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### Key Points:

- Dust flux to the Sargasso Sea estimated from aluminum in the surface ocean is 4–9 times that estimated from aluminum in aerosols and rain
- The discrepancy likely reflects an underestimate in the assumed 5 years replacement time for dissolved aluminum in a 200 m surface mixed layer
- A replacement time of ~20–50 years brings the two estimates into accord, but may yield artifacts due to lateral transport of surface waters

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Dust Deposition to the Sargasso Sea: A Comparison of Estimates Using Aluminum in the Surface Ocean Versus Aerosols and Rainwater

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**Abstract** As a major component of mineral aerosol that partially dissolves in seawater, aluminum (Al) has been used to estimate time-averaged dust fluxes to the ocean, based on dissolved aluminum (DAI) concentrations and an assumed replacement time of DAI ( $\tau_{\text{DAI}}$ ) in the surface mixed layer, along with the fractional solubility ( $Al_s$ ) and mass abundance of Al in dust. We apply this method in the Bermuda region using seasonal water-column DAI concentrations, and compare our results with fluxes estimated from Al measured in Bermuda aerosols and rain. Inventories of DAI over the maximum mixed-layer depth of 200 m and empirically derived  $Al_s$  values yield mean dust fluxes of 5.3–11.2 g m<sup>-2</sup> yr<sup>-1</sup>, around 4- to 9-fold higher than fluxes estimated from aerosols and rain, which average 1.2 g m<sup>-2</sup> yr<sup>-1</sup> over a 318-day sampling period. This discrepancy likely reflects an underestimate in  $\tau_{\text{DAI}}$  and lateral transport of DAI in the surface ocean.

**Plain Language Summary** The rate at which soil dust from the continents is deposited to surface ocean waters is important, because dust is a major source of some chemical elements to the ocean, including the essential micronutrient iron, which controls primary production in some ocean regions. In this study we compare two different methods of estimating dust deposition rate to the North Atlantic Ocean near Bermuda, using dust, rain and seawater samples collected during calendar year 2019. Both methods use the chemical element aluminum (Al), which is present in soil dust in a relatively constant proportion. The first method estimates dust deposition using measurements of Al in dust and rain collected on Bermuda, and the second uses measurements of dissolved Al in the upper ocean near Bermuda. We find that the second method yields deposition rates 4- to 9-times higher than the first method, probably because the amount of time dissolved Al remains in the upper ocean in the study region is longer than typically assumed. Increasing that time can bring the two estimates into agreement, although that may introduce problems due to lateral movement of ocean waters, such that the estimated dust deposition is not representative of the ocean region around Bermuda.

## 1. Introduction

Deposition of mineral aerosol (dust) is a major source of bioactive trace elements to the surface ocean, yet this biogeochemically important flux remains difficult to constrain, complicated by large inter-annual and shorter-term variability (Anderson et al., 2016; Duce & Tindale, 1991; Kok et al., 2020; Mahowald et al., 2009). On average, dust contains aluminum (Al) in a proportion close to its mass abundance in the upper continental crust (UCC, Lawrence & Neff, 2009; Trapp et al., 2010; Zhang et al., 2015). Accordingly, dry and wet dust deposition can be estimated from Al concentrations in aerosols and rainwater, if aerosol deposition velocity and rainfall, respectively, are known. Alternately, the partial dissolution of aerosol Al in seawater (Buck et al., 2013; Maring & Duce, 1987; Measures et al., 2010) and an apparent several-year replacement time of dissolved aluminum (DAI) in ocean surface waters (Jickells et al., 1994; Oriens & Bruland, 1986) allow time-averaged dust deposition to be estimated from the surface-ocean inventory of DAI using the *measurement of Al for dust calculation in oceanic waters* (MADCOW) model (Measures & Brown, 1996; Measures & Vink, 2000).

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For the MADCOW method, there are substantial uncertainties associated with the replacement time of DAI in the surface ocean (Dammshäuser et al., 2011; Xu & Weber, 2021) and the fractional solubility of aerosol Al in seawater (Baker et al., 2006; Measures et al., 2010). To further examine these issues, we utilize data from seasonally resolved aerosol and rainwater samples and contemporaneous oceanic water-column samples collected from the Bermuda region to derive two independent estimates of dust deposition, based on (a) total Al in aerosols and rain and (b) the DAI inventory of the ocean surface mixed layer (SML). Bermuda and the adjacent Sargasso Sea are particularly well suited for this study, given the rich context of oceanographic observations collected as part of the ongoing Bermuda Atlantic Time-series Study (BATS, Lomas et al., 2013; Steinberg et al., 2001), and decades of atmospheric sampling at Tudor Hill, Bermuda, which has documented the regularly elevated transport of North African dust to this region during the summer months (Arimoto et al., 1995; Sorooshian et al., 2020).

## 2. Methods

Aerosol, rainwater and water-column samples together with hydrographic observations were collected from the Bermuda region of the North Atlantic Subtropical Gyre (Sargasso Sea), as part of the Bermuda Atlantic Iron Time-series (BAIT) project, GEOTRACES Process Study GApr13 (Sedwick et al., 2023). Sample collection, processing and analysis are summarized below, with complete details provided in Supporting Information S1.

### 2.1. Collection and Processing of Aerosol and Rain Samples From Bermuda

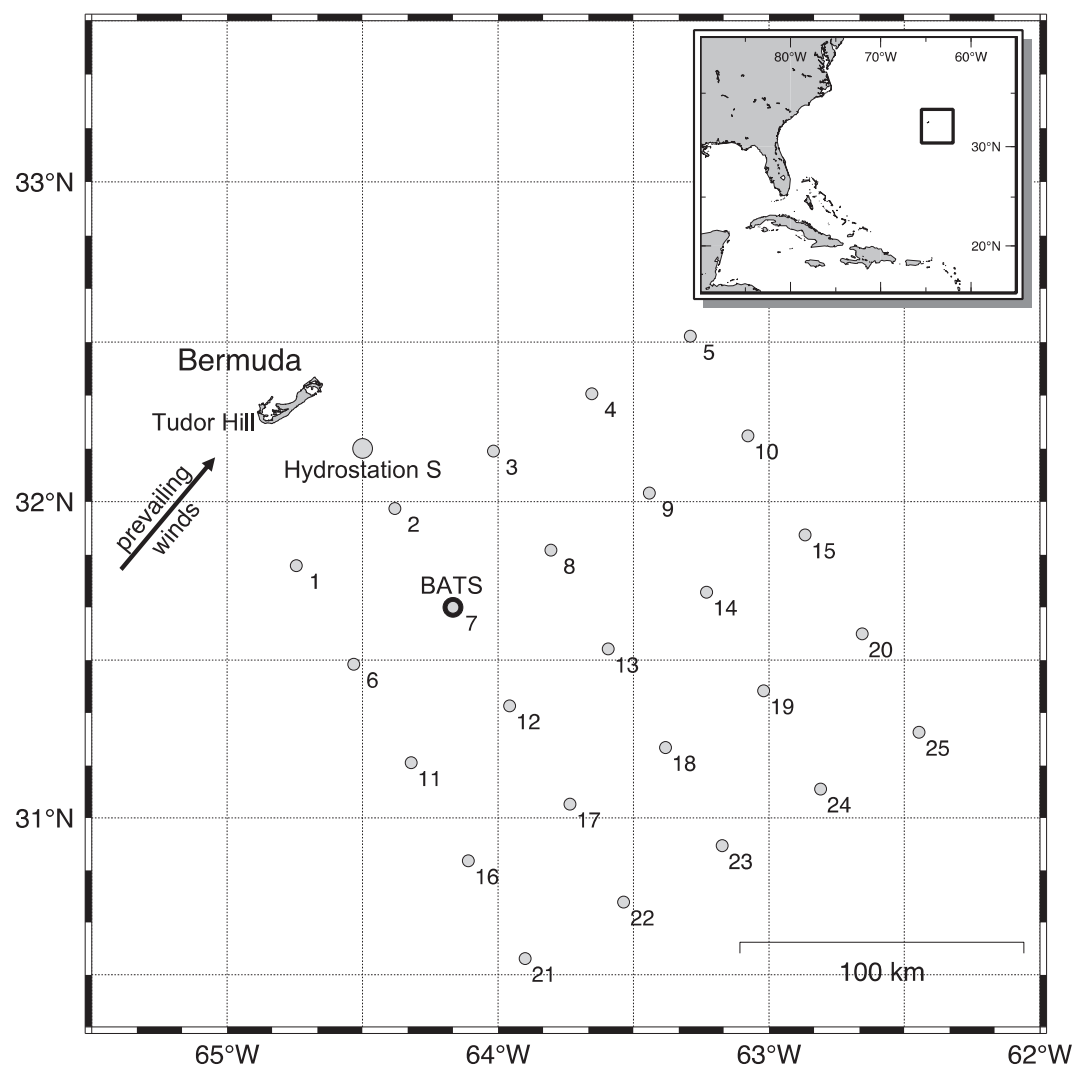
Composite samples of bulk aerosol and rainwater and associated field blanks were collected from the sampling tower at Tudor Hill, Bermuda (32°15.9'N 64°52.6'W, Figure 1) on an approximately weekly basis over the period from November 2018 to March 2020 as described by Sedwick et al. (2023). Aerosols were collected on acid-cleaned Whatman-41 cellulose filters using a high-volume aerosol sampler. For analysis of total aerosol aluminum, portions of the aerosol sample filters were digested with a mixture of ultrapure concentrated acids and hydrogen peroxide using microwave heating. Replicate portions of each aerosol sample filter were also subjected to a flow-through leaching procedure using 250 mL of high-purity deionized water (DIW), and the leachate acidified to 0.4% (v/v) with 6 M ultrapure hydrochloric acid for analysis of “DIW-soluble aerosol Al”; the same filter portions were then subjected to a batch leach with 25% acetic acid and 0.02 M hydroxylamine hydrochloride at 90°C (Kadko et al., 2020), with the supernatant leachate evaporated then diluted with 2% (v/v) concentrated ultrapure nitric for analysis of “HOAc-soluble aerosol Al.” Rainwater samples were collected in acid-cleaned 2-L wide-mouth fluorinated high density polyethylene bottles using an automatic rain sampler, and subsequently acidified to 0.4% (v/v) with 6 M ultrapure hydrochloric acid; after 1 week the acidified samples were transferred into acid-cleaned 125 mL low-density polyethylene bottles for analysis of “total-dissolvable aluminum” (TDAI) after storage at room temperature for more than 6 months. Field blanks for aerosol and rainwater were processed in the same manner as the samples.

### 2.2. Collection and Processing of Water-Column Samples From the BATS Region

Water-column samples and hydrographic data were collected from the BATS site (31°40'N, 64°10'W) and adjacent BATS spatial stations (Figure 1) on cruises during 10–15 March (spring), 11–17 May (early summer), 16–22 August (late summer), and 15–21 November (fall) of 2019 aboard RV *Endeavor* or RV *Atlantic Explorer*, as described by Sedwick et al. (2023). Samples were collected in 5-L external-closure Niskin-X samplers mounted on a custom-built trace-metal clean carousel and conductivity-temperature-depth sensor deployed on a non-metallic line. During the August cruise, near-surface (~0.3 m depth) samples were collected in a Niskin-X sampler deployed from a small inflatable boat. Upon recovery, the seawater samples were filtered through pre-cleaned 0.2-μm pore AcroPak Supor filter capsules using filtered nitrogen gas. For DAI analysis, the seawater filtrate was collected in acid-cleaned 100 mL low-density polyethylene bottles (Bel-Art), and acidified to pH 1.7 post-cruise by addition of 6 M ultrapure hydrochloric acid.

### 2.3. Analysis of Aluminum in Aerosol Digests, Aerosol Leachates, and Rainwater

Aluminum was determined in the aerosol digest and leachate solutions, acidified rainwater, and associated field blanks by inductively coupled plasma mass spectrometry. Analytical precision is estimated at better than ±10% (one relative standard deviation of the mean) for total aerosol Al, DIW-soluble aerosol Al, HOAc-soluble aerosol

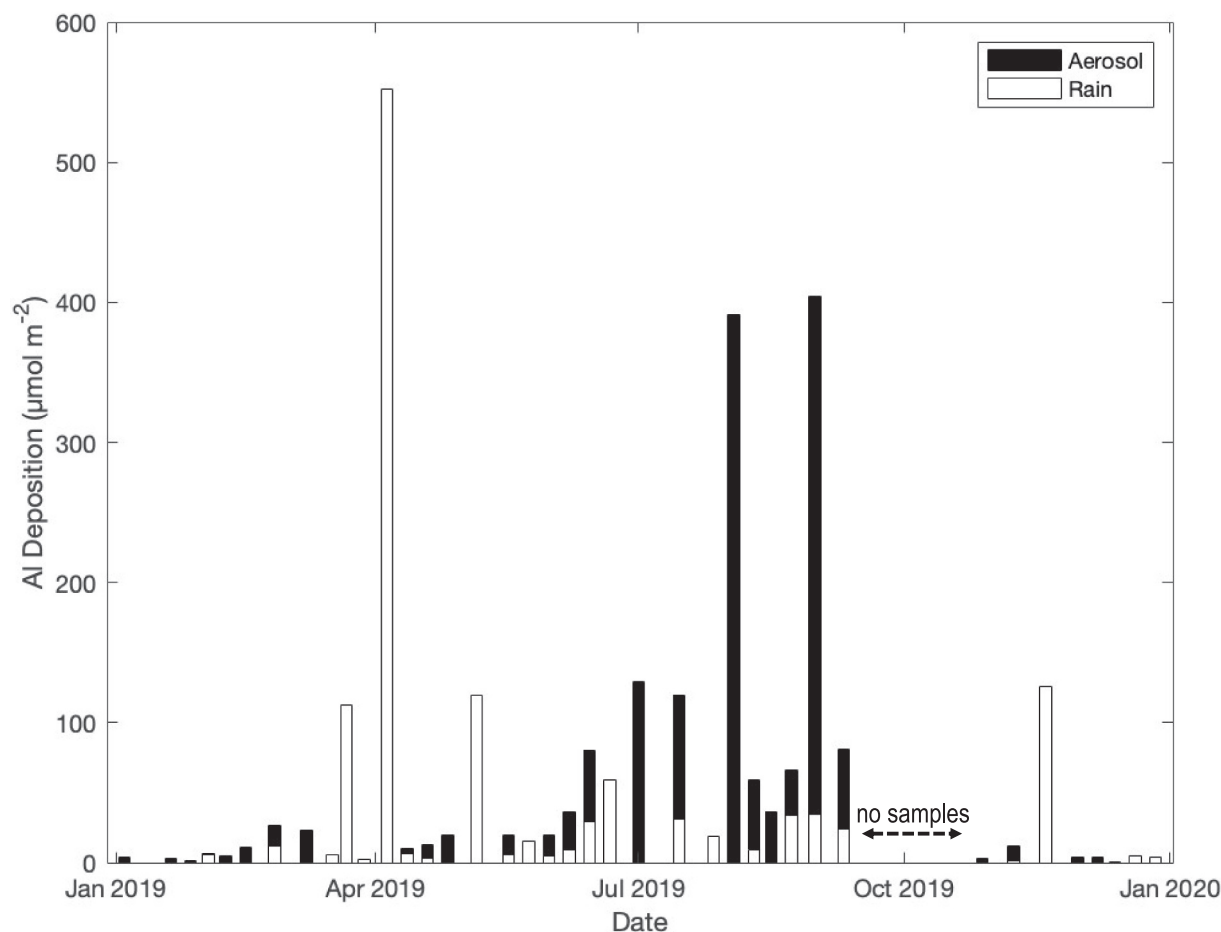


**Figure 1.** Study region showing locations of Tudor Hill on Bermuda, where aerosols and rainwater were collected, and the Bermuda Atlantic Time-series Study (BATS) site (station 7) and adjacent BATS spatial stations, where oceanic water-column samples were collected (map created by Paul Lethaby).

Al, and TDAI in rainwater. Our estimated detection limits correspond to atmospheric loadings of 0.021, 0.126, and 0.003  $\text{nmol m}^{-3}$  for total aerosol Al, DIW-soluble aerosol Al and HOAc-soluble aerosol Al, respectively, and a rainwater TDAI concentration of 150  $\text{nmol L}^{-1}$ , based on average sample volumes of around 3,000  $\text{m}^3$  of air and 100 mL of rainwater. Atmospheric loadings of total aerosol aluminum, DIW-soluble aerosol aluminum, and HOAc-soluble aerosol aluminum were calculated from the Al concentrations of the aerosol digest and leach solutions, and corresponding volumes of air sampled.

#### 2.4. Analysis of DAI in Water-Column Samples

Post-cruise determinations of DAI in the filtered, acidified water-column samples used flow injection analysis with fluorometric detection, without in-line preconcentration, following the method of Resing and Measures (1994). Repeated separate determinations of a low-concentration seawater sample yielded a DAI concentration of  $3.19 \pm 0.68 \text{ nM}$  ( $n = 12$ ), indicating an estimated analytical uncertainty of  $\pm 21\%$  (one relative standard deviation of the mean) and detection limit of 2.04 nM (three standard deviations of the mean). Repeated separate analyses of the GEOTRACES GSP reference seawater yielded a mean concentration of  $1.52 \pm 0.28 \text{ nM}$  ( $n = 6$ ), although there is as yet no consensus value available for this material.



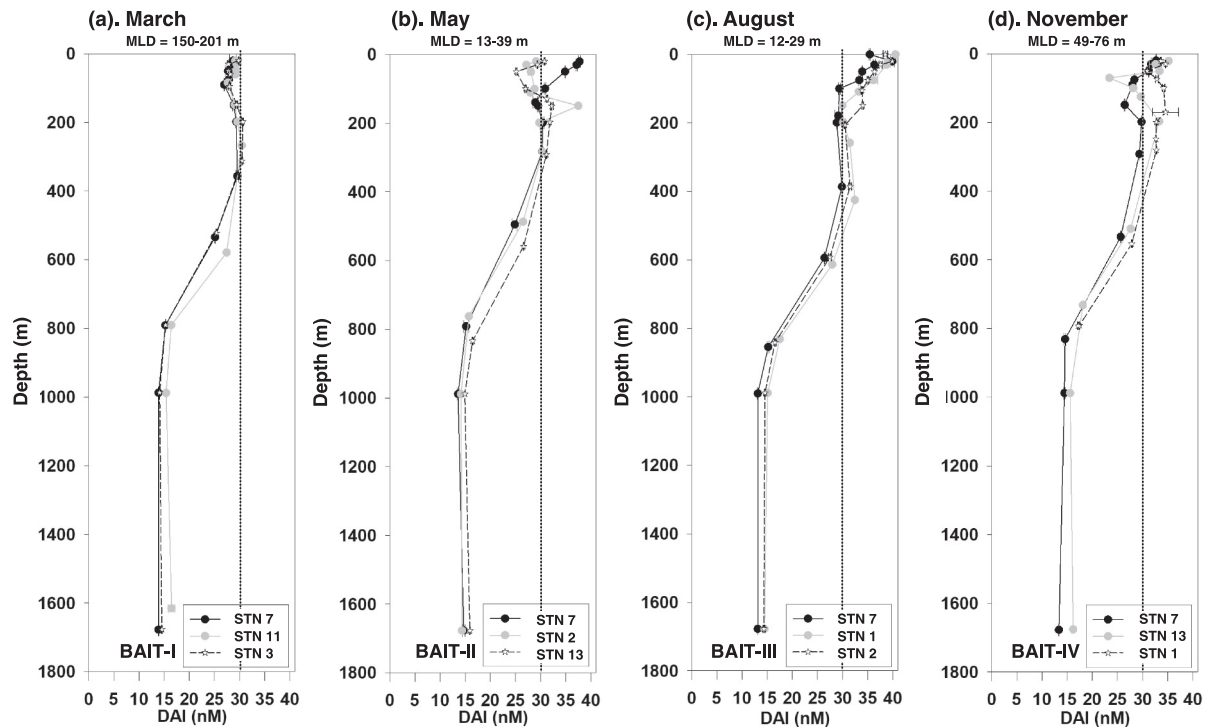
**Figure 2.** Dry deposition (black bars) and wet deposition (white bars) of total Al and TDAI, respectively, for each approximately 1-week sampling period, estimated from analysis of aerosols and rain collected at the Tudor Hill tower on Bermuda. Deposition is calculated from the product of the Al deposition flux ( $\mu\text{mol m}^{-2} \text{d}^{-1}$ ) and each total  $\sim$ weekly sampling period (d). The annualized mean deposition of Al is estimated from the sum of all Al deposited (dry and wet) during these sampling periods divided by the total aerosol sampling days during 2019 (318 days). Dust mass flux is derived by assuming a upper continental crust mass abundance of 8% Al (see Section 3.1).

### 3. Results

Our aerosol, rainwater and water-column aluminum data allow two independent estimates of dust flux to the Bermuda region, averaged over an annual timescale. The first estimate uses our measurements of total aluminum in aerosols and TDAI in rainwater to calculate an annualized average dust flux to the study region. The second estimate utilizes the MADCOW model (Measures & Brown, 1996), which calculates dust flux from the water-column inventory of DAI in the SML of the BATS region, assuming a fixed replacement time (or mean residence time) for DAI therein. These two estimates are discussed below.

#### 3.1. Dust Flux Estimated From Al in Aerosols and Rainwater on Bermuda

Dry deposition of Al was calculated for each aerosol sampling period (Figure 2) from the atmospheric loading of total aerosol Al multiplied by the aerosol deposition velocity and the duration of each  $\sim$ weekly sampling period. For the aerosol dry deposition velocity, we assume a constant value of  $1 \text{ cm s}^{-1}$ , consistent with previous estimates for this region (Arimoto et al., 2003; Kandler et al., 2018; Tian et al., 2008) and a value derived from a global relationship between bulk deposition velocities estimated with beryllium-7 and rainfall rate (Kadko et al., 2020). From this we calculate an annualized mean dry deposition of Al ( $5.53 \mu\text{mol m}^{-2} \text{d}^{-1}$ ) from the sum of the total aerosol Al deposited during all sampling periods divided by the total duration of the aerosol sampling days during calendar year 2019 (318 days); we note that the passage of Hurricane Humberto over Bermuda and a resulting power outage interrupted our Tudor Hill sampling from 16 September to 21 October (that 5-week



**Figure 3.** Vertical concentration profiles of dissolved aluminum (DAI) from samples collected during the Bermuda Atlantic Iron Time-series project cruises in (a) March, (b) May, (c) August, and (d) November of 2019. The range of mixed layer depths observed on each cruise is shown at top of each panel. For reference, the vertical dotted line in each panel indicates a DAI concentration of 30 nM.

unsampled period is excluded from our calculation of mean aeolian flux). When converted to a mass basis and divided by the mass abundance of Al in aerosol, which we assume equal to the UCC abundance of 8% (Li, 2000; Taylor & McLennan, 1985), this yields an annualized mean dry dust deposition of  $0.68 \text{ g m}^{-2} \text{ yr}^{-1}$ . Using a similar calculation scheme, our measurements of total-dissolvable Al in rainwater can be combined with measured rainfall amounts for each sampling period to estimate the total wet deposition of Al during each sampling period (Figure 2). Again assuming a UCC mass abundance of 8% for Al in dust yields an annualized mean wet dust deposition of  $0.52 \text{ g m}^{-2} \text{ yr}^{-1}$ . Summing our estimates for dry and wet dust deposition then yield a mean total dust deposition of  $1.20 \text{ g m}^{-2} \text{ yr}^{-1}$  for the Tudor Hill sampling site during our 318-day sampling period.

### 3.2. Dust Flux Estimated From Water-Column DAI Inventory in the BATS Region

The water-column based dust flux estimate using the MADCOW model (Measures & Brown, 1996; Measures & Vink, 2000) assumes that the DAI inventory in the SML ( $\text{DAI}_{\text{SML}}$ ) is at steady state, and that the replacement time of DAI ( $\tau_{\text{DAI}}$ ) in the SML, with respect to atmospheric input, is 5 years. The atmospheric deposition flux of seawater-soluble aluminum is then calculated from  $\text{DAI}_{\text{SML}}$  divided by  $\tau_{\text{DAI}}$ . Finally, the aeolian flux of seawater-soluble aluminum is divided by the fractional solubility of aerosol aluminum in seawater ( $\text{Al}_s$ ) and the mass abundance of aluminum in dust ( $\text{Al}_{\text{dust}}$ ) to yield the total dust deposition flux in  $\text{g m}^{-2} \text{ yr}^{-1}$ , following Equation 1:

$$\text{dust flux} = \text{DAI}_{\text{SML}} / (\tau_{\text{DAI}} \times \text{Al}_s \times \text{Al}_{\text{dust}}) \quad (1)$$

Water-column DAI concentration profiles from the four seasonal BAIT cruises are shown in Figure 3, along with the corresponding depth ranges of the SML as estimated from the criteria of de Boyer Montegut et al. (2004), using a temperature threshold of  $0.2^\circ\text{C}$  relative to reference pressures of either 10 or 30 dbar. As is typical for the BATS region (Lomas et al., 2013), we observed a substantial seasonal variation in SML depth, which ranged from  $\sim 150$  to  $200 \text{ m}$  in early spring (BAIT-I cruise), to  $\sim 10$ – $30 \text{ m}$  in late summer (BAIT-III cruise). Our DAI measurements reveal both seasonal and lateral variations in the distribution of DAI in the upper water column. A

**Table 1**

*Mean Water-Column Dissolved Aluminum (DAI) Inventory in the Upper 200 m and Associated MADCOW-Derived Dust Flux Estimates, Along With Overall Mean Dust Flux Estimated From Al in Bermuda Aerosols and Rain, and Dust Flux Estimated From DAI Accumulation in 30 m Surface Mixed Layer Over Summer*

Cruise	BAIT-I	BAIT-II	BAIT-III	BAIT-IV	Overall mean
DAI inventory ( $\mu\text{mol m}^{-2}$ )	$5,740 \pm 70$	$6,150 \pm 300$	$6,760 \pm 350$	$6,620 \pm 840$	$6,320 \pm 590$
Dust deposition <sup>a</sup> using $\text{Al}_{\text{S-DIW}}$ ( $\text{g m}^{-2} \text{yr}^{-1}$ )	$10.2 \pm 0.1$	$10.9 \pm 0.5$	$12.0 \pm 0.6$	$11.8 \pm 1.5$	$11.2 \pm 1.0$
Dust deposition <sup>b</sup> using $\text{Al}_{\text{S-DIW+HOAc}}$ ( $\text{g m}^{-2} \text{yr}^{-1}$ )	$4.8 \pm 0.1$	$5.1 \pm 0.3$	$5.6 \pm 0.3$	$5.5 \pm 0.7$	$5.3 \pm 0.5$
Dust deposition <sup>c</sup> from Al in Bermuda aerosols and rain ( $\text{g m}^{-2} \text{yr}^{-1}$ )	—	—	—	—	1.20
Dust deposition <sup>d</sup> from summer DAI accumulation ( $\text{g m}^{-2} \text{yr}^{-1}$ )	—	—	—	—	1.5–6.3

<sup>a</sup>MADCOW-derived estimate using fractional solubility based on DIW-leach of aerosols. <sup>b</sup>MADCOW-derived estimate using fractional solubility based on DIW + HOAc leach of aerosols. <sup>c</sup>Estimated from measurements of Al in aerosols and rain collected on Bermuda. <sup>d</sup>Estimated from DAI accumulation in 30 m surface mixed layer over April–August period (see Section 4).

seasonal accumulation of  $\sim 5$ – $10$  nM DAI is evident in near-surface samples collected on the August (BAIT-III) and November (BAIT-IV) cruises, suggesting that atmospheric input exceeded scavenging removal from the shallow SML during the summer months. There are also lateral concentration differences of  $\sim 5$  nM in the upper water column between adjacent BATS spatial stations during the May (BAIT II) and November (BAIT IV) cruises (Figure 3); such lateral variability has also been observed for dissolved iron and is likely related to the mesoscale eddies that move through the region (Sedwick et al., 2020, 2023).

We choose a SML depth of 200 m, corresponding to the maximum mixed layer depth observed during our four cruises (Figure 3), which should thus account for the impact of seasonal turnover over an annual cycle. For each station sampled, the inventory of DAI in the SML was estimated by summing the product of sample concentrations and the thickness of depth layers, where each layer was defined by the midpoints between successive sampling depths. The mean DAI SML inventory for each cruise was then used to calculate a dust deposition flux from Equation 1, assuming a  $\tau_{\text{DAI}}$  value of 5 years and mass abundance of 8% for Al in dust (Measures & Vink, 2000). For the fractional solubility of aerosol aluminum in seawater, we use the mean of values determined from the DIW and DIW + HOAc leaches performed on our aerosol samples ( $\text{Al}_{\text{S-DIW}} = 3.8\%$  and  $\text{Al}_{\text{S-DIW+HOAc}} = 8.1\%$ , respectively), which Shelley et al. (2018) argue should define the lower and upper limits of aerosol aluminum dissolution in seawater. These dust deposition estimates are shown in Table 1, and yield overall annual mean fluxes of  $11.2 \text{ g m}^{-2} \text{yr}^{-1}$  (based on  $\text{Al}_{\text{S-DIW}}$ ) and  $5.3 \text{ g m}^{-2} \text{yr}^{-1}$  (based on  $\text{Al}_{\text{S-DIW+HOAc}}$ ).

#### 4. Discussion

The dust flux estimates derived from our aerosol and rain samples agree reasonably well with previous, similarly derived estimates for the Bermuda region ( $1.5$ – $1.9 \text{ g m}^{-2} \text{yr}^{-1}$ , Jickells et al., 1994), as well as estimates derived from sediment-trap samples ( $1.7 \text{ g m}^{-2} \text{yr}^{-1}$ , Jickells et al., 1998) and the fluxes obtained from global-scale dust models ( $2.2 \text{ g m}^{-2} \text{yr}^{-1}$ , Prospero, 1996;  $2$ – $4 \text{ g m}^{-2} \text{yr}^{-1}$ , Albani et al., 2014;  $1.0$ – $3.2 \text{ g m}^{-2} \text{yr}^{-1}$ , Kok et al., 2020; and  $\sim 1 \text{ g m}^{-2} \text{yr}^{-1}$ , Xu & Weber, 2021). Nonetheless, our calculated dust fluxes are likely underestimates, because they exclude dust deposited when winds were  $< 1 \text{ m s}^{-1}$  or outside the  $210^\circ$ – $315^\circ$  directional sector required for aerosol sample collection at Tudor Hill (this sector control is intended to avoid sampling aerosols from local sources), which sometimes accounted for more than half of each weekly collection period. In addition, the measured rainwater TDAI concentrations are likely to considerably underestimate the total Al concentrations of those samples; indeed, Tian et al. (2008) report that TDAI in Bermuda rainwater accounted for 39%, on average, of the total Al concentrations measured following strong acid digestion. On that basis, our wet deposition estimate might be adjusted upwards to  $1.33 \text{ g m}^{-2} \text{yr}^{-1}$  ( $= 0.52/0.39 \text{ g m}^{-2} \text{yr}^{-1}$ ), yielding a total dust deposition of around  $2 \text{ g m}^{-2} \text{yr}^{-1}$ .

A striking result from this study is the poor agreement between the two different methods used to estimate dust deposition (Table 1), with the mean flux calculated from DAI inventories in the SML in the BATS region ( $5.3$ – $11.2 \text{ g m}^{-2} \text{yr}^{-1}$ ) being more than 4- to 9-fold greater than the flux calculated from total Al in aerosols and rainwater on Bermuda ( $1.20 \text{ g m}^{-2} \text{yr}^{-1}$ ). Even if our aerosol- and rain-based dust flux is underestimated by a factor of two, as discussed above, then the MADCOW-based flux estimate remains more than 2- to 4-fold higher.



There are two factors that may substantially contribute to this discrepancy, both of which are related to the assumed replacement time (residence time) of DAI in the SML,  $\tau_{\text{DAI}}$ . First is the assumed  $\tau_{\text{DAI}}$  value of 5 years (Measures & Vink, 2000), which is based on previous field studies (Jickells et al., 1994; Oriens & Bruland, 1986). Importantly, those studies estimate  $\tau_{\text{DAI}}$  for SML depths in the range of 25–100 m. Measures and Vink (2000) used MADCOW to calculate a mean dust deposition of  $2.6 \text{ g m}^{-2} \text{ yr}^{-1}$  for the Sargasso Sea—a value that is closer to our aerosol- and rain-based flux estimate—although they assume a SML depth of only 30 m, which is not representative of the full depth of mixing over an annual cycle.

For the SML depth of 200 m that we have chosen to account for vertical mixing in the BATS region over an annual timescale, a larger value of  $\tau_{\text{DAI}}$  may be expected, given that there will be a larger inventory of DAI in the deeper SML. Noting that our dust flux calculated using the MADCOW model is 4.4- to 9.3-fold greater than that calculated using aluminum in aerosols and rainwater, these two flux estimates can be brought into accord if the value of  $\tau_{\text{DAI}}$  is increased to 22 or 46.5 years, for estimates based on aerosol-Al fractional solubilities of 8.1% and 3.1%, respectively (see Equation 1); more modest increases in  $\tau_{\text{DAI}}$  are required if we accept that our aerosol- and rain-based flux estimates are likely on the low side. Such longer replacement times are consistent with a recent modeling study of the ocean aluminum cycle constrained by assimilation of surface DAI concentrations (Xu & Weber, 2021), which yields a  $\tau_{\text{DAI}}$  of 10.8 years for a SML depth of 55 m (H. Xu, pers. comm.). However, decadal-scale replacement times remain at odds with  $\tau_{\text{DAI}}$  estimates of less than 5 years based on thorium supply (Hayes et al., 2018).

A second factor that may contribute to the disagreement between our two dust flux estimates concerns the potential impact of lateral transport on the inventory of DAI in the upper water column over time periods on the order of  $\tau_{\text{DAI}}$ . In the Sargasso Sea, the mean regional-scale surface circulation extends over  $\sim 1,000 \text{ km}$  of latitude, with surface waters in the BATS region carried southwest until they are steered northeast by Gulf Stream (Jenkins & Doney, 2003; Steinberg et al., 2001). For a mean advective speed in the range of  $3\text{--}6 \times 10^{-2} \text{ m s}^{-1}$  (Artigue et al., 2021; Jenkins & Doney, 2003), surface waters in the BATS region could circulate several times around a gyre with a circumference of  $\sim 3,000 \text{ km}$  over a decade—a time period of the same order as our inferred replacement time of DAI in the SML. An important implication of this rough calculation is that the inventory of DAI in the SML of the BATS region could reflect advective inputs from waters located well to the southwest of Bermuda, where higher dust deposition fluxes and perhaps inputs from the Gulf Stream result in higher DAI concentrations in surface waters (Artigue et al., 2021; Middag et al., 2015; Shelley et al., 2012). If so, then the DAI input flux calculated using the MADCOW method will represent contributions from those sources *in addition to* dust deposition in the Bermuda region, thereby overestimating local dust deposition.

Here an alternate approach is to apply a MADCOW-type estimate to the period between the March and August cruises, when dust deposition is typically maximum and the DAI inventory of the shallow, summer SML is clearly *not in steady state*. During that  $\sim 5$ -month period, the SML had shoaled to a depth closer to the 30 m value used by Measures and Vink (2000), and DAI concentrations in that shallow layer increased from  $\sim 30$  to  $\sim 35\text{--}40 \text{ nM}$ . Assuming that this concentration increase reflects accumulation of dust-derived DAI in the shallow SML, and neglecting scavenging losses of DAI during those summer months, when primary production is low (Tin et al., 2016), we can estimate a *seasonal* dust flux using Equation 1. For this calculation, we replace  $\tau_{\text{DAI}}$  by a period of 5 months—essentially a *response time* during which the DAI inventory increased by  $\sim 150\text{--}300 \mu\text{mol m}^{-2}$  over the upper  $\sim 30 \text{ m}$ . This yields a summer dust flux in the range of  $1.5\text{--}6.3 \text{ g m}^{-2} \text{ yr}^{-1}$  (Table 1) using mean  $\text{Al}_s$  values derived from our Tudor Hill aerosol samples. The lower end of this range is in accord with the dust flux estimated from Al in our aerosol and rain samples ( $1.2 \text{ g m}^{-2} \text{ yr}^{-1}$ ), suggesting that the much shorter timescale of this calculation, relative to a decadal-order replacement time of DAI over the full annual range of vertical mixing in the BATS region, renders it less susceptible to artifacts due to lateral advection.

## 5. Concluding Remarks

Our unique, seasonally resolved data sets from the upper water column in the BATS region and the atmospheric marine boundary layer over Bermuda provide insight into limitations of the MADCOW model for estimating time-averaged dust flux to ocean regions where seasonal mixing is relatively deep. Specifically, the required assumption of steady state for the DAI inventory in the SML over annual timescales requires that we account for the full range of vertical mixing during that period, which typically extends to at least 200 m in waters of the Bermuda region. Adopting that deeper SML in turn requires that the DAI replacement time must also be increased

beyond the previously assumed value of 5 years (Measures & Vink, 2000; Measures et al., 2005). Doing so brings the MADCOW-based estimate into better accord with the flux estimates derived from Al in aerosols and rain collected on Bermuda.

However, adopting a longer replacement time for DAI then introduces the possibility of substantial lateral transport of surface waters over the timescale of the calculation, meaning that the MADCOW-derived fluxes will likely reflect DAI inputs from dust deposition and other sources well beyond our immediate study region. Importantly, similar considerations will apply to the potential use of other elements as tracers of dust deposition, such as titanium and gallium, which share some geochemical similarities to Al, but appear to have even longer replacement times in surface ocean waters (Dammshäuser et al., 2011; Ho et al., 2019).

## Data Availability Statement

Data presented and discussed here, and associated metadata, are available from the U.S. National Science Foundation's Biological and Chemical Oceanography Data Management Office at: <https://www.bco-dmo.org/project/822807> (Sedwick & Sohst, 2025; Sedwick et al., 2024, 2025).

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