

Deposition of ^7Be to Bermuda and the regional ocean: Environmental factors affecting estimates of atmospheric flux to the ocean

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[1] The distribution of ^7Be in ocean surface waters is used as tracer of upper ocean transport and atmospheric deposition processes. However, there is very little ocean deposition data available to characterize the temporal and spatial input of ^7Be to the oceans and test model results. Here we measure the deposition of ^7Be in bulk collectors at two sites at Bermuda over a span of nearly 2 years (April 2007 to January 2009) and compare these rates to the flux required to sustain the inventory of ^7Be measured in the nearby Sargasso Sea. The Tudor Hill collector site undersampled (by ~40%) both the rainfall compared to other Bermuda sites and the ^7Be flux required for the ocean inventory. On the other hand, the ^7Be flux captured at the Bermuda Institute of Ocean Sciences station site ($0.048 \text{ dpm cm}^{-2} \text{ d}^{-1}$) matched that expected from the ocean observations. Previously measured long-term atmospheric concentration of ^7Be in surface air at Bermuda was used to estimate deposition velocities and scavenging ratios, and our estimates in this marine environment were found to be similar to those measured in continental regions. The deposition of ^7Be to the oceans is overwhelmingly determined by wet processes; dry deposition to the ocean surface accounts for only a few percent, at most, of the total deposition to the ocean. We place these measurements in a longer-term and large-scale spatial context by using climatological rainfall data on Bermuda and ocean rainfall estimates from the Global Precipitation Climatology Program and Tropical Rainfall Measuring Mission.

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1. Introduction

[2] The atmospheric input of numerous chemical species into the global ocean equals or exceeds that from river sources [Duce *et al.*, 1991]. Assessment of this input is difficult, however, because measurements of deposition rates to the ocean are rare. Furthermore, such measurements are susceptible to problems associated with temporal and spatial variability of precipitation and of the concentration of species within precipitation. Natural radionuclides delivered to the ocean from the atmosphere are useful tracers for characterizing global-scale transport and deposition. For example, the global distribution of ^7Be 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 aerosol species including those from anthropogenic sources.

[3] The accumulation of ^7Be in the upper ocean provides a means of assessing ^7Be deposition on a global scale. The

ocean inventory of ^7Be is largely based on measurements of the vertical distribution of ^7Be in the oceans, most notably in the work of Young and Silker [1980], who presented data from a large number of globally distributed ocean sites. These data have been widely used to constrain the estimates of ^7Be deposition rates to the ocean. More recently, ocean profiles have also been measured in the northeastern Atlantic [Kadko and Olson, 1996] and the western Arctic [Kadko, 2000; Kadko and Swart, 2004]. There have been very few measurements made of the deposition rates of ^7Be in rain at ocean sites [Turekian *et al.*, 1983; Kim *et al.*, 1999]; studies at coastal sites are more common [e.g., Olsen *et al.*, 1985, 1986; Dibb, 1989; Todd *et al.*, 1989; Baskaran *et al.*, 1993]. To our knowledge, no concurrent measurements of ^7Be deposition with ocean profile measurements have been made prior to this study. It is well known that rainfall rates measured on islands are not necessarily representative of rainfall rates to the surrounding ocean, even at small Pacific atolls that are commonly used for satellite validation [Huffman *et al.*, 2007]. Consequently, the measured deposition rates of chemical species in island rain might not accurately reflect regional ocean deposition rates. To address this issue, we assess the atmospheric flux of ^7Be measured at two sites at Bermuda over a nearly 2 year period. These data were compared to the

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standing crop of ^7Be measured in the nearby Sargasso Sea. The objective was to determine how well the flux measured on the island matched that required to support the observed ocean inventory. This, by inference, provides a measure of how well the flux of other species measured in Bermuda rainfall reflects the actual oceanic deposition to the nearby ocean. We extend the analysis of this short-term data set by placing it in the context of long-term rainfall measurements on Bermuda and on the regional ocean using satellite rainfall data sets. Finally, to place our measurements in a global context, we compare the ^7Be measurements and deposition parameters on Bermuda with those made in other (mostly continental) regions.

2. Background ^7Be

[4] Be-7 is a cosmic ray-produced isotope (half-life of 53.3 days) that is deposited on the ocean surface primarily by rainfall and is 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]. In the low-particle environment of the open ocean, particularly within the oligotrophic regime of the Sargasso Sea, ^7Be is considered conservative with respect to particle scavenging removal [e.g., *Silker*, 1972; *Andrews et al.*, 2008].

[5] While most of the production of ^7Be occurs in the stratosphere, the long residence time of aerosols in the stratosphere relative to the short half-life of ^7Be dictates that the tropospheric production of ^7Be determines the flux to the Earth's surface, although intrusions of stratospheric air into the troposphere are, at times, also important [*Feely et al.*, 1989]. Therefore, over broad oceanic regions, the ^7Be flux and water column inventory vary as a function of rainfall, and, consequently, inventories in ocean surface waters are inversely related to salinity [*Young and Silker*, 1980; *Kadko and Olson*, 1996]. The water column inventory represents an integration of the flux over approximately the previous mean-life (77 days) of the isotope, assuming that advective processes represent a small component of the isotopic budget. While advective effects cannot be discounted from individual ocean profiles, long-term averages as compiled in this study likely mitigate the effect of these complications.

3. Methods

3.1. Atmospheric Deposition Measurements

[6] The ^7Be flux was determined at two sites on the island of Bermuda. One was atop the 23 m sampling tower at Tudor Hill (TH), located on a steep slope at the extreme western end of Bermuda ($32^\circ 15.95'\text{N}$, $64^\circ 52.65'\text{W}$) with the tower base 28 m above sea level. The second collection site was established at the Bermuda Institute of Ocean Sciences (BIOS) located 21 km northeast of TH, close to the eastern end of the island ($32^\circ 22.28'\text{N}$, $64^\circ 41.80'\text{W}$). The collector was situated near ground level, clear of surrounding vegetation, at a meteorologic 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.

[7] To estimate the atmospheric deposition of ^7Be , plastic buckets (660.5 cm^2) were deployed for collection of fallout over known time periods. Typically, collection times were

about 3 weeks but varied as a function of rainfall, i.e., were longer when rainfall was less abundant. After collection, dilute HCl and stable Be yield tracer were added to the buckets. The ^7Be was then coprecipitated with iron hydroxide, and the precipitate was dried in plastic petri dishes. The sample was subsequently counted by gamma spectrometry. The counting system was calibrated for all samples by preparing a commercial standard in a geometry identical to the samples. The stable Be in the precipitate was then measured by atomic absorption to calculate the ^7Be recovery (generally $\sim 70\%$) during precipitation.

[8] At both sites, rainfall was monitored by rain gauges (model 03319; Cole Parmer Instrument Co.), a cylindrical tube with a 10 cm diameter knife-edge opening.

3.2. Measurement of ^7Be Profiles at Ocean Stations

[9] The Bermuda deposition results were compared to the ocean inventory determined from ^7Be profiles, collected monthly, from June–November 2007 and April–November 2008 in the vicinity of the U.S. Joint Global Ocean Flux Study Bermuda Atlantic Time-series Study (BATS) ($31^\circ 50'\text{N}$, $64^\circ 10'\text{W}$) and hydrostation S ($32^\circ 10'\text{N}$, $64^\circ 30'\text{W}$) in the Sargasso Sea. Samples were collected from the R/V *Atlantic Explorer* and subsequently processed by methods described in detail elsewhere [*Kadko*, 2009]. Briefly, ^7Be was collected at various depths by pumping 400–700 L of seawater via a 1.5 inch 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. 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 ^7Be concentrations were expected (typically at depths greater than 190 m), two samples were taken from the same depth to assure adequate signal, and the ashed fibers were combined and placed in Marinelli beakers to accommodate the large ash volume. For such samples, up to $\sim 1200\text{ L}$ were analyzed.

[10] The ^7Be has a readily identifiable gamma peak at 478 keV. 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. Results

[11] The ^7Be flux measurements taken on Bermuda are shown in Table 1. Sampling began at the TH site in April 2007 in anticipation of the collection of oceanic ^7Be profiles initiated in June 2007. The measured ocean inventories, and the calculated steady state fluxes to support them, are shown in Table 2.

Table 1. ⁷Be Flux Measurements

Sample	Start Date	End Date	Rain Volume (mm)		Flux (dpm cm ⁻² d ⁻¹)	
			Tudor Hill	BIOS ^a	Tudor Hill	BIOS
F1	18 Apr 2007	8 May 2007	39.8	49	0.0207 ± 0.0006	
F2	8 May 2007	14 May 2007	72.2	106.4	0.1417 ± 0.0067	
F3	14 May 2007	21 May 2007	106.1	108.7	0.0705 ± 0.0019	
F4	22 May 2007	30 May 2007	39.9	42.9	0.0127 ± 0.0011	
F5	30 May 2007	18 Jun 2007	74.6	97.8	0.02845 ± 0.0013	
F6	18 Jun 2007	25 Jun 2007	21.8	33.3	0.01235 ± 0.0011	
F7	25 Jun 2007	3 Jul 2007	60.5	52.1	0.0487 ± 0.0030	
F8	3 Jul 2007	23 Jul 2007	28.9	24.9	0.0210 ± 0.0007	
F9	23 Jul 2007	7 Aug 2007	137.7	244.3	0.0593 ± 0.0018	
F10	7 Aug 2007	16 Aug 2007	16.1	38.9	0.0157 ± 0.0011	
F12	16 Aug 2007	28 Aug 2007	31.2	54.9	0.0297 ± 0.0019	0.0332 ± 0.0023
F13	28 Aug 2007	3 Sep 2007	34.1	27.2	0.0580 ± 0.0057	0.1069 ± 0.0072
F15	3 Sep 2007	18 Sep 2007	10	42.4	0.0088 ± 0.0008	0.0075 ± 0.0010
F17	18 Sep 2007	2 Oct 2007	54.2	79.8	0.0701 ± 0.0020	0.1127 ± 0.0065
F20	2 Oct 2007	15 Oct 2007	115.8	99.8	0.0228 ± 0.0022	0.0560 ± 0.0039
F22	15 Oct 2007	30 Oct 2007	57.6	68.58	0.0176 ± 0.0015	0.0176 ± 0.0014
F24	30 Oct 2007	12 Nov 2007	46.8	105.4	0.0434 ± 0.0038	0.0850 ± 0.0059
F26	12 Nov 2007	3 Dec 2007	48.4	45.9	0.0300 ± 0.0020	0.0468 ± 0.0035
F28	3 Dec 2007	18 Dec 2007	53.2	26.9	0.0365 ± 0.0013	0.0731 ± 0.0023
F30	18 Dec 2007	7 Jan 2008	12.8	77.2	0.0370 ± 0.0005	0.0402 ± 0.0004
F32	7 Jan 2008	28 Jan 2008	28.4	127.3	0.0132 ± 0.0003	0.0252 ± 0.0004
F34	28 Jan 2008	18 Feb 2008	22.9	42.4	0.0056 ± 0.0002	0.0196 ± 0.0005
F36	18 Feb 2008	19 Mar 2008	45.7	81.5	0.0217 ± 0.0006	0.0449 ± 0.0010
F38	19 Mar 2008	27 Mar 2008	103.6	127	0.01745 ± 0.0012	0.0241 ± 0.0014
F39	27 Mar 2008	18 Apr 2008	68.1	93.98		0.0755 ± 0.0017
F40	18 Apr 2008	7 May 2008	150.2	186.18		0.1013 ± 0.0030
F41	7 May 2008	3 Jun 2008	228.5	209.8		0.0823 ± 0.0015
F42	3 Jun 2008	2 Jul 2008	13.2	17.2		0.0115 ± 0.0002
F43	2 Jul 2008	24 Jul 2008	70.3	211.8		0.0234 ± 0.0007
F44	24 Jul 2008	19 Aug 2008	82.8	124.5		0.0436 ± 0.0030
F45	19 Aug 2008	18 Sep 2008	51.2	90.9		0.0452 ± 0.0020
F46	18 Sep 2008	13 Oct 2008	93.9	128.5		0.04085 ± 0.0013
F47	13 Oct 2008	7 Nov 2008	72.5	85.34		0.0488 ± 0.0011
F48	7 Nov 2008	21 Nov 2008	31.4	40.1		0.0551 ± 0.0029
F49	21 Nov 2008	11 Dec 2008	18	32		0.0232 ± 0.0016
F50	11 Dec 2008	19 Dec 2008	30.2	66.8		0.0488 ± 0.0024
F51	19 Dec 2008	9 Jan 2009	14.2	44.45		no sample
F52	9 Jan 2009	29 Jan 2009	56.1	86.36		0.0572 ± 0.0031
Average					0.0302 ± 0.0223 ^b	0.0483 ± 0.0271 ^c

^aBermuda Institute of Ocean Sciences.^bFor 18 April 2007 to 27 March 2008.^cFor 16 August 2007 to 21 November 2008 (see section 4).**Table 2.** Water Column Inventories

	Site S		BATS	
	Inventory (dpm m ⁻²)	Flux ^a (dpm cm ² d ⁻¹)	Inventory (dpm m ⁻²)	Flux ^a (dpm cm ² d ⁻¹)
2007				
Jun	46,538	0.0605	43,469	0.0565
Jul	24,809	0.0323	29,104	0.0378
Aug	31,468	0.0409	30,964	0.0403
Sep	39,388	0.0512	35,698	0.0464
Oct			28,567	0.0371
Nov	36,449	0.0474	37,053	0.0482
2007 Average	35,730	0.0464 ± 0.0106	34,142	0.0444 ± 0.007
2008				
Apr	33,961	0.0442	43,795	0.0569
May	43,860	0.0570	47,635	0.0619
Jun	39,315	0.0511	31,835	0.0414
Aug	31,888	0.0415	35,814	0.0466
Sep	35,231	0.0458	33,322	0.0433
Oct	30,115	0.0392	31,084	0.0404
Nov	50,468	0.0656	54,314	0.0706
2008 Average	37,834	0.0492 ± 0.0094	39,686	0.0516 ± 0.012
2007–2008 Average	36,957	0.0481 ± 0.0100	37,127	0.0483 ± 0.0103

^aSteady state flux required to support observed inventory.

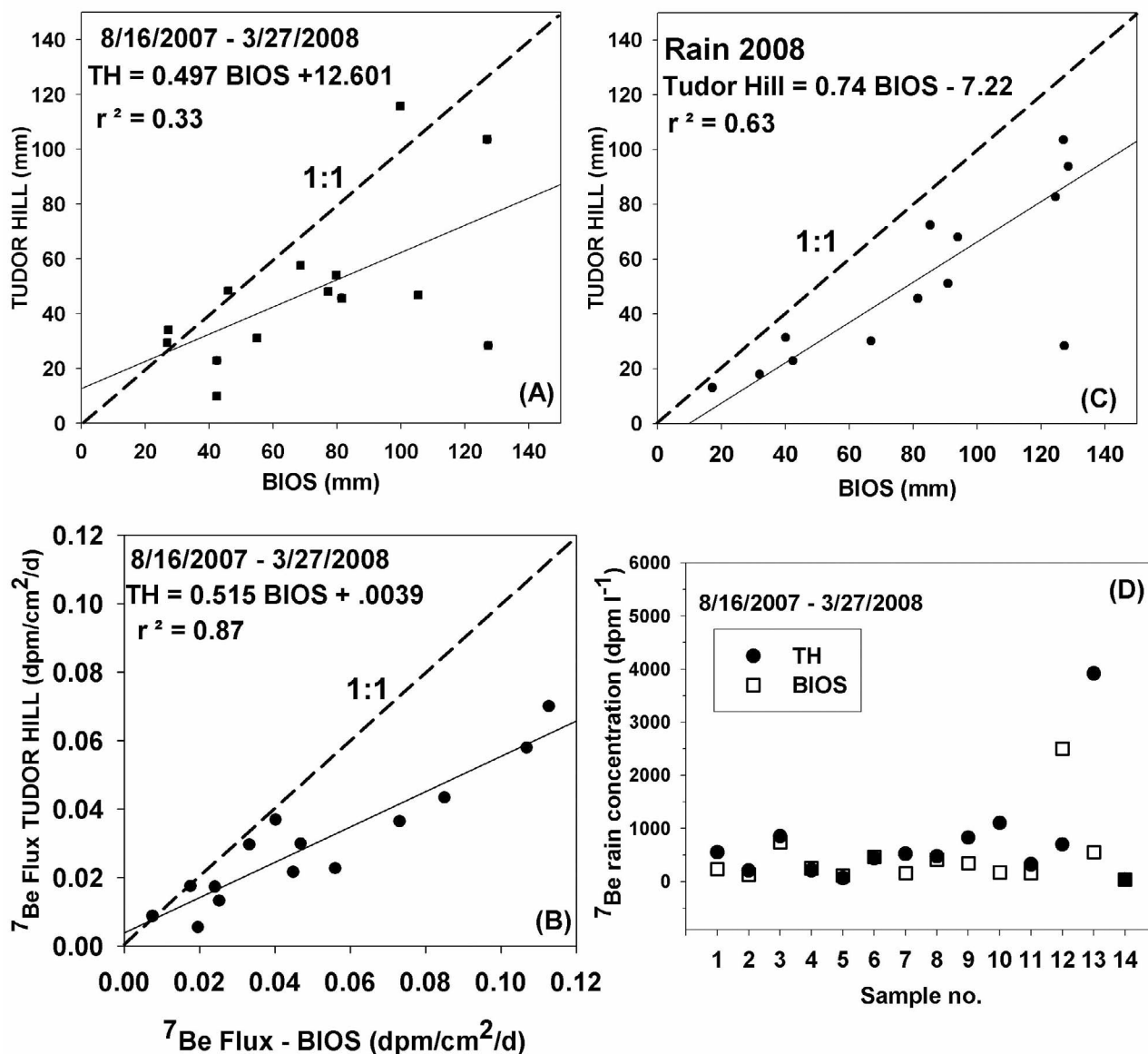


Figure 1. Rainfall and ^7Be comparison between the two flux measurement sites at Bermuda. Over the ^7Be flux intercomparison period (16 August 2007 to 27 March 2008), the Bermuda Institute of Ocean Sciences (BIOS) station site had higher (a) rainfall and (b) ^7Be flux than the TH site. (c) Over the year 2008, ~35% more rain was recorded at the BIOS location than the Tudor Hill site. (d) ^7Be concentrations for both sites plotted against sampling interval for the intercomparison period.

[12] The initial ocean sampling suggested a mismatch between the flux measured at TH and that required to sustain the observed ocean inventory. The average flux to the Sargasso Sea based on the ^7Be inventories at sites S and BATS for 18–20 June was $0.0585 \text{ dpm cm}^{-2} \text{ d}^{-1}$, while the average measured flux at TH from 18 April to 25 June was only $0.037 \text{ dpm cm}^{-2} \text{ d}^{-1}$. If the TH deposition rates were representative of the true oceanic deposition rate, this would have suggested a large advective input of ^7Be into the Sargasso Sea, with important implications for transport of nutrients and carbon into the region [e.g., Michaels *et al.*, 1994; Toggweiler, 1994]. To test the possibility that the TH flux measurements were undersampling the ^7Be flux, it was decided to perform parallel ^7Be measurements at an alternate

site on a nonelevated position on the BIOS campus. The two sites were sampled concurrently between 16 August 2007 and 27 March 2008. Figures 1a and 1b show that over this period, the flux and rainfall determined at BIOS were systematically greater than at the TH location. The deficit of ^7Be flux at TH relative to the BIOS site was nearly identical to the relative deficit in rainfall. For this period, the weighted mean flux at TH was $0.027 \text{ dpm cm}^{-2} \text{ d}^{-1}$, while at BIOS it was $0.0455 \text{ dpm cm}^{-2} \text{ d}^{-1}$, a factor of 1.69 greater than at TH. The rain at TH was 532 mm compared to 824 mm at BIOS, 1.55 greater than at TH. In 2008, BIOS recorded 1.35 times higher rain accumulation than TH for the entire year (Figure 1c). The similarity between the deficit of rain and flux over the intercomparison period suggests that the

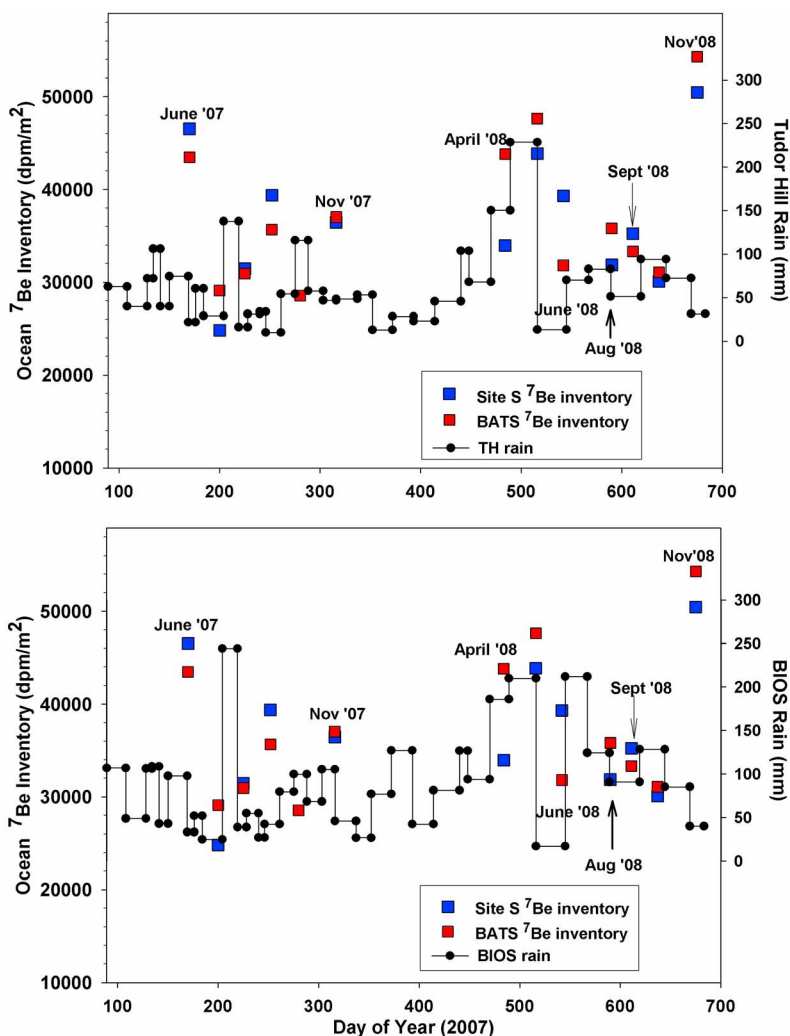


Figure 2. A comparison of the rainfall accumulation over each rainfall collection period (segments bracketed by black dots) and ocean inventory (red squares, Bermuda Atlantic Time-series Study (BATS); blue squares, site S). (top) Tudor Hill rain. (bottom) BIOS rain.

^7Be concentration in the rain for the two sites was similar (Figure 1d). The average oceanic ^7Be flux determined from the ocean inventory for the sampling period between June 2007 and November 2008 was $0.048 \pm 0.010 \text{ dpm cm}^{-2} \text{ d}^{-1}$, nearly identical for the two oceanic locations (Table 2). This matched the weighted (by collection time) average flux determined in precipitation at the BIOS site (16 August 2007 to 21 November 2008) of $0.048 \pm 0.027 \text{ dpm cm}^{-2} \text{ d}^{-1}$ (standard deviation of the mean). *Turekian et al.* [1983] previously reported a similar value for precipitation collected at the same site in Bermuda ($0.047 \text{ dpm cm}^{-2} \text{ d}^{-1}$). The weighted mean flux between the period 18 April 2007 and 21 November 2008, which brackets the ocean sampling period, was constructed as $(1.7 \times \text{TH flux (for the period 18 April 2007 to 16 August 2008)} + \text{BIOS flux (for the period 16 August 2008 to 21 November 2008)})$ and equals $0.051 \text{ dpm cm}^{-2} \text{ d}^{-1}$, which again, within the uncertainty of the measurements, would support the ocean inventory. The factor 1.7 applied to the TH result is derived from the entire intercomparison period between TH and BIOS. Figure 1c

shows that the difference in rainfall persisted over the entire year (2008); the rainfall accumulation for TH and BIOS was 996 and 1549 mm, respectively, yielding a relative deficit of 0.64 at TH. These results suggest the TH location was undersampling the rain and associated ^7Be flux and, by inference, would underestimate the delivery by rain of other atmospheric aerosol species to the ocean. In section 5.1 we discuss more fully the issues of rainfall amounts and their representativeness.

[13] Figure 2 shows a plot of the rainfall accumulation at BIOS and TH compared to the ocean inventories at sites S and BATS. There is a qualitative similarity between the rainfall at the two sites, and the inventories generally reflect the trends in rainfall. In Figure 3a, the running average of the flux measurements is seen to track the changes in ocean inventories over time. Note that for the TH data plotted here, a factor of only 1.4 is applied as was observed for this partial segment of the comparison period (16 August 2007 to 12 November 2007). In Figure 3b, the cumulative ^7Be ocean inventory expected from progressive input and decay of the measured

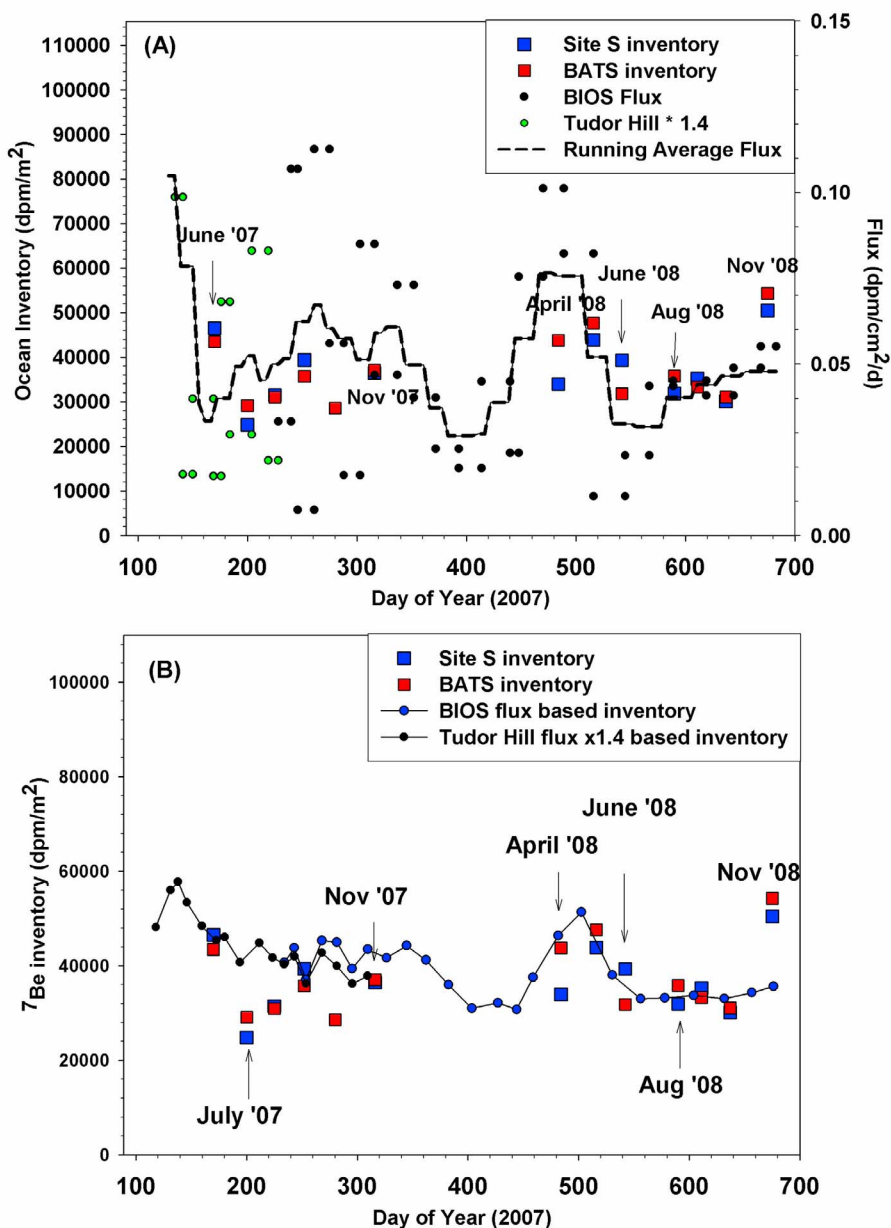


Figure 3. (a) Individual flux measurements (black dots, BIOS; green dots, Tudor Hill \times 1.4), ocean inventories (red squares, BATS; blue squares, site S), and the running average of the flux measurements. (b) Cumulative ^7Be ocean inventory expected from progressive input and decay of the measured flux.

flux is plotted [e.g., *Dibb*, 1989]. The calculated flux generally predicts the observed ocean inventory.

5. Discussion

[14] Our study shows that there is a substantial difference in the rainfall amounts and ^7Be deposition rates measured at the two deposition sampling sites, and that the rate at BIOS closely matches that necessary to sustain the water column inventory of ^7Be at the two ocean profile sites. To assess the significance of our measured rates relative to the ocean standing inventory, we must address several issues. The first is to establish how well our measured rain rates compare

with long-term rainfall measured at Bermuda. The second is to assess the representativeness of rainfall rates measured on Bermuda relative to that on the surrounding ocean. It is well known that islands generate their own climate and that, consequently, the measured rainfall may be biased. We consider these issues below. We also make use of earlier measurements of ^7Be air concentration on Bermuda to constrain various deposition parameters often used in estimating the deposition of aerosols to the oceans.

5.1. Historical Rainfall Measurement on Bermuda

[15] Rainfall rates are highly variable both spatially and temporally. To relate our measurements of ^7Be deposition as

Table 3. Bermuda Rainfall Estimates

Source	Location	Start Date	End Date	Amount (cm/yr)
Bermuda Meteorological Service ^a	airport	1949	2008	146
Playle	Somerset	1974	1999	152
Galloway, University of Virginia	Harbour Radio Tower	Apr 1980	Jun 1988	150
Galloway, University of Virginia	Tudor Hill	Aug 1989	Feb 1997	94
University of Miami	BIOS	Jan 2007	Jan 2009	146
University of Miami	Tudor Hill	Jan 2007	Jan 2009	108
Global Precipitation Climatology Project (GPCP) (V2.1) ^b	2.5° × 2.5° box	1998	2004	124
Tropical Rainfall Measuring Mission (TRMM) (3B-43) ^{b,c}	2.5° × 2.5° box	1998	2004	123

^aThis includes the data collected by the U.S. Navy (1949–1994).

^bThe GPCP and TRMM data are the average for a 2.5 × 2.5 grid box that contains Bermuda.

^cTRMM data multiplied by 1.16.

representative of Bermuda and the adjacent ocean, we need to place our measurements in a larger time-space context. To this end, we use rainfall data from various sources, summarized in Table 3. The annual averages are shown in Figure 4. The longest record of rainfall on Bermuda is that obtained by the U.S. Navy (USN) (1949–1994) at the airfield located immediately to the south of BIOS on St. David’s Island (32.36°N, 64.70°W). When the Navy departed, measurements were continued by the Bermuda Meteorological Service (BMS) at the same location, now the Bermuda International Airport. The average annual long-term (1949–2008) rainfall rate on Bermuda is 146 cm. In most months, the rates fall into a relatively narrow range, 11–13 cm/month, with a dry spell in April–May (~8 cm) and a sharp peak in October (16 cm/month).

[16] A long-term data set was also obtained by a weather enthusiast, B. Playle, who made systematic measurements from 1974 to 1999 and provided the data to the USN-BMS. The readings were taken in Somerset on the extreme western end of Bermuda at a site 3.2 km north of TH. As seen in Figure 4, the Somerset data closely track those at the USN-BMS site and suggest that rainfall is relatively uniform across the island. In another study, *Galloway et al.* [1993] presented a data set from an automatic collector started in April 1980 and ending in February 1997. Over the period April 1980 to June 1988, the collector was located at Harbour Radio Tower, Bermuda (HRT), 32°22’N/64°41’W, in St. George’s on the eastern end of the island a short distance from BIOS and the USN-BMS airport site. In August 1989, the collection site was moved to the atmo-

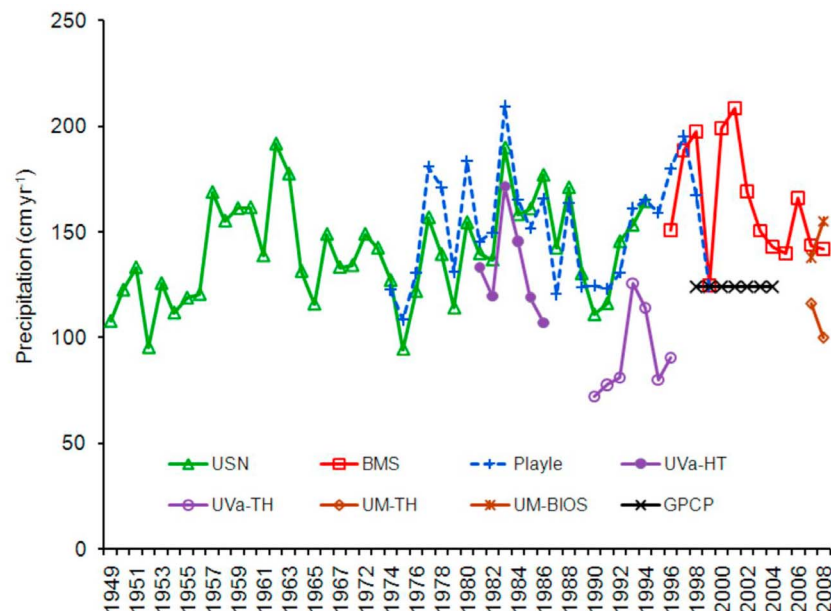


Figure 4. Precipitation measurements on Bermuda. Annual rainfall measurements made on Bermuda over the period 1949 to 2008 are summarized. USN, U.S. Navy at the air field; BMS, Bermuda Meteorological Service at the air field, now serving as the airport; Playle, a private individual residing in Somerset; UVa-HT, University of Virginia using a collector located at Harbour Tower; UVa-TH, University of Virginia using a collector located at Tudor Hill; UM-BIOS, University of Miami sampler at BIOS; UM-TH, University of Miami sampler at Tudor Hill; GPCP, Global Precipitation Climatology Project estimate of precipitation to Bermuda Region.

spheric chemistry station located at TH, the same location used for the University of Miami (UM) measurements made for this work, where it operated until February 1997. The data from these two sites are shown in Figure 4. There is a clear difference between the two sites. For the most part, the data at HRT closely track the USN data and the Playle data from Somerset; in contrast, the TH data fall well below these data. The mean annual rainfall at HRT was 150 cm/yr, and that at TH was 94 cm/yr. Thus the HRT rate is 1.6 times that at TH. Also shown in Figure 4 are the measurements made for this work (UM) at TH and BIOS for the years 2007 and 2008. The BIOS station yielded 146 cm/yr, very similar to that measured by BMS (~ 142 cm yr $^{-1}$) over the same period. For TH, the annual rate calculated in this manner is 108 cm, with the resulting ratio of BIOS to TH for the UM data at 1.35, comparable to the University of Virginia (UVa) ratio of HRT to TH data, which was 1.6.

[17] These diverse data sets lead to a wide range of estimates. One clear conclusion is that there is a substantial and consistent difference between measurements made at samplers located at TH by both UM and UVa and all other samplers on the island. Especially notable is the difference between measurements made at TH and those made at Somerset, 3.2 km to the north, and at sites in the eastern end of Bermuda (BIOS, Harbour Radio Tower, and airport) being about 40%–60% higher than the TH measurements. The UVa and UM measurements are quite consistent in this respect. This difference could be attributed to a number of causes. One could be related to a real difference in rainfall across the island. The other could be due to the location of the precipitation collector at TH, on the top of a 20 m tall tower placed on the side of a steep bluff. This site is often exposed to strong winds both because of its location on the coast and because of the effects of the hill on the wind at the site. Rain collector efficiency is very sensitive to wind velocity; to obtain accurate rainfall data, wind screens must be used [Sieck *et al.*, 2007; Strangeways, 2010].

[18] To place the Bermuda deposition data in a larger context, we compare our data with large-area data sets of ocean precipitation. The first is from the Global Precipitation Climatology Project (GPCP), a monthly $2.5^\circ \times 2.5^\circ$ latitude-longitude gridded data set [Adler *et al.*, 2003]. The product is based on a merged analysis that incorporates precipitation estimates from low-orbit satellite microwave data, geosynchronous-orbit satellite infrared data, and surface rain gauge observations. These show that Bermuda is located in a region where there is a relatively large synoptic scale gradient in rainfall rates with higher rates to the north and lower rates to the south. The GPCP (version 2.1) mean rainfall estimate for the years 1998–2004 obtained for the 2.5×2.5 box centered on Bermuda is 124 cm yr $^{-1}$ (D. Bolvin, Science Systems and Applications, Inc., NASA Goddard Space Flight Center, personal communication, 2010). The second large-scale precipitation data set is obtained from the Tropical Rainfall Measuring Mission (TRMM) [Huffman *et al.*, 2007]. The TRMM (3B-43) average across the same 2.5° box used in the GPCP estimate is 106 cm yr $^{-1}$. TRMM estimates are believed to be low by about 16% [Adler *et al.*, 2003]. If this correction is applied, the deposition estimate would be 123 cm yr $^{-1}$.

[19] The longer-term measurements made by the BMS, UVa at HRT, and the large-area estimates from GPCP and TRMM yield rates that fall into a rather narrow range, from

123 to 146 cm yr $^{-1}$, which exceeds that measured in all cases at TH. However, it should be noted that the GPCP value is the average for the years 1998–2004 when the measured rainfall rates on Bermuda were above average (Figure 4). Another problem with this comparison is that the GPCP data are averaged over a large area; as Bermuda is located in a region with a large rainfall gradient, the large averaging box could encompass a large range of rainfall rates that do not necessarily reflect rainfall on Bermuda. Nonetheless, the relatively close agreement between the GPCP and TRMM values and those measured on Bermuda provide some assurance that the measurements on Bermuda (excluding TH) are reflective in a broad way of deposition to the surrounding ocean.

5.2. Deposition Parameters

[20] The accurate assessment of atmospheric inputs to the oceans is difficult because of the dearth of measurements and the large temporal and spatial variability of deposition. Consequently, the ocean community must rely 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], but these models themselves are unconstrained as to the amounts of rainfall delivered to the ocean and the parameterization of aerosol removal processes. Alternatively, natural radionuclides such as ^7Be , which are delivered to the ocean from the atmosphere, provide useful tracers for the input of atmospherically derived chemical species into the ocean. For such species, the source terms are definable, and measurements are not readily contaminated within normal environmental conditions [Turekian *et al.*, 1983].

[21] The flux (F_i) of an element into the ocean is described by the sum of wet and dry deposition processes, respectively [e.g., Duce *et al.*, 1991]:

$$F_i = R \times S \times C_{p_i} \times \rho_r / \rho + C_{p_i} \times Vd, \quad (1)$$

where R is the precipitation rate, S is the scavenging ratio (the ratio of the concentration of a substance in rain, C_r , to that on particles, C_{p_i}), ρ_r is the density of water, ρ is the density of air (1200 g m $^{-3}$), and Vd is the dry deposition velocity (cm s $^{-1}$).

[22] The ocean community has largely resorted to the use of canonical values of Vd [Duce *et al.*, 1991] that are related to particle size distribution. The size distribution of ^7Be radioactivity is predominantly submicrometer with an activity mean diameter of about 0.5 μm [Winkler *et al.*, 1998]. For such aerosol particles, Vd is 0.1 cm s $^{-1}$ plus or minus a factor of 3. Calculating from concentrations in air using $F_i = C_{p_i} \times Vd$, with this canonical value (0.1 cm/s) and the measured ^7Be air concentration on Bermuda [Arimoto *et al.*, 1999], 4.4 mBq m $^{-3}$ (2.64×10^{-7} dpm cm $^{-3}$), the dry deposition rate would be 2.3×10^{-3} dpm cm $^{-2}$ d $^{-1}$, only 5% of the measured flux of 0.048 dpm cm $^{-2}$ d $^{-1}$ derived from the ocean inventory and rain collection. Thus wet deposition dominates. The ^7Be concentration used here falls within the range of concentrations of ^7Be measured in a broad range of (mostly continental) environments 3.98 to 5.9 mBq m $^{-3}$ [Akata *et al.*, 2008].

[23] Neglecting dry deposition, we can calculate the scavenging ratio using our ^7Be flux measurements. The required deposition rate to maintain the water column inventory of

⁷Be is 0.048 dpm cm⁻² d⁻¹. Using the annual rainfall rate for Bermuda of 146 cm (0.40 cm d⁻¹) based on long-term measurements (1949–2008) from the Bermuda Meteorological Service (<http://www.weather.bm/climate.asp>), and the air concentration (Cp_i) from *Arimoto et al.* [1999],

$$F_i = R \times S \times Cp_i \times \rho^{-1},$$

$$F_{7Be} = Cp_{7Be}(R \times S \times \rho^{-1}) = 0.048 \text{ dpm cm}^{-2} \text{ d}^{-1} \quad (2)$$

$$= 2.64 \times 10^{-7} \times 0.40 \times S \times 833.$$

This yields $S = 546$. A lower limit of this term is derived ($S \sim 328$) if the Tudor Hill flux is used.

[24] These scavenging ratios are comparable to those reported in the literature, which mostly report on continental measurements. Scavenging ratios of various chemical species in rain were measured over a 2.5 year period from 1989 to 1991 based on measurements at TH [Galloway et al., 1993]. The scavenging ratios measured for nss-SO₄, NO₃, and MSA were 210, 342, and 240, respectively. The scavenging ratio for Na was considerably higher, 640, because Na is concentrated primarily in the marine boundary layer. Akata et al. [2008] summarized a wide range of reports (mostly at continental sites) and obtained scavenging ratios of ⁷Be that ranged from 130 to 2100 with a mean value of 640. An extended study by Todd et al. [1989] along the coast of Virginia at ~37°N (that is, in a marine-influenced environment in roughly the same latitude of Bermuda) obtained annual values of 370 and 375 in 1983 and 1984.

[25] Finally, the ⁷Be deposition rates we observe on Bermuda and the nearby BATS site are consistent with average rates determined by runs of the ECHAM5 model for the period 1986–1990 (U. Heikkilä, personal communication, 2010). This model run has been validated against a large number of ⁷Be surface air and deposition observations from different continents and was found to reproduce the observations within a factor of 2 [Heikkilä et al., 2008, Figure 2].

6. Conclusions

[26] We have presented concurrent estimates of ⁷Be deposition from island-based bulk collectors and ocean profile measurements to constrain estimates of ocean deposition rates by wet and dry deposition. Excluding measurements at Tudor Hill, rainfall amounts and ⁷Be flux measurements on Bermuda are generally consistent with those to the surrounding ocean. Wet deposition accounts for over 95% of the total deposition to the adjacent ocean. Dry deposition to the ocean surface, calculated using the deposition velocity of 0.1 cm s⁻¹, commonly used for submicrometer aerosols, accounts for only a few percent at most of the total ⁷Be deposition. In oceanic regions where routine rain collection is not possible, our results suggest that ⁷Be flux estimates based on the ocean ⁷Be inventory allows an independent assessment of the rain (and scavenging) rates over the ocean, a very difficult parameter to constrain, and one that will be invaluable in assessing the atmospheric input of trace elements.

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