Tropical rains controlling deposition of Saharan dust across the North Atlantic Ocean

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Key Points

1. Dust deposition fluxes collected while settling in the tropical North Atlantic show highest dust deposition in summer and autumn
2. Wet deposition of Saharan dust dominates over the tropical North Atlantic from July to October
3. Model simulations corroborate sediment trap and satellite observations of wet and dry dust deposition

Keywords

Mineral dust
Atlantic Ocean
Dust deposition
Wet deposition
Ocean fertilization

Abstract

Mineral dust plays an important role in the atmospheric radiation budget as well as in the ocean carbon cycle through fertilization and by ballasting of settling organic matter. However, observational records of open-ocean dust deposition are sparse. Here, we present the spatial and temporal evolution of Saharan dust deposition over 2 years from marine sediment traps across the North Atlantic, directly below the core of the Saharan dust plume, with highest dust fluxes observed in summer. We combined the observed deposition fluxes with model simulations and satellite observations and argue that dust deposition in the Atlantic is predominantly controlled by summer rains. The dominant depositional pathway changes from wet deposition in summer to dry deposition in winter. Wet deposition has previously been suggested to increase the release of dust-derived nutrients and their bioavailability, which may be a key contributor to surface-ocean productivity in remote and oligotrophic parts of the oceans.

Plain Language Summary

Large quantities of dust are emitted from the Sahara Desert and blown over the (sub)tropical North Atlantic Ocean. Atmospheric dust plays an important role in the Earth's climate, but observations of open-ocean deposition are sparse. Here, we present a continuous, 2-year record of dust deposition across the North Atlantic Ocean, between Africa and the Caribbean. We compare and evaluate measurements of dust deposition with model simulations and satellite observations. The combined results reveal that tropical rains dominate dust deposition across the Atlantic. In particular, the rain is responsible for the high dust deposition during summer and may also affect the nutrients that are carried with the dust. Since previous studies suggest that the nutrients that reach the surface ocean with the rain are more readily available for phytoplankton productivity, the wet deposition of dust potentially has a major impact on the ocean carbon cycle.

1. Introduction

Large quantities of mineral dust are blown from the Sahara Desert across the Atlantic (Yu et al., 2015). This dust impacts the atmosphere by altering the atmospheric radiation budget (Ryder et al., 2013) and by serving as cloud condensation nuclei (Twohy et al., 2009) and ice nuclei (Atkinson et al., 2013). Dust deposition also influences the ocean carbon cycle through the delivery of nutrients, which stimulate phytoplankton growth (Mills et al., 2004; Pabortsava et al., 2017) and by ballasting organic particles settling to the ocean floor (Bressac et al., 2014; Pabortsava et al., 2017; Van der Jagt et al., 2018). These effects on the atmosphere and ocean depend on atmospheric dust concentrations and deposition fluxes, and the size, shape and mineralogy of the dust particles (Shao et al., 2011). Dust particle size varies seasonally along the transect (Van der Does et al., 2016), which could in turn be related to compositional variations along the downwind trajectory as a result of preferential settling of coarser and heavier dust particles, related to their mineralogy (Korte et al., 2017).

The seasonal variability of dust transport and deposition over the Atlantic is subject to many factors. According to remote-sensing observations, maximum dust emission occurs in summer, while dust emissions are at a minimum in winter and autumn (Adams et al., 2012). Several dust-transporting wind systems are active, transporting the dust at different altitudes in the atmosphere during the different seasons (Stuut et al., 2005; Muhs, 2013; Yu et al., 2019). First, dust transport occurs year-round in the shallow north-easterly trade-wind layer. Furthermore, dust transport is influenced by the intertropical convergence zone (ITCZ; Nicholson, 2000), with the center of the main dust cloud migrating from 10–20°N in summer to 0–10°N in winter (Adams et al., 2012). With the southward migration of the ITCZ in winter the Harmattan winds intensify, transporting dust at altitudes between 0 and 3 km over the Atlantic (Muhs, 2013; Tsamalis et al., 2013). During summer when the ITCZ

migrates north, dust transport across the Atlantic occurs within the Saharan Air Layer at maximum altitudes of 5 to 7 km (Muhs, 2013; Weinzierl et al., 2017). The particle size of the deposited dust shows coarser grains at proximal locations, and finer-grained material at more distant sites (Mahowald et al., 2014; Van der Does et al., 2016), although giant mineral dust particles (>75 μm) were also identified thousands of kilometres from their sources (Van der Does et al., 2018).

The magnitude of dust deposition over the ocean is a function of the travelled distance, with more deposition close to the source (Sarnthein et al., 1981; Goudie & Middleton, 2001). Atmospheric dust particles can be removed from the atmosphere by dry and wet depositional processes (Zender et al., 2003; Bergametti & Forêt, 2014). Atmospheric processing of dust is of great interest since dust processing in aerosols under low pH conditions favors the leaching of important nutrients (Meskhidze et al., 2005; Korte et al., 2018), while the iron in dust is made more soluble in clouds due to reactions with oxalate (Johnson & Meskhidze, 2013), which are subsequently released into the surface ocean (Spokes et al., 1994) and lead to increased primary production and chlorophyll-a (Ridame et al., 2014). This may lead to higher solubility of iron and other nutrients in wet deposition (Spokes et al., 1994). In contrast, a study by Korte et al. (2018) suggests that dry-deposited dust has a much lower nutrient bio-availability. Also, since satellite instruments cannot detect wet dust deposition below the clouds, a multidisciplinary approach combining *in situ* observations and satellite remote sensing with model simulations should provide a better understanding of processes involved in dust transport and deposition type, and help to constrain the biogeochemical impact of dust deposition.

So far, transport and deposition of Saharan dust across the Atlantic Ocean was studied at sea during research expeditions (Stuut et al., 2005; Baker et al., 2010; Weinzierl et al., 2017), or monitored by sediment-trap collection at stations only at the eastern (Bory & Newton, 2000; Neuer et al., 2004; Brust et al., 2011) and western sides of the Atlantic (Carlson & Prospero, 1972; Jickells et al., 1998), and estimated by remote sensing (Yu et al., 2015). Here, we present Saharan dust deposition fluxes collected while settling in the tropical North Atlantic Ocean, along a transect below the core of the Saharan dust plume (at 12°N).

2. Methods

We monitored 2 years of Saharan dust deposition fluxes at four sites across the Atlantic Ocean using time-series from submarine sediment traps (M1–M4). The traps were moored at ~1,200 m water depth, three positioned along a transect at 12°N, and a fourth at 13°N (map Fig. 3), which sampled synchronously from October 2012 to October 2014 (Table S1). The transect is complemented by 2 years (2013–2014) of Saharan dust concentrations measured on Barbados (13°N, 59°W). We combined these observational data with dry- and wet-dust deposition fluxes simulated using the Community Earth System Model (CESM 1.2.2; Hurrell et al., 2013), and dust optical depth (DOD) and precipitation remote-sensing estimates along the transect were obtained using Moderate Resolution Imaging Spectroradiometer (MODIS) and Tropical Rainfall Measuring Mission (TRMM) datasets, respectively. These methods are described in detail in the Supporting Information.

3. Results and Discussion

3.1 Mineral Dust Fluxes

Residual-mass fluxes are often taken to represent the lithogenic or mineral dust fraction, and result from subtracting the weights of biogenic constituents from the total mass (e.g. Jickells et al., 1998; Fischer et al., 2016; Korte et al., 2017), defined as

Residual mass = total mass – $CaCO_3$ – $BSiO_2$ – OM,

where $CaCO_3$ represents calcium carbonates, $BSiO_2$ is biogenic silica, and OM is the organic matter. For the exact calculations, see Korte et al. (2017). All three fractions are determined by using specific calculation factors that are, due to, for example, an uncertain water content for $BSiO_2$ and OM, globally not well constrained and the range is often not taken into account. Since this representation of the lithogenic or mineral dust fraction is obtained indirectly, this fraction is associated with several uncertainties related to the conversion factors and measurements of the individual biogenic fractions. Here, we present dust-mass fluxes which were obtained by directly weighing the dust fraction, and are therefore more accurate and representative for the dust flux. In our method, the biogenic components were chemically removed and this resulted in the dust fraction only. These dust mass fluxes are significantly smaller than the residual-mass fluxes (Fig. 1) determined by Korte et al. (2017) on the same samples as were used here. Dust-mass fluxes at M1, M2 and M4 presented here are all highly correlated with the residual-mass flux (Korte et al., 2017), with R^2 values ranging between 0.69 and 0.78 and slopes decreasing downwind from 0.57 to 0.25 and 0.15, respectively. On average, the residual-mass fluxes are 5.8 times higher than the dust mass fluxes (standard deviation = 3.3), corroborating the conclusion by Korte et al. (2017) that the residual fraction most likely overestimates dust fluxes due to various assumptions and uncertainties in calculating the masses of biogenic matter. Additional components of the residual fraction potentially include biogenic phosphates and sulfates, particles of volcanogenic, cosmogenic and anthropogenic origin, as well as crystal water associated with the opaline silica and the OH content of organic matter and clay minerals (Korte et al., 2017). Although the residual fraction does reflect the temporal changes in dust deposition at each site, the absolute dust fluxes are severely overestimated and differ between sites. Alternative methods of estimating the dust deposition fluxes use particular lithogenic elements as proxies, mainly Al and Ti (e.g. Bory et al., 2002; Pabortsava et al., 2017), or automated mineral classification by EDX (Brust et al., 2011), but these methods also rely on several assumptions involving the composition and size of the dust particles, and therefore direct measurements of dust deposition fluxes are considered to be more accurate.

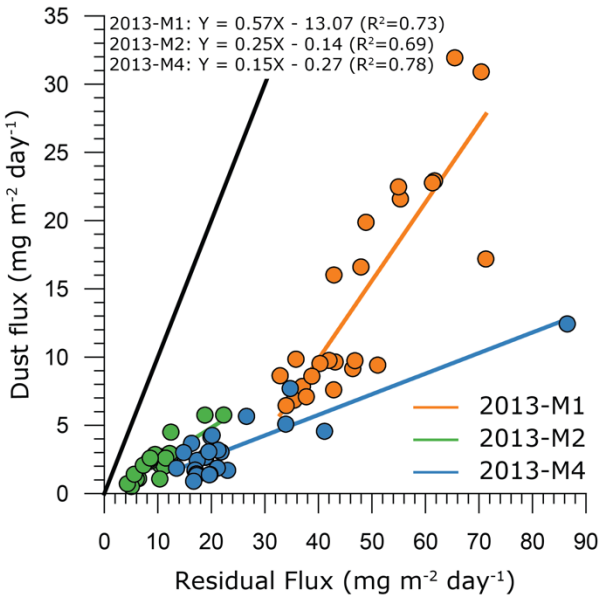

Figure 1. Dust-mass flux (established here) versus residual-mass flux (data from Korte et al., 2017) for 2013-M1–M4. Black line represents 1:1 relation.

3.2 Dust deposition along the transect

Dust-mass fluxes are highest at M1, located about 700 km from the west African coast, reaching maximum values of >30 mg m^{-2} day^{-1} and an average value of 14.3 mg m^{-2} day^{-1} in 2013 (Fig. 2). This is about 1.5 times higher than the lithogenic fluxes observed close to the Canary Islands by Neuer et al. (2004) and Brust et al. (2011), who observe average deposition fluxes of 10 and 9.5 mg m^{-2} day^{-1}, and a maximum deposition flux in winter of up to 54 and 43 mg m^2 day^{-1}, respectively. The episodic high fluxes in winter are likely due to dust transport at lower altitudes during this time and therefore deposited closer to the source (Van der Does et al., 2016), while the overall lower fluxes probably relate to the more northern position of the Canary Island traps, away from the center of the main dust cloud. In our sediment traps, dust deposition also decreases sharply towards the north, to 2.5 and 2.9 mg m^{-2} day^{-1} at M2 in 2013 and 2014, and towards the west to 3.7 mg m^{-2} day^{-1} at M3 in 2014, and 3.3 and 2.3 mg m^{-2} day^{-1} at M4 in 2013 and 2014 (Table 1). These fluxes are lower than the average 8 mg m^{-2} day^{-1} deposition estimates by Pabortsava et al. (2017) in the oligotrophic gyre north of M2. This sharp down-wind decrease was also observed in sediment traps offshore Mauritania by Bory et al. (2002), who found a decrease from 50 to 9 mg m^{-2} day^{-1}, at 500 and 1500 km from the coast, respectively.

Table 1. Average dust deposition fluxes in mg m^{-2} day^{-1} for all the sediment traps in 2013 and 2014. For 2014-M1 only the winter season was sampled completely.

sediment trap	2013-M1	2013-M2	2013-M4	2014-M1	2014-M2	2014-M3	2014-M4
annual average	14.3	2.5	3.3		2.9	3.7	2.3
spring average	9.4	1.2	1.9		2.3	1.9	1.3
summer average	22.6	3.3	4.7		2.8	4.0	3.2
autumn average	15.4	3.0	4.5		4.2	5.8	3.8
winter average	8.5	2.4	1.8	9.7	2.5	3.3	1.5

Satellite observations by CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) show that an estimated 182 Tg of dust is emitted from the African continent at 15°W toward the Atlantic Ocean annually (Yu et al., 2015). At 35°W this atmospheric dust load is reduced to 132 Tg, and at 75°W to 43 Tg, which translates into losses of 14.0 and 12.4 mg m^{-2} day^{-1} between 15–35°W and 35–75°W, respectively. Although the satellite observations exhibit a great uncertainty of ±45-70% (Yu et al., 2015), their estimates are very similar to the average dust deposition observed for the eastern Atlantic in the sediment traps at M1. However, in the central and western Atlantic the observed deposition fluxes in the sediment traps M2–M4 are considerably lower than the fluxes derived from CALIOP estimates (12.4 mg m^2 day^{-1}). Instead of deposition into the ocean, this decrease in atmospheric dust load derived from CALIOP estimates may have resulted from dispersal of the dust cloud out of the studied area. Also, the recent publication by Yu et al. (2019) presents dust deposition derived from different satellite products, which shows much higher deposition fluxes than observed in our sediment traps, for all four seasons, although the general seasonal and spatial trends are very similar. They also point out that the dust fluxes show high correlations between the different satellite products, but with large offsets in the magnitude of dust fluxes. They further state that dust deposition occurs further south than the observed DOD pattern due to the effective removal of dust by precipitation at the southern edge of dust plumes, related to the ITCZ (Yu et al., 2019). This highlights the importance of *in situ* observations of dust deposition over the Atlantic, since most open-ocean records of dust transport and deposition are either indirect observations, for example by remote sensing, or simulated by models, which both require validation against *in situ* records.

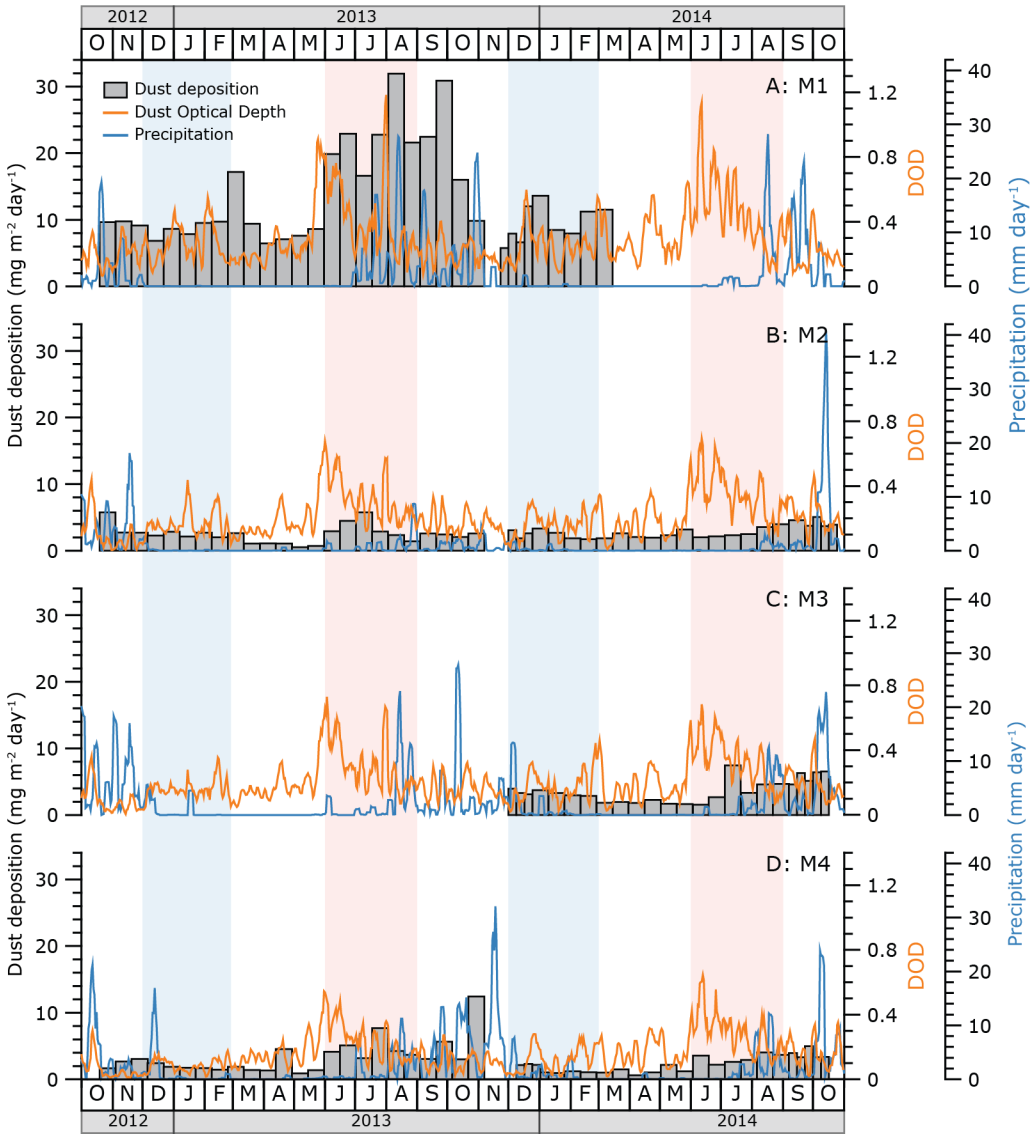

Figure 2. Time series of Saharan dust and precipitation across the Atlantic. A–D: Mass fluxes of Saharan mineral dust (particles >0.4µm) collected by sediment traps (bars), DOD (orange lines), and precipitation (TRMM; blue lines), at stations M1–M4. Width of the gray bars corresponds to the respective 8- or 16-day collection intervals. Blue and red areas mark winter and summer seasons, respectively.

Seasonality of dust deposition observed in the sediment traps is well defined across the Atlantic, with highest fluxes in summer and autumn, and lowest fluxes in winter and spring (Table 1). The seasonal amplitude decreases downwind, from east to west, with largest seasonal differences observed at station M1, in line with the downwind decrease in seasonality observed in the particle size of the deposited dust (Van der Does et al., 2016). Satellites have also observed highest dust fluxes in the Atlantic in summer, although contrary to the observations in the sediment traps, the satellites observed lowest fluxes in autumn and reaches less far west (Yu et al., 2019). Consistent with increased deposition fluxes in the traps, atmospheric dust loads measured as DOD were also highest in early summer (Fig. 2). As virtually no precipitation occurs along the transect from winter until June (Fig. 2), only dry deposition can account for the fluxes intercepted during this time by the sediment traps. Later in summer, DOD decreases at the same time when precipitation increases (June-July), although deposition fluxes in the traps remain high. This suggests that the dust is washed out of the atmosphere through precipitation (Fig. 2), resulting in low DOD values, but high (wet-)deposition fluxes. This

trend continued until November, and this anti-correlation between atmospheric dust load and dust deposition in some regions and seasons is consistent with model simulations (Mahowald et al., 2003). In winter, wet deposition likely occurs more to the south than the studied transect, close to the ITCZ (Yu et al., 2019).

If the dust is indeed deposited by wet deposition, the satellite data suggest a complete washout of atmospheric dust at the first incidence of precipitation, not directly related to the quantity or duration of precipitation (Fig. 2). Bory et al. (2002) also found wet deposition of dust to dominate in summer, on the eastern side of the Atlantic, and found that wet deposition mainly takes place in a few major events, during which the majority of the annual deposition is established. Such major wet-deposition events were also found by Neuer et al. (2004), with deposition fluxes of 160 mg m^{-2} day^{-1}. In short, from combining sediment trap and satellite data we infer low amounts of dry deposition in winter and spring, and a sharp increase at the beginning of summer. This is followed by high amounts of wet deposition from late June until November.

Station M2 is positioned about 160 km north of station M3 and received lower dust fluxes and less seasonality (Table 1), although multiple-year records, especially at M3, should be considered for a better understanding of seasonal differences between these sites. Less seasonality at M2 could result from the larger distance from the center of the Saharan dust plume. The position of the central axis of the main dust cloud is closely linked to the position of the ITCZ, which migrates seasonally (Ben-Ami et al., 2012; Meng et al., 2017).

Reported seasonality of dust concentrations and deposition is different on either side of the Atlantic, peaking in winter in the east (Ratmeyer et al., 1999; Bory et al., 2002; Neuer et al., 2004; Skonieczny et al., 2013; Fomba et al., 2014; Fischer et al., 2016), and in summer in the west (Jickells et al., 1998; Prospero et al., 2014). These differences can be related to seasonal atmospheric dynamics like the altitude at which the dust is transported, with dust at lower altitudes deposited close to the source in the east, while dust transported at higher altitudes can reach the remote western Atlantic (Van der Does et al., 2016; Yu et al., 2019). In addition, most of the studies on the eastern side of the Atlantic were carried out at latitudes above the main dust cloud and the northern position of the ITCZ, which could result in the opposing seasonal signal compared to the western Atlantic. The sampling method can also influence the seasonal signal of dust fluxes, as both dry and wet deposition are collected by the sediment traps, while only “dry” dust collected directly from the atmosphere is sampled at Barbados. The difference in dust deposition between sampling sites highlights the importance of dust collection between locations close to the source (eastern Atlantic) and further downwind (western Atlantic). The transect of sampled dust presented here shows year-round dust deposition across the Atlantic, peaking in summer most prominently at the eastern end of the transect.

3.3 Modeled dust deposition

When comparing the model results to the observed deposition fluxes in the sediment traps, it shows that the model performs well in terms of simulating the seasonality of dust deposition, but performs less well in terms of simulating the magnitude of dust deposition along this transect. As Albani et al. (2014) report, the model overestimates dust deposition and dust surface concentrations in the North Atlantic, but the overall seasonality of dust concentration is captured well. Given the overall uncertainties in modeling the mineral dust cycle with earth system models (Huneeus et al., 2011), the model can still be considered a useful tool to put the observations used in our study into perspective. The fluxes of total dust deposition (sum of wet and dry deposition, Fig. 3A) simulated by the earth system model show a similarly strong decrease from east to west, albeit more gradual than the

downwind decrease in fluxes observed in the sediment traps (Fig. 3B). More importantly, the model results show a sharp seasonal contrast between dry and wet deposition. Modeled wet deposition (blue bars) is highest in summer and autumn across the tropical Atlantic, related to precipitation activity along with the meridional ITCZ movement (Nicholson, 2000). Modeled dry deposition (red bars) is highest in winter and spring in the east and decreases westward in both magnitude and seasonal amplitude. These simulated deposition maxima in winter and spring (dry deposition) and late summer and autumn (wet deposition) in the Atlantic Ocean are corroborated by the sediment-trap observations and the incidence of precipitation, with the (wet) deposition maxima in summer being the highest (Fig. 3B). From east to west, the relative contribution of dry deposition to total deposition in winter and spring decreases, making wet deposition in summer and autumn the dominant modeled deposition mode in particular in the western Atlantic. It accounts for most of the total annual dust deposition there and determines the seasonal signature of total deposition. This result also matches dust deposition observed in Florida at the western side of the Atlantic, where wet deposition was found to dominate (Prospero et al., 2010).

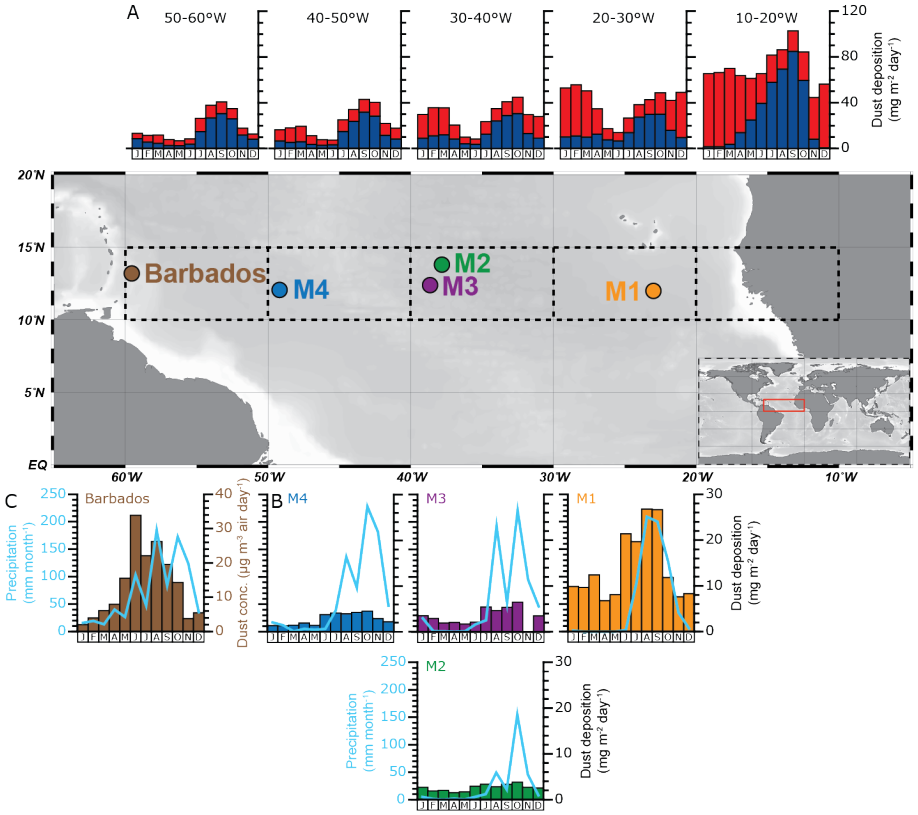

Figure 3. A: Simulated trans-Atlantic dust deposition fluxes (multi-year 5°×10° lat-lon averages in dashed boxes on map): dry deposition (red bars) stacked upon wet deposition (blue bars), their sum displays the total dust deposition (dry + wet). B: Annual cycle of dust deposition in the submarine sediment traps M1–M4 (bars; right Y-axes) with monthly precipitation (TRMM; blue lines; left Y-axes), for 2013–2014. C: Annual cycle of atmospheric dust concentrations at Barbados (bars; right Y-axis) and averaged monthly precipitation (TRMM, blue line; left Y-axis) for 2013–2014. The map was created using Ocean Data View (Schlitzer, 2018).

3.4 Dust concentrations at Barbados

On Barbados in the far western Atlantic, atmospheric dust loads are highest in summer and autumn (Fig. 3C), which is in line with the long-term observational record on Barbados (Prospero et al., 2014). Given that wet deposition dominates over the Atlantic during summer and autumn, the Saharan dust

arriving at Barbados is therefore likely the remaining dust not washed out by rains during its trans-Atlantic crossing, and to a smaller extent dry deposition. This is also evidenced by the atmospheric dust concentrations at Barbados that peak (June) before precipitation peaks across the Atlantic (August–October; Fig. 3). Prospero and Nees (1986) have initially found a relationship between Barbados dust concentrations and previous-year Sahel precipitation, although additional data of later studies did not support this relationship anymore (e.g. Prospero, 1996). Our results demonstrate that dust concentrations at Barbados could be affected not only by precipitation-related processes in the source region, but also by precipitation during long-range transport across the Atlantic Ocean, independent of dust generation and emission. Given our current findings, we argue that precipitation over the North Atlantic could be another important factor in influencing Saharan dust *transport* across the Atlantic and *concentrations* over Central America and the Caribbean. Investigating the relation between Barbados dust concentrations and rainfall over the Atlantic over a longer time scale should shed more light on the dominance of wet deposition over the Atlantic Ocean, and how this influences dust concentrations in the Caribbean.

3.5 Ocean fertilization

The results presented here show that most Saharan dust over the tropical North Atlantic Ocean is deposited with precipitation. Wet-dust deposition has important implications for transferring atmospheric dust-borne nutrients into the ocean, as low pH conditions in aerosols and clouds favor the leaching of nutrients from the dust particles (Desboeufs et al., 2001; Pulido-Villena et al., 2014; Korte et al., 2018). This may stimulate phytoplankton production, which in turn impacts atmospheric CO_2 levels (Mills et al., 2004; Guieu et al., 2014; Jickells et al., 2014; Pabortsava et al., 2017). Bottle-incubation experiments with Saharan dust additions along the same transect as in the present study have shown that Saharan dust indeed has the potential to deliver macro- and micronutrients (e.g. phosphate, silica, and dissolved iron) if exposed to acidic conditions before deposition (Korte et al., 2018). Not surprisingly, the concentration of nutrients released is in turn related to the amount of dust deposited, highlighting the importance for accurate dust flux measurements.

4. Conclusions

Our combined results of *in situ* observations of dust deposition fluxes, model simulations and remote sensing provide deeper insights into Saharan dust deposition across the Atlantic Ocean. Direct observations on dust deposition in such remote areas are sparse but essential for model calibrations, which underscores the uniqueness of the current dataset. We show that dust transport and deposition are highest during summer and demonstrate that wet deposition is the main mechanism for depositing Saharan dust into the tropical Atlantic Ocean: nearly all dust deposition in the western Atlantic originates from wet deposition during summer. As a result, dust arriving in the Caribbean is the remainder of dust escaping wash-out by precipitation along its passage from east to west. Wet-deposition events may lead to increased release of macro- and micronutrients that are important for the ocean's carbon cycle. Consequently, wet deposition of bio-available nutrients from mineral dust may be driving increased surface-ocean productivity, particularly in oligotrophic parts of the world's oceans. Considering future changes in global and regional precipitation patterns, enhanced droughts and the many uncertainties regarding the dust cycle, an improved understanding of these mechanisms is key to future climate predictions and to a better understanding of the impact of mineral dust on global carbon budgets through ocean fertilization. In addition, our findings could be used for the interpretation of sediment archives and long-term paleo dust records in the ocean.

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Data Availability

All data used in this manuscript are stored at https://doi.org/10.1594/PANGAEA.912241

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