]. Here, we describe how the ability to readily derive the atmospheric flux of ^{7}Be from its ocean inventory provides the means to link the chemical concentration data of aerosols to flux. We develop a novel approach utilizing the ^{7}Be inventory of the upper ocean and

the ^{7}Be and trace metal concentration of aerosols to provide estimates of the atmospheric deposition of trace elements into the subtropical Atlantic BATS (Bermuda Time-Series) site, and compare these results to direct flux measurement made on the nearby island of Bermuda. The unique observing facilities at this locale allowed both time series ocean measurements and nearby island atmospheric monitoring which provided an ideal testing opportunity. Ultimately, the goal is to apply this method to remote ocean areas where fixed, land sampling stations do not exist.

2. Background

Be-7 is a cosmic-ray-produced isotope (half-life = 53.3 day) that is deposited to the ocean surface primarily by precipitation and subsequently homogenized within the surface mixed layer [e.g., *Silker*, 1972; *Aaboe et al.*, 1981; *Young and Silker*, 1980; *Kadko and Olson*, 1996; *Kadko*, 2000, 2009; *Kadko and Prospero*, 2011; *Kadko and Johns*, 2011]. Therefore, the ^{7}Be flux and water column inventory vary as a function of rainfall, and over broad oceanic regions are relatively constant. This is manifested by the observation that water column inventories are negatively related to surface salinity [*Young and Silker*, 1980; *Kadko and Olson*, 1996; *Kadko and Johns*, 2011]. In the absence of physical removal processes other than radioactive decay, the water column inventory of the isotope represents an integration of the atmospheric input flux over approximately the previous mean life (77 day) of the isotope, making it relevant to studies encompassing seasonal timescales. With regard to particle scavenging, ^{7}Be is considered a conservative tracer in the ocean regime. Numerous studies have shown that ^{7}Be is quite soluble in the relatively low-particle environment of the open ocean, thus allowing particle scavenging losses to be ignored. *Silker* [1972] found that <10% of ^{7}Be was on the insoluble fraction in the water column during his study at Bermuda, and *Andrews et al.* [2008] pumped ~1000 L of seawater through a 1 μm Hytrex filter from surface water of the Sargasso Sea and observed that the particulate matter had a ^{7}Be activity below the detection limit. *Aaboe et al.* [1981] measured ^{7}Be in the western Sargasso Sea and found that the calculated ocean standing crop is equal to that predicted from precipitation collectors, as have *Kadko and Prospero* [2011] from the same area. This suggests a conservative behavior in this environment.

2.1. ^{7}Be Input

Atmospheric deposition of ^{7}Be has been routinely measured by precipitation collection on land for many years, and long-term, systematic studies of precipitation have shown that temporal variation of ^{7}Be deposition occurs over time scales ranging from single precipitation events to seasons [e.g., *Olsen et al.*, 1985; *Dibb*, 1989; *Todd et al.*, 1989; *Baskaran et al.*, 1993]. In Bermuda for example, a correlation between monthly precipitation and ^{7}Be flux was observed over the course of an 8 month study [*Turekian et al.*, 1983], and more recently over a nearly 2 year period at the same location [*Kadko and Prospero*, 2011]. This correlation is not surprising as wet deposition dominates the ^{7}Be flux [*Kadko and Prospero*, 2011].

The standing crop (inventory) of ^{7}Be in the surface ocean also affords a way to determine the depositional flux of this isotope. Given the relatively short half-life of ^{7}Be, at steady state the input flux of ^{7}Be (atoms min^{-1} m^{-2}) should be balanced by the ^{7}Be inventory, or decay rate, integrated over the water column (dpm m^{-2}). Note that the standing crop of ^{7}Be in the ocean is relatively insensitive to short-term (daily–weekly) variability of ^{7}Be input since the standing crop at any one time has a seasonal mean life, lending an inertia to the system with respect to short term input events. *Aaboe et al.* [1981] calculated open ocean standing crops of ^{7}Be by measuring mixed layer activities and assuming an exponential decrease in ^{7}Be below the mixed layer. The results were comparable to that expected from previously collected precipitation data, indicating that the ^{7}Be flux for an oceanic location can be estimated from water column inventories. *Kadko and Prospero* [2011] made the first concurrent measurements of ^{7}Be deposition from rain samples (on Bermuda) with ocean profile measurements of ^{7}Be inventories at the nearby US JGOFS Bermuda Atlantic Time-Series study (BATS) and Hydrostation S in the Sargasso Sea. Between April 2007 and November 2008, the average oceanic ^{7}Be flux determined from the ocean inventory was nearly identical at the two sites (480 ± 100 dpm m^{-2} d^{-1}),which matched the flux determined at Bermuda by precipitation collection (weighted average 480 ± 270 dpm m^{-2} d^{-1}). This value is quite similar to that reported earlier for precipitation collected at the same site in Bermuda (470 dpm m^{-2} d^{-1}) [*Turekian et al.*, 1983]. The 1-dimensional approach assumed here (no lateral effects) appears valid due to the relatively short half-life of

[7]Be and the constancy of [7]Be deposition over broad latitudinal bands [e.g., *Young and Silker*, 1980; *Kadko and Olson*, 1996; *Kadko and Prospero*, 2011; *Kadko and Johns*, 2011].

Of relevance to this work, the chemical concentration data obtained from ship board or land-based aerosol samplers in and of themselves cannot yield the deposition flux of trace elements; a method is required to transform concentration measurements into flux. The ability to derive the atmospheric flux of [7]Be from its ocean inventory provides a key linkage between the atmospheric concentrations of chemical species and their deposition to the ocean.

3. Methods

3.1. Atmospheric Deposition Measurements

Rainfall samples were collected between 15 June 2011 and 27 July 2013 at the Bermuda Institute of Ocean Sciences (BIOS) located near the eastern end of the island (32°22.28′N, 64°41.80′W). Collectors were situated near ground level, clear of surrounding vegetation, at the meteorological monitoring station in front of the BIOS laboratory, about 10 m above sea level. This is a Bermuda Air Quality Program site used for ambient air quality monitoring. To estimate the atmospheric deposition of [7]Be, plastic buckets (660.5 cm^2) were deployed for collection of fallout over ∼3 week intervals that varied as a function of rainfall, i.e., longer deployment when rainfall was less abundant. After collection, dilute HCl and stable Be yield tracer were added to the buckets. The [7]Be was then coprecipitated with iron hydroxide, and the precipitate was dried in plastic Petri dishes. The sample was subsequently counted by gamma spectrometry. Be-7 has a readily identifiable gamma peak at 478 keV. The counting system was calibrated for all samples by preparing a commercial standard in geometry identical to the samples. The stable Be in the precipitate was then measured by ICPMS to calculate the [7]Be recovery (generally>70%) during the coprecipitation procedure.

For trace elements, all the sampling equipment was precleaned with appropriate acid washing and handled using "clean lab" protocols. Wet deposition was collected for trace element analysis using an N-CON (ADS 00–120) automated wet deposition collector deployed at the BIOS site described above. The N-CON sampler only opened when it was raining. Inside the sampler bucket, two identical acid-washed 2L FEP-Teflon receiving bottles attached to 180 cm^2 polycarbonate funnels via PTFE Teflon vented "collars" were nested in order to collect duplicate samples for every deployment interval. Deployments averaged 3 weeks in duration yielding sample volumes between 100 and 1500 mL. The receiving bottles were sealed upon recovery and shipped to FSU for processing and analysis. At FSU, the samples were acidified to 0.045M HCl and 0.048M HNO$_3$ using ultrapure acids (Optima, Fisher Scientific), then exposed to UV radiation (254 nm UVA; 730 microwatts/cm^2)for at least 48 h to desorb the collected trace elements from the bottle walls [*Landing et al.*,1998]. FEP Teflon bottles are about 50% transparent to the UVA radiation produced using low pressure Hg vapor lamps. The samples were then analyzed using an Agilent 7500cs quadrupole ICPMS equipped with an octopole collision/reaction cell (CRC) filled with He and H$_2$ for simultaneous analysis of As and Se in addition to the other trace elements. The analysis was calibrated using multielement standards in the same acid matrix. A check standard was analyzed every 10th sample to make minor drift corrections. Analytical sensitivities (cps) for each element were compared to spiked samples (standard additions) to confirm that there was no signal suppression or enhancement due to slight matrix differences between samples and standards. Trace element concentrations for the samples were then multiplied by the rain depth at the site (as recorded by a tipping bucket rain gauge) to calculate wet deposition fluxes for each deployment interval.

3.2. Aerosol Measurements

Aerosol samples were collected between 26 June 2011 and 27 July 2013 at the same location as the rain collectors. This site was chosen over the Tudor Hill site on the western end of the island that has been used as a clean air site but has been shown to undersample (by ∼40%) rainfall amounts compared to other Bermuda sites, as well as the [7]Be flux compared to that captured at BIOS and required to sustain the ocean [7]Be inventory [*Kadko and Prospero*, 2011]. Aerosol samples were obtained with a Tisch TE-5170V-BL high-volume aerosol sampler, modified to collect 12 replicate samples on acid-washed [*Wallace et al.*, 1977; *Baker et al.*, 2006] 47mm diameter Whatman-41 (W-41) filters, using procedures identical to those used for the US GEOTRACES aerosol program [*Morton et al.*, 2013]. The particle collection efficiency of W-41 filters is well established: >99% for non-sea salt sulfate, 95% for nitrate marine aerosols, and 99% for mineral dust [*Li-Jones and Prospero*, 1998]. Although they report data for only a few elements, *Wallace et al.* [1977] and

Baker et al. [2006] have shown that the blanks and detection limits can be significantly improved with acid washing of W-41 filters. In contrast, *Morton et al.* [2013] reported that prewashing W-41 filters had a more modest effect on lowering the filter blanks. In order to minimize the filter blanks for our experiment, the W-41 filters were precleaned before deployment using three cycles of leaching with 0.5M HCl (Optima) then rinsing with ultrahigh purity water (UHP water) according to trace element protocols in a HEPA-filtered laminar flow hood [*Morton et al.*, 2013; similar to *Baker et al.*, 2006].

For determinations of total aerosol metal loadings (Na, Mg, Al, P, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Rb, Sr, Zr, Cd, Sb, Ba, La, Ce, Nd, Pb, Th, U), the W-41 filter discs were digested in tightly capped Teflon vials using sequential additions of concentrated acid solutions: 1.0 mL 15.8M nitric acid (Optima) followed by 0.5 mL of 15.8M HNO_3 with 0.1 mL of 29M hydrofluoric acid (Optima) at 150°C [*Morton et al.*, 2013]. Any solution remaining after each digestion step was heated to near dryness. After the final digestion and evaporation step, the samples were resuspended in 0.4 mL of 15.8M HNO_3, then diluted to 20 mL with UHP water (final concentration of 0.32 M nitric acid) for analysis by quadrupole ICP-MS (Agilent 7500cs). All filter digestions and trace metal determinations were performed under Class-100 clean laboratory conditions at the National High Magnetic Field Laboratory at Florida State University (FSU).

For ^{7}Be, three of the 47 mm aerosol samples were stacked in a plastic Petri dish and counted by gamma spectroscopy. As described above, this configuration was calibrated with a commercially prepared mixed solution of known gamma activities.

3.3. Measurement of ^{7}Be Profiles at Ocean Stations

The ocean inventory of ^{7}Be was determined from profiles collected in the vicinity of the U.S. Joint Global Ocean Flux Study Bermuda Atlantic Time-Series Study (BATS) (31°50′N, 64°10′W) and Hydrostation S (32°10′N, 64°30′W) in the Sargasso Sea. Samples were collected from the R/V Atlantic Explorer and subsequently processed by methods described in detail elsewhere [*Kadko and Prospero*, 2011]. Briefly, ^{7}Be was collected at various depths by pumping 400–700 L of seawater via a 1.5 in hose through iron-impregnated acrylic fibers [*Lal et al.*, 1988; *Krishnaswami et al.*, 1972; *Lee et al.*, 1991]. The efficiency of the fiber for extraction of Be from seawater was determined by adding 500 mL of a 1000 ppm Be atomic absorption standard to a drum containing seawater. The seawater was pumped through an iron fiber cartridge and at every 100 L the Be content of the cartridge effluent was measured by atomic absorption. From these data, the integrated Be extraction efficiencies were calculated. For sample volumes in the range 400–700 L, based on several trials, the extraction efficiencies were respectively, 82 ± 3%–76 ± 2%. On land, the fibers were dried and ashed. The ash was subsequently pressed into a pellet (5.8 cm diameter) and placed on a low-background germanium gamma detector. In some cases where low ^{7}Be concentrations were expected (typically at depths greater than ∼190 m), two samples were taken from the same depth to ensure adequate signal, and the ashed fibers were combined and placed in Marinelli beakers to accommodate the large ash volume. For such samples, up to ∼1200 L were extracted and analyzed. The detector was calibrated for the pellet and Marinelli beaker geometries by adding a commercially prepared mixed solution of known gamma activities to an ashed fiber, pressing the ash into a pellet or adding the ash to a beaker, and counting the activities to derive a calibration curve. The uncertainty of the extraction efficiency (4%) and the detector efficiency (2%) was, in all cases, smaller than the statistical counting error and the uncertainty in the blank.

4. Approach: Determining Trace Element Flux

The flux (F_i) of an aerosol element into the ocean can be described as the sum of wet and dry deposition processes, respectively:

$$F_i = R \cdot S \cdot Cp_i \cdot \rho + Cp_i \cdot Vd \qquad (1)$$

where F_i = flux to the oceans (μg/m/d), R = precipitation rate (m_{rain}/d), S = washout ratio (kg_{air}/kg_{rain}; the concentration in rain (μg/kg_{rain}) divided by the aerosol concentration (μg/kg_{air})), Cp_i = aerosol concentration (μg/m^3_{air}), ρ = density of water over the density of air ((∼1000 kg_{rain}/m^3_{rain})/(∼1.2 kg_{air}/m^3_{air}) = ∼833 ($kg_{rain} \cdot m^3_{air}$)/($m^3_{rain} \cdot kg_{air}$)), and Vd = aerosol dry deposition velocity (m/d). The dry deposition velocity is difficult to describe properly because of variability arising from changing humidity, wind speed, and particle size. It has been estimated that for submicron aerosol particles Vd varies by a factor of 3 [*Duce et al.*, 1991]. There are equally large if not greater uncertainties associated with wet deposition estimates [*Prospero et al.*,

2010]. The rain rate over the ocean is very difficult to constrain as obviously one cannot "be there" to capture rain events which are patchy and episodic, and remote determinations from, for example, Microwave Imager (TMI) and precipitation radar (PR) suffer in accuracy [e.g., *Ikai and Nakamura*, 2003]. Here, we use the known flux of ^{7}Be from the ocean inventory to avoid the pitfalls associated with determination of these parameters so that the flux of a trace element "i" can be calculated. The ratio of the atmospheric flux of trace element "i" to that of ^{7}Be is:

$$\frac{\text{Total Atmospheric Flux}_{(I)} = Cp_i * [R \cdot S \cdot \rho + Vd]_i}{\text{Total Atmospheric Flux}_{(7Be)} = Cp_{Be} * [R \cdot S \cdot \rho + Vd]_{Be}^7} \tag{2}$$

For ^{7}Be, S is large (on the order of 400–600) and Vd is small (on the order of 0.1 cm/s), so wet deposition dominates the ^{7}Be flux [e.g., *Kadko and Prospero*, 2011]. For mineral dust, S is smaller (150–300) but Vd is larger (~1cm/s). Assuming therefore that the right-hand terms in brackets roughly cancel:

$$\text{Total Atmospheric Flux}_{(i)} \cong \text{Total Atmospheric Flux}_{(7Be)} \cdot \frac{(Cp_i)}{(Cp_{Be})} \tag{3}$$

The term Flux$_{7Be}$ is derived from the integrated ^{7}Be (Σ^7Be) of the water column (described in section 2.1), and Cp$_{Be}$ and Cp$_i$ are the measured ^{7}Be activity and trace element concentrations, respectively, on aerosols, such that:

$$\text{Total Atmospheric Flux}_{(I)} \cong \left[\text{Ocean}\Sigma^7\text{Be} \right] \cdot \frac{(Cp_i)}{(Cp_{Be})} \tag{4}$$

This approach is analogous to the use of ^{234}Th as a tracer of ocean particle flux to estimate particulate organic carbon export from the surface ocean which requires determination of both the ^{234}Th activity distribution (in order to calculate ^{234}Th fluxes) and an estimate of the C$_{org}$/^{234}Th ratio on sinking particles, to derive C fluxes [e.g., *Buesseler et al.*, 2006]:

$$\text{Flux}_{carbon} = \text{Flux}_{234Th} \cdot \frac{[C_{org}]_{particle}}{[^{234}\text{Th}]_{particle}} \tag{5}$$

We now present data which indicate that the assumptions inherent in equation (4) are reasonable, i.e., that estimates of the atmospheric flux of trace elements (TE) can be made by multiplying the ocean inventory of ^{7}Be by the [TE/^{7}Be] ratio in bulk aerosols. It has already been demonstrated that a necessary condition of this assumption, that the ^{7}Be flux can be estimated by the ocean ^{7}Be inventory, is valid [*Aaboe et al.*, 1981; *Kadko and Prospero*, 2011]. Additional data are presented below. The other condition is that TE/^{7}Be ratios measured on aerosols are indicative of those on particles crossing the air-sea interface. This is tested by comparing fluxes derived from equation (4) to direct flux measurements made on the nearby island of Bermuda.

5. Results

The ^{7}Be inventories collected during 2011–2013 are shown in Table 1. They average 39,600± 10,800 dpm/m^2. These are plotted, together with the inventories collected from earlier work (2007–2008) [*Kadko and Prospero*, 2011], in Figure 1. The average of all inventories (2007–2008; 2011–2013), is 37,800 ± 8,600 dpm/m^2 (equivalent, at steady state, to a flux of 490 ± 100 dpm/m^2/d). Generally, the inventories from Hydrostation S and BATS are quite similar for any sampling period, with one exception occurring in May 2012 when the respective profiles had different T-S characteristics. Also shown are the average measured ^{7}Be rain flux on Bermuda during 2007–2008 and the running (2 month) average ^{7}Be rain flux from 2008 [*Kadko and Prospero*, 2011]. The rain flux (right axis) corresponds to the ocean inventory (left axis) that the flux would support at steady state. The average ^{7}Be rain flux measured on Bermuda during 2007–2008 matches the average ocean ^{7}Be inventory. For the 2011–2013 sampling period, the Bermuda rain flux was 350 dpm/m^2/d. While within 30% of the ocean inventory, the data are biased to lower values as the sampling started and ended during the summer months when the rain flux has been shown to be lower. Also, there was anomalously low precipitation, and corresponding low ^{7}Be flux in the spring of 2012; however, there was no concurrent ocean inventory measured at that time with which to compare the rain flux.

Table 1. [7]Be Inventories From BATS and Hydrostation "S" 2011–2013[a]

Date	Site S (dpm/m^2)	BATS (dpm/m^2)
18 Aug 2011		35,775
20 Aug 2011	37,796	
31 Oct2011	35,253	
2 Nov 2011		37,468
10 Feb 2012		39,764
20 May 2012		60,379
23 May 2012	45,592	
21 Aug 2012		49,635
22 Aug 2012	46,528	
4 July 2013	23,418	
8 July 2013		23,725

[a]BATS (31°50′N, 64°10′W), Hydrostation S (32°10′N, 64°30′W).

Figure 1 also shows the monthly average rainfall for Bermuda. The seasonal variation in rainfall is reflected in the ocean [7]Be inventories which nonetheless generally agree to within ∼20% of the annual average. It has been shown that the [7]Be ocean inventory calculated from progressive input and decay of the measured rain flux generally predicts the observed ocean inventory [*Kadko and Prospero*, 2011].

The [7]Be activities measured on aerosols are presented in Figure 2. Our results are similar to those of *Arimoto et al.* [1999], showing a moderate seasonality with a July–August minimum associated with lower altitude SE air mass trajectories. This corresponds to the summer African dust injection that extends across the Atlantic and is evident in the aerosol iron concentrations and the Fe/[7]Be ratios (Figure 2). Our aerosol TE concentrations, especially the pronounced seasonality in aerosol Fe concentrations, agree well with other published data for Bermuda and the Sargasso Sea [e.g., *Duce et al.*, 1976; *Arimoto et al.*, 2003; *Tian et al.*, 2008; *Sholkovitz and Sedwick*, 2006; *Shelley et al.*, 2012] as do our rainfall TE concentrations from this area [e.g., *Church et al.*, 1984; *Kim et al.*, 1999; *Kieber et al.*, 2003].
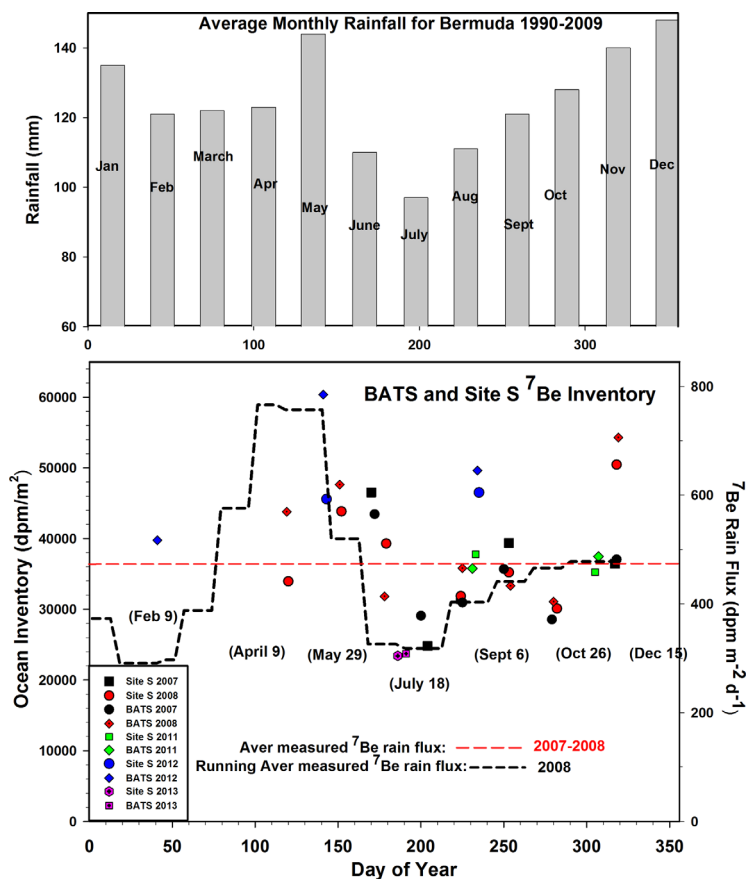

Figure 1. (bottom) The [7]Be ocean inventories collected during 2007–2008 (red and black symbols) [*Kadko and Prospero*, 2011] and 2011–2013 (green, blue and purple symbols, this work). Also shown are the average measured [7]Be rain flux during 2007–2008 and the running average [7]Be rain flux from 2008 [*Kadko and Prospero*, 2011]. The rain flux (right axis) corresponds to the ocean inventory (left axis) the flux would support at steady state. The average measured [7]Be rain flux matches the average ocean [7]Be inventory (see text). (top) The average monthly rainfall from Bermuda 1990–2009 (Climatic Research Unit of University of East Anglia).
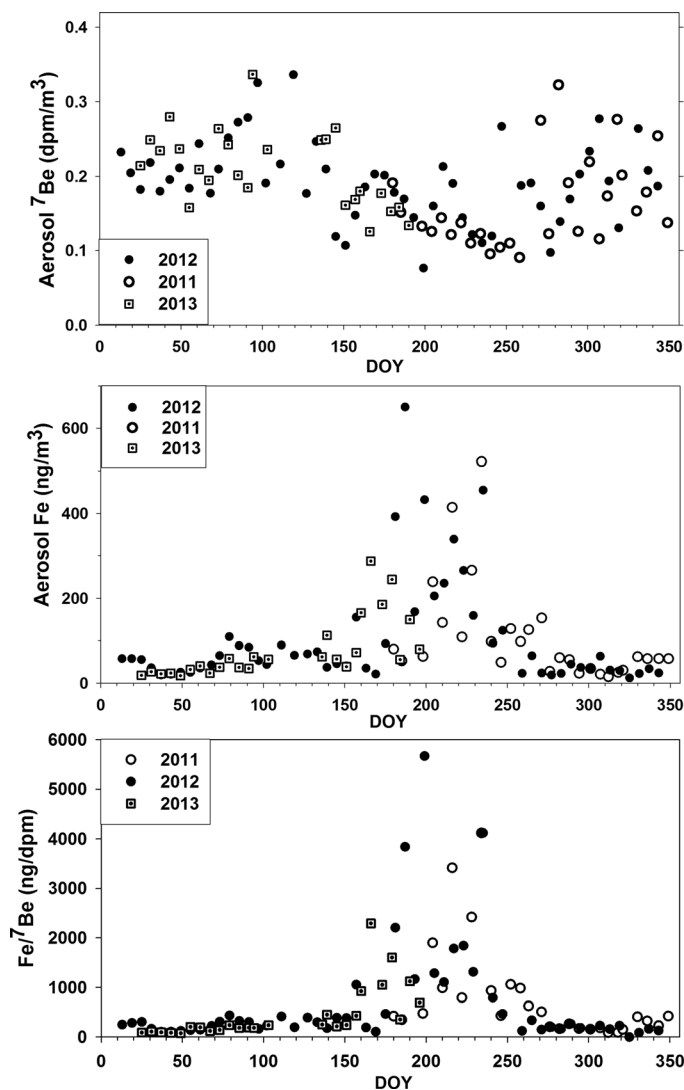
The weekly atmospheric deposition of Fe for 2011–2013, calculated from the yearly ^{7}Be flux (determined from the average ocean inventory; 490 dpm/m^2/d) multiplied by Fe/^{7}Be ratios in aerosols is presented in Figure 3. The integration of the weekly Fe flux over the full 24 month sampling period yields 94 mg (1680 μmol)/m^2/yr. Annual fluxes for other important bioactive trace elements (Mn, Co, Ni, Cu, Zn, Cd, and Pb) are shown in Table 2. The calculation based on the yearly average ^{7}Be flux is compared to that using individual ocean inventories applied to the Fe/^{7}Be ratio from the actual time interval when they were collected. Little difference is seen in the results (Figure 3). Similarly, the calculation based on seasonal (90 day period) averages applied to the corresponding Fe/^{7}Be ratios is shown (Figure 3). Because the ocean inventory does not vary much during the year, both cases show that flux variation derives mainly from seasonal differences in the Fe/^{7}Be ratio (Figure 3).

Figure 2. Aerosol data for the period 26 June 2011 to 27 July 2013. (top) ^{7}Be activities. (middle) Aerosol Fe concentrations. (bottom) Aerosol Fe/^{7}Be ratios. Strong seasonality is observed, particularly for aerosol Fe concentrations. Saharan dust arrives in July/August with SE trajectories.

In Table 2, the results of these flux calculations using the ^{7}Be method are compared to the independently derived flux measurements based on our aerosol and rainfall sampling at the BIOS station on Bermuda. We divide our 2 year sampling period into 12 month periods (July 2011–June 2012 and July 2012–June 2013), and also present the fluxes integrated over the entire 24 month sampling period. The TE fluxes based on the ^{7}Be method are shown first, and were calculated by summing the weekly fluxes obtained using the aerosol TE and ^{7}Be data for each sampling period and equation (4) (with a ^{7}Be decay inventory of 490 dpm/m^2/d). The TE fluxes from wet deposition were calculated by summing the rainfall TE "wet-only" fluxes over each 12 month period. The dry deposition fluxes were calculated by multiplying the aerosol TE concentrations by a dry deposition velocity of 1000 m/d (equivalent to 1.16 cm/s) and summing the weekly fluxes over each 12 month period. Using AEROCE data from the Bermuda West (Tudor Hill) Tower, *Arimoto et al.* [2003] demonstrated that aerosol TEI dry deposition fluxes could be apportioned into a recycled "sea salt" component (with a deposition velocity on the order of 3 cm/s), a crustal component (with a deposition velocity of 1 cm/s), and a "non-sea-salt/non-crustal" component (with a deposition velocity of 0.1 cm/s). We have data for aerosol Fe and Zn in common with the data set used by *Arimoto et al.* [2003], and the impact of using varying dry deposition velocities will be addressed below. Finally, in Table 2, the measured Wet Deposition and calculated Dry Deposition are summed and divided by the fluxes calculated using the ^{7}Be method
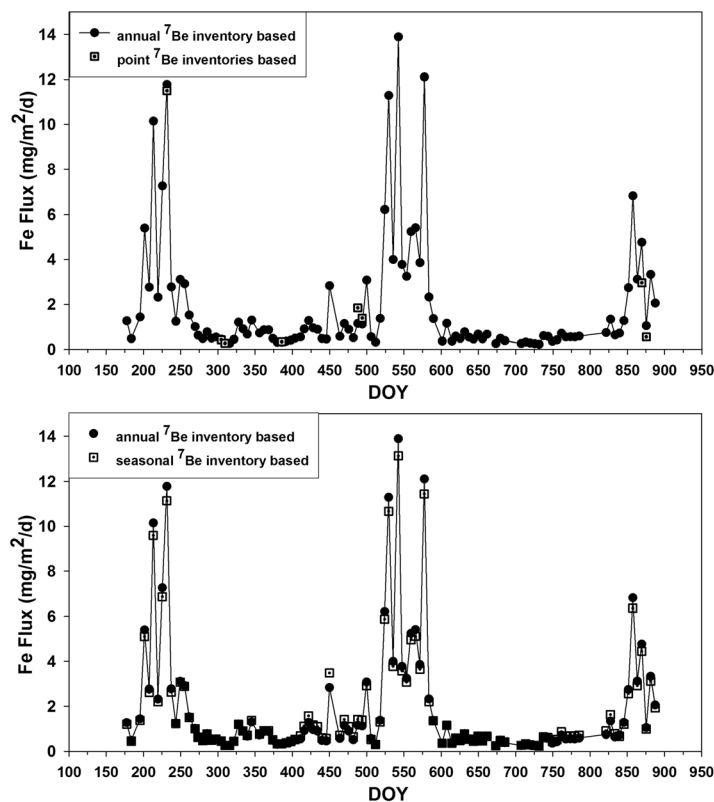
Figure 3. (top) The atmospheric deposition of Fe for 2011–2013, calculated from the yearly ^{7}Be flux (determined from the average ocean inventory; 490 dpm/m^2/d) multiplied by Fe/^{7}Be ratios in aerosols. This is compared to the atmospheric deposition calculated from the ^{7}Be flux determined from individual ocean inventory measurements (open squares). (bottom) The atmospheric deposition of Fe for 2011–2013 calculated from the yearly ^{7}Be flux compared to that calculated from the ^{7}Be flux determined from seasonally averaged ocean ^{7}Be inventory measurements (open squares). Because the ocean inventory does not vary much during the year, both cases show that flux variation derives mainly from seasonal differences in the Fe/^{7}Be ratios.

(equation (4)) to demonstrate how well these more "traditional methods" for estimating atmospheric TE fluxes agree with the ^{7}Be method.

For Mn, while the measured wet deposition fluxes agreed very well for both years (and also with previously published wet-only fluxes of 8–10 μmol/m^2/yr; *Kim et al.* [1999]), the aerosol Mn/^{7}Be ratios were slightly higher in Year 2 resulting in slightly higher fluxes. Overall, the "traditional method" yielded a total atmospheric flux that was 74% of the flux we obtained from the ^{7}Be method, and aerosol dry deposition accounted for ∼ 44 % of the total (wet + dry) amount.

The aerosol Fe concentrations (and Fe/^{7}Be ratios) were slightly higher in Year 2, yielding a slightly higher dry deposition flux and a slightly higher flux using the ^{7}Be method. Wet deposition for Fe was significantly higher in Year 1 due to the capture of the highest aerosol Fe concentrations by concurrent rain events (4–25 August 2011). The "traditional method" yielded fluxes that ranged from 41 to 58% of the fluxes derived using the ^{7}Be method, and dry deposition accounted for 63% of the total (wet + dry) atmospheric Fe flux (comparable to the 70% estimated by *Tian et al.* [2008]). Our measured wet deposition agrees well with the 1982 and 1996 wet-only Fe fluxes reported by *Kim et al.* [1999] and our estimated *aerosol* Fe dry deposition (529 μmol/m^2/yr) is within the interannual variability of the 1100 (±∼50%) μmol/m^2/yr reported by *Arimoto et al.* [2003] based on AEROCE data (1988–1993) from the Bermuda West (Tudor Hill) Tower. *Arimoto et al.* [2003] also concluded that the aerosol Fe dry deposition flux was dominated by mineral dust (95%), such that a single dry deposition velocity of 1 cm/s was appropriate to use.

Shelley et al. [2012] reported aerosol Co concentrations from summer 2003 (400–1000 pg/m^3; a "high dust" year) and summer 2008 (4–11 pg/m^3; a "low dust" year) near the BATS station in the Sargasso Sea. We measured summertime aerosol Co concentrations at the BIOS site ranging from 130 to 300 pg/m^3, suggesting that our sampling period reflected an "intermediate dust" condition during both summers. All of the Co fluxes were similar for both years, and the "traditional method" yielded a total flux that was 63% of the flux estimated using the ^{7}Be method, and dry deposition accounted for 52% of the total.

For Ni, Cu, and Cd, the aerosol concentrations and TEI/^{7}Be ratios were both lower in Year 2 while the wet deposition was higher. Our wet deposition Ni and Cu fluxes are both within the ranges reported by *Kim et al.* [1999] while our wet deposition Cd flux was much lower. Overall, the "traditional method" yielded total fluxes that were 64%, 53%, and 81% of the fluxes estimated using the ^{7}Be method for Ni, Cu, and Cd, respectively. Dry deposition accounted for 55%, 62%, and 46% of the total deposition for Ni, Cu, and Cd, respectively.

Table 2. Comparison of Collection-Based and ^{7}Be Method Annual TEI Atmospheric Deposition Fluxes (μmol m^{-2}y^{-1})[a]

Year 1 Data: July 2011–June 2012	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
^{7}Be method	30.2	1570	0.944	14.7	38.2	28.1	0.157	1.64
Measured wet deposition	14.0	414	0.283	1.75	3.98	9.90	0.035	0.540
Calculated dry deposition (V_d=1.16 cm s^{-1})	9.93	496	0.308	5.12	11.9	10.4	0.056	0.623
Total (wet+dry)/^{7}Be method	79%	58%	63%	47%	42%	72%	58%	71%
Year 2 data: July 2012–June 2013	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
^{7}Be method	38.0	1790	0.860	10.3	22.0	29.6	0.096	1.60
Measured Wet deposition	13.7	175	0.265	4.96	7.63	21.2	0.073	0.520
Calculated Dry deposition (V_d=1.16 cm s^{-1})	12.6	563	0.279	3.55	8.08	11.1	0.038	0.575
Total (wet+dry)/^{7}Be method	69%	41%	63%	83%	71%	109%	116%	68%
24 Month Weighted Average: July 2011–June 2013	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
^{7}Be method	34.1	1680	0.902	12.5	30.1	28.9	0.127	1.62
Measured Wet deposition	14.1	308	0.274	3.61	6.09	16.4	0.056	0.540
Reported Wet Deposition [*Kim et al.*, 1999]	7.5–10	130–250		1.4–6.0	1.6–16	21–28	0.2–0.8	0.3–7.0
Calculated Dry deposition (V_d=1.16 cm s^{-1})	11.2	529	0.293	4.34	9.98	10.8	0.047	0.599
Reported Dry Deposition [*Arimoto et al.*, 2003]		1100				3		
Total (wet+dry)/^{7}Be method	74%	50%	63%	64%	53%	94%	81%	70%

[a]Three significant figures are shown for convenience.

For Zn, the aerosol Zn concentrations and Zn/^{7}Be ratios were comparable in both years, while the wet deposition was higher in Year 2 (but still within the range reported by *Kim et al.* [1998]). Overall, the "traditional method" yielded a total flux that was 94% of the flux estimated using the ^{7}Be method, and dry deposition accounted for 40% of the total. *Arimoto et al.* [2003] reported that 3% of the aerosol Zn concentration at Bermuda was associated with sea-salt particles (Vd=3 cm/s), 14% was associated with mineral dust (Vd=1 cm/s), and 83% was associated with "non-sea-salt/non-crustal" aerosols (Vd=0.1 cm/s). If we use the same apportionment, and apply the same three dry deposition velocities to our aerosol Zn data, we calculate an aerosol Zn dry deposition flux (3.0 μmol/m^2/yr) that agrees with the flux calculated by *Arimoto et al.* [2003], yet is a factor of 3.6 lower than the flux of 10.8 μmol/m^2/yr calculated using a single Vd=1.16 cm/s, shown in Table 2. It is important to point out that the aerosol Zn concentrations we measured from samples collected at the BIOS biostation agree very well with those collected from the Tudor Hill sampling tower (reported by *Arimoto et al.*, 2003, and *Duce et al.*, 1976], and they also agree with our preliminary data from a subset of aerosol samples that were collected on the tower (using sector control) concurrently with the samples collected at the biostation. In other words, there is no evidence that the aerosols collected at the bio-station are contaminated relative to those collected on the tower. The aerosol Zn dry deposition flux we report in Table 2 is 3.7 times higher than the flux calculated by *Arimoto et al.* [2003] simply because we used a single dry deposition velocity (1000 m/d) while *Arimoto et al.* [2003] apportioned the aerosol Zn into three size classes with different deposition velocities. If we use the same approach (yielding an aerosol Zn dry deposition flux of 2.9 μmol/m^2/yr) the "traditional method" yields a total flux that is 67% of the flux calculated using the ^{7}Be method, and dry deposition accounts for 15% of the total.

All of the atmospheric Pb fluxes agreed well between Year 1 and Year 2, and the Pb wet deposition was well within the range reported by *Kim et al.* [1998]. Overall, the "traditional method" yielded a total flux that was 70% of the flux estimated using the ^{7}Be method, and dry deposition accounted for 53% of the total.

Overall, the sum of the measured wet deposition plus the estimated dry deposition is lower than the annual flux calculated using the ^{7}Be method by 6–50%. Particularly for those elements associated with coarser mineral grains (Mn, Fe, Co), the annual wet plus dry deposition flux would match the annual flux from the ^{7}Be method if the dry deposition velocity was closer to 2000 m/d (2.31 cm/s) which is not an unreasonable value and within the uncertainty of this parameter. The differences could also be accounted for if the ratio of the right hand terms (in brackets) in equation (2) ranged from 0.83 to 0.71, instead of 1.0 (as was initially assumed). There are four "unknowns" in the right hand ratio; the rainfall scavenging ratios for a given trace element and for ^{7}Be, and the dry deposition velocities for a given trace element and for ^{7}Be. While these parameters can be constrained to a first approximation (section 4.0), the estimated values for these parameters all range over a factor of 2 or more. Considering Fe for example, using Vd = 1000 m/d and S = 200, and for ^{7}Be, Vd=86 m/d and S = 500, then the right hand ratio is 0.86 for the average Bermuda rainfall rate (0.004 m/d).

Either factor (higher dry deposition velocity for aerosol trace elements or nonunity in the ratio of the right hand terms in equation (2)) can account for the relatively modest disagreement between the annual trace element fluxes determined by these two methods (Table 2). Regardless of this conundrum, we conclude that the [7]Be method allows one to calculate the total annual deposition for aerosol trace elements, with an error on the order of less than 50%, without relying on quantification of the depositional parameters. This degree of uncertainty is relatively small, and demonstrates the utility of the [7]Be method for estimating the deposition of aerosol trace elements to the oceans.

6. Conclusions

Given the relatively low seasonal variability in the aerosol [7]Be concentrations over the Sargasso Sea, the atmospheric flux of [7]Be is roughly equivalent to the [7]Be ocean decay inventory. This observation provides a key linkage between the atmospheric concentrations of chemical species and their deposition to the oceans. In the subtropical N. Atlantic, the ocean inventory of [7]Be is relatively constant, such that a single profile represents the average atmospheric flux to within ∼20%, whereas the aerosol TE/[7]Be ratios may experience greater variability due to seasonal differences in rainfall rates and in air masses reaching Bermuda. We have demonstrated that estimates of the atmospheric fluxes of trace elements can be made by multiplying the ocean inventory of [7]Be by the [TE/[7]Be] ratio in bulk aerosols. Flux estimates for trace elements made by the [7]Be ocean inventory method are comparable (to within 50% or better) to fluxes derived from measured wet deposition plus estimated dry deposition for samples collected on the island of Bermuda.

To estimate the atmospheric flux of trace element to the oceans on time scales on the order of weeks to months, it is only necessary to characterize the aerosol TE/[7]Be ratios on those same time scales. To estimate the annual trace element deposition in oceanic regions where one expects strong seasonal variability in aerosol TE/[7]Be ratios, for example downwind of continental mineral dust sources, it would be necessary to collect enough aerosol samples to characterize the annual "average" TE/[7]Be ratios. This would require seasonal sampling of aerosols augmented perhaps with satellite tools such as the Total Ozone Mapping Spectrometer (TOMS) which measures the relative amount of aerosols in the atmosphere.

The situation at Bermuda allowed testing of our ocean-based method through comparison with convenient land locations. Our results suggest that this method would be useful for remote areas where fixed sampling stations do not exist; that is, the majority of the global ocean.

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