

Dear Dr. Schwarz,

Thank you very much for handling the review process of our manuscript. We received three positive reviews from anonymous reviewers with constructive comments improving the readability and understanding of our manuscript. For this, we would like to thank the three reviewers, and we now acknowledge this in the manuscript.

Firstly, we received comments on the title of the manuscript, which is crucial for the public reading. We agree to the reviewers' comments and decided to change the title from: "Compositional changes in present-day transatlantic Saharan dust deposition", to: "Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic".

Secondly, the introduction of the manuscript was modified, concentrating on the uniqueness of our experimental set-up of simultaneous particle flux sampling across the entire Atlantic Ocean, including Saharan dust particles.

We received comments on the horizontal dust fluxes collected on the Mauritanian coast. Initially, we used these pure dust samples mostly showing that their element composition is very similar to that of the residual (lithogenic) mass fraction in our samples from the deep ocean, i.e. are derived from the same dust source. Relationships between particle size and the meteorology of dust deposition have recently been accepted as an ACP-Discussion paper by Friese et al., (doi:10.5194/acp-2017-131). However, in order for our manuscript to be clear as such, we added meteorological data to the dust fluxes to set these into context.

Furthermore, questions were raised on the XRF method we used to evaluate the element composition of the samples. We agree that we should elaborate on these XRF-analyses, since they provide a well-accepted and much applied easy and quick way to analyse wet sediment cores but have not been applied to dry sediment trap material before.

Additional concerns about the methods, analyses and calculations of the marine particle fluxes were taken into account and the appropriate parts of the manuscript were rephrased accordingly. In addition, more information is now provided to clarify the methods as such, also for a wider scientific audience. Lastly, minor comments were implemented to promote the readability.

Modifications in response to referee #1 are colored in red, to referee #2 in green and to referee #3 in blue. The comments of the reviewers are in normal text and our reply in italics. We hope you agree with us that the manuscript has much improved and merits publication in ACP.

On behalf of all co-authors, yours sincerely,

Laura Korte

Anonymous Referee #1

Comment Referee #1:

The present manuscript deals with the changes in Aeolian dust and marine fluxes on transect from Western Africa to the Caribbean. Sediment traps were deployed in several depths in the ocean, as well as a horizontal flux sampler was on the continent. Seasonal cycles are discussed. A lot of data material is presented and set into context of previous literature. The paper was a pleasure to read for clarity. Some spelling errors remain, which should be taken care of by the technical editing. I wonder though, whether the title is appropriate, as much of the manuscript deals rather with total mass fluxes and changes in total mass flux composition (including biogenic, carbonate), but a minor part only is dedicated to Saharan dust composition. Being on the atmospheric side, I can't comment on the oceanic sediment-specific techniques. A few minor remarks are made below; in particular the XRF data handling should be re-considered.

Reply to comment:

We agree that the title can be clearer, therefore we changed it to:

Changes made in manuscript:

“Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic”.

Comment Referee #1:

P6L9: How was the dust removed from the MWAC bottles – liquid suspension or dry?

Reply to comment:

When we receive the MWAC bottles from Iwik, they are most often also dusty on the outside, and this dust is first removed by rinsing and wiping the bottle from the outside. Afterwards the dust inside the bottles is shaken out by tapping on the bottle avoiding any liquids leading to possible chemical leaching of the dust.

Changes made in manuscript:

We adapted the following sentence in the manuscript:

Page 6, line 17 & 18

“From the land-based dust collectors, all dust was removed from each sample bottle by loosening and shaking the dry dust out of the bottles. The removed dust was weighed on a micro-balance.”

Comment Referee #1:

P9L17: Goossens and Offer (2000) give the efficiency for a size distribution with a mass median of 30 μm (geometric) (their Fig. 8). Van der Does et al. (2016) show mass median grain sizes between 4 and < 20 μm for the sediment traps. How was the size distribution in the MWAC samplers? If the mass median was considerably smaller than 30 μm , a significant overestimation of collection efficiency / underestimation of mass flux would need to be regarded; see Mendez et al. 2016, <http://dx.doi.org/10.1016/j.aeolia.2016.02.003>

Reply to comment:

The reviewer is right in this regard. The efficiency of the MWAC samplers depends on the particle size of collected material and as Mendez et al. (2016) showed, the efficiency decreases with decreasing particle size (PM₁₀ to PM₁). The median grain sizes of the dust presented in our manuscript, however, range between 25 and 50 μm with an average of 38 μm . Therefore, we rely on the efficiency range of 75 to 90 % given by Goossens and Offer (2000). More details on the grain-size distributions of the MWAC samples are now available in Friese et al., 2017 (ACPD).

Changes made in manuscript:

We adapted the following sentence in the manuscript:

Page 6, line 24 – 26

“The sampling efficiency of the MWAC samplers is between 75 % and 90 % for 30 µm dust (Goossens and Offer, 2000), which is within a similar size fraction of the Iwik dust (Friese et al., 2017).”

Comment Referee #1:

P7L33-35: As the intensity readings are used and it is calculated with, I would suggest not calling this qualitatively. It does not become clear from the manuscript, whether the procedure of Weltje and Tjallingii (2008) was performed (i.e. log ratios were calculated, relative detection efficiency and specific matrix calibration for the particular setup were obtained), or whether the raw intensity readings were used.

Reply to comment:

We concur with the Reviewer and clarified the section concerning the XRF analyses, in order to avoid confusion. We analyzed homogenized dry sediment trap samples, which largely avoids difficulties typical for XRF scanning on wet sediment cores, such as interference of interstitial water and/or sample roughness effects (as described in Tjallingii et al., 2007 and Weltje and Tjallingii, 2008). Therefore the intensities we measure closely resemble concentration data (i.e. follow linear calibration lines). To elaborate this we extended and adapted the section on XRF scanning of the dust samples. We now show that our XRF intensities closely resemble variability of concentration data, by addition of X-Y plots of concentration data to XRF intensities showing high correlations for our target elements (Appendix S1). Also, we replaced the reference to Weltje and Tjallingii (2008) to that of Tjallingii et al. (2007) to avoid the confusion about data processing, as Tjallingii et al. (2007) already shows that dry samples follow much better concentration data. The measurements we did on our dry dust samples can hence be approached statistically similar as concentration data.

Changes made in manuscript:

We changed the XRF paragraph in the method section:

Page 8, line 1 – 12

“The elemental composition of each sediment trap sample was determined by X-ray fluorescence (XRF) using the Avaatech XRF core scanner (Richter et al., 2006). This analytical technique has the important advantage that it is non-destructive, allowing that very small-size samples – such as sediment trap samples – can be used for other analyses after measurement. XRF scanning results in semi-quantitative compositional data (Richter et al., 2006), being expressed as intensities (i.e. counts or counts per second), which we normalize to the total counts to take into account the closed sum of geochemical data. We analyse our data as normalized element intensities, using the advantage that XRF-scan measurements on homogenised dry sediment trap samples largely avoid physical properties biasing, e.g. wet down-core XRF measurements (Tjallingii et al., 2007; Weltje and Tjallingii, 2008). For dry-powder samples, Tjallingii et al. (2007) showed that element intensities are proportional to their chemical concentration, which we confirm by measuring 13 standards with various matrices, including marine sediments that have a similar matrix to sediment trap samples (Supplement S1).”

Comment Referee #1:

P7L34-35: “these two elements”: Weltje and Tjallingii suggest a method based on two elements. If you normalize to the total counts here, you do right that, what Weltje and Tjallingii try to avoid by using two different elements. Please consider revising the data processing or write more clearly, if the method of Weltje and Tjallingii is used.

Reply to comment:

As described above (P7L34-35) we clarified the section about XR-scan analyses, elaborating on the method used and making clear that our data closely resembles concentration data.

Comment Referee #1:

P8L25: If there were wind speed measurements available at the MWAC station, it should be considered giving the information together with the horizontal mass flux, as averages or as times, where the wind speed was higher than an appropriate deflation threshold. That way, it could be estimated, whether emission was locally dominated.

Reply to comment:

We now downloaded monthly wind speed data around the MWAC station from Giovanni of the Goddard Earth Sciences Data and Information Services Center (GES DISC) provided by NASA. They show average speeds of around 3 to 7 m/s with higher wind speeds in summer and lower wind speeds in fall. For an accurate deflation threshold, we do not have enough information since the main factor affecting the threshold velocity is the interparticle cohesion force which is dependent, amongst others, on the particle size, density and soil moisture (Pye, 1987). However, according to Pye (1987), the threshold velocities for desert pavements are between 1.5 and 3 m/s. Since the average wind speeds are higher throughout the entire sampling period, we suggest that the captured dust is not only locally dominated and also might come from adjoining areas. For more information, see Friese et al. (2017), in which exactly these processes are described.

Changes made in manuscript:

We added the following sentences to the manuscript:

Page 9, line 10 – 16

“The total mass fluxes of the MWAC samplers at 290 cm height show significant month-to-month differences (Fig. 2). Highest dust fluxes of around 360 and 230 g m⁻² d⁻¹ were found during spring in April 2013 and during summer in August 2013, respectively. Local wind speeds at 10 m above the displacement height (MERRA model, NASA GES DISC) show monthly average velocities between 3 and 7 m s⁻¹ with higher wind speeds in early summer and lower wind speeds in fall. Throughout the entire sampling period, the wind speeds are above desert pavement threshold velocities of 1.5 to 3 m s⁻¹ (Pye, 1987), suggesting also dust contribution from adjoining areas.”

Comment Referee #1:

P9L3: This is actually an import.

Reply to comment:

This is an interesting remark, which probably results from the different “languages” that are used within the different scientific disciplines. Depending from which way you look at it, it might be import, yes. However, we talk about the export fluxes from the surface to the deep ocean which are intercepted by the sediment traps. Therefore, we leave it as ‘export’ since this is the common way in oceanography to express downward particle fluxes.

Comment Referee #1:

P9L15 and P9L38: If there are significant differences between the upper and lower traps recorded: where does the additional mass come from, where does the lost mass go to? The differences are not small.

Reply to comment:

There are differences in the upper and lower traps at site M2 and M4: there is less material caught in the lower trap in comparison to the upper trap at site M4 and there is more material collected by the lower trap in comparison to the upper trap at site M2. While the situation at site M4 shows the ‘normal’ situation, the situation at site M2 is challenging. The sediment traps are 2300 m apart, over which distance the sinking particles are potentially subject to a variety of processes, such as remineralization, disaggregation, or repacking by zooplankton. This happened at site M4 with the result in lower particle fluxes in the lower trap. However, in addition to the biological processes, advection of water displaces particles on the horizontal plane might take place, resulting in a greater catchment area of the sinking particles (Waniek et al. 2000). We attribute the additional mass in the lower trap at M2 to be the result of this greater catchment area of the lower trap.

Changes made in manuscript:

Page 22, line 2 – 7

“However, the settling pathway might be different, as indicated by the higher total mass fluxes in the lower trap than the upper trap at site M2. Over the 2300 m distance of the upper and lower trap, the sinking particles are potentially subject to a variety of processes including remineralisation, disaggregation, or repacking, as well as

horizontal movement of the particles, resulting in a greater catchment area (Waniek et al. 2000). Therefore, the enrichment in the lower trap at site M2 might result from the greater catchment area of the deeper trap (Siegel and Deuser, 1997;Waniek et al., 2000).”

Comment Referee #1:

Fig. 4 is on the small side. In particular the relative mass percent are difficult to compare. I suggest having an additional column of cumulative/stacked plots in white/gray/green/orange instead of the small separate plots. I would also suggest dividing the x axes by seasons instead of 2-month intervals, as seasons are discussed in the text. Moreover, vertical grid lines at the season boundaries would surely help to read particularly the upper plots.

Reply to comment:

We fully agree with the reviewer. The figure is on the small side but like this it is showing all the data on one page. We tested several options and we think that this is the best way to present the data. However, we acknowledge the suggestion of the seasonal grid which makes the traps more comparable. In addition, we provide the entire data set in a table on Pangaea for download.

Changes made in manuscript:

Seasonal grid added in Fig 4. (now 3) and Fig. 5 (now 4).

Comment Referee #1:

Fig. 5: What are the diamonds in the modal grain size?

Reply to comment:

The diamonds in figure 5 in the grain size data at site M3 were considered as outliers (van der Does et al. 2016).

Changes made in manuscript:

We added the following clause in the figure caption:

Page 15, line 3 to 5

“Black line represents modal grain sizes of Saharan dust of the same sample, diamonds in M3 are outliers (data Van der Does et al. 2016).”

Comment Referee #1:

P13L14: “Saharan dust is characterized by exclusively lithogenic elements”: I suggest “We identify Saharan dust by elements”, as in particular K in the atmosphere might have different sources (biomass burning), which might be not relevant in your case.

Reply to comment:

Yes, we agree.

Changes made in manuscript:

We adopted the mentioned suggestion in the manuscript:

Page 16, line 15 to 18

“We identify the residual mass fraction as lithogenic fraction by elements such as titanium (Ti), aluminium (Al), iron (Fe) and potassium (K). These elements are incorporated in mineral dust, in especially in aluminosilicates and feldspar, oxides and hydroxides and are incorporated in crystal lattice (Scheuven et al., 2013).”

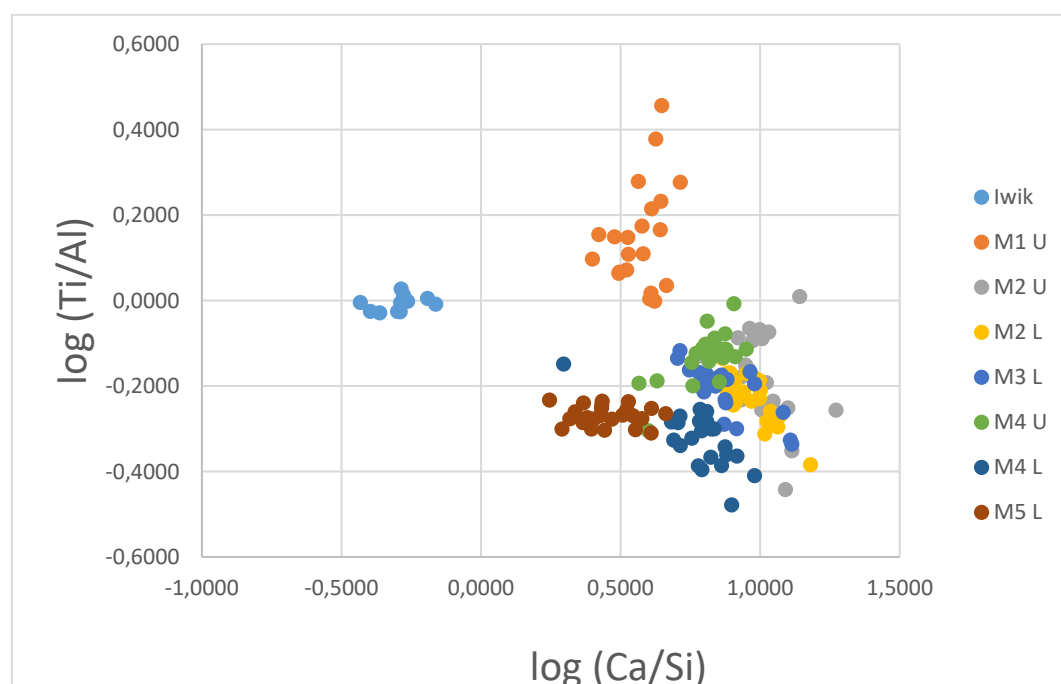
Comment Referee #1:

P13L19: If the procedure of Weltje and Tjallingii was implemented, the following comment can be neglected, otherwise: Correlating relative readings (‘normalized intensities’) can produce quickly spurious correlations, in

particular, if one of the major elements shows independent variations (e.g., Ca, Si). I highly suggest not doing that, but investigating elemental ratios instead; e.g., Ti/Al vs. Ca/Si.

Reply to comment:

We would like to demonstrate with the XRF data that the calculated residual mass fraction (TM - CaCO₃ - OM - BSiO₂) fully includes the lithogenic fraction in the sediment trap samples and is of the same composition as the pure Saharan dust samples at Iwik. This is best shown by plotting the individual lithogenic elements against the residual mass fraction (Fig. 7 (now 6) in manuscript). We explored the suggested ratios as well (see figure below) but we think that this only complicates the visualization since lithogenic (Ti/Al) and both lithogenic and biogenic elements (Ca/Si) are compared to each other. Therefore, we did not change it in the manuscript. Moreover, as we now strengthened our section on the XRF-scan measurements (see comment to P7L33-35), we are convinced that our measurements closely resemble concentration data, and as such a correlation of XRF data to residual mass fraction is warranted.



Comment Referee #1:

Fig. 9: What are the units on the y-axis?

Reply to comment:

Figure 9 (now 8) is showing the biogenic silica measured by sequential leaching (y-axis) versus the total silica measured by XRF (x-axis). Indeed, the y-axis was not properly labelled. It gives the biogenic silica as weight percentages (wt %).

Changes made in manuscript:

We changed the axis name in figure 8 (Page 19):

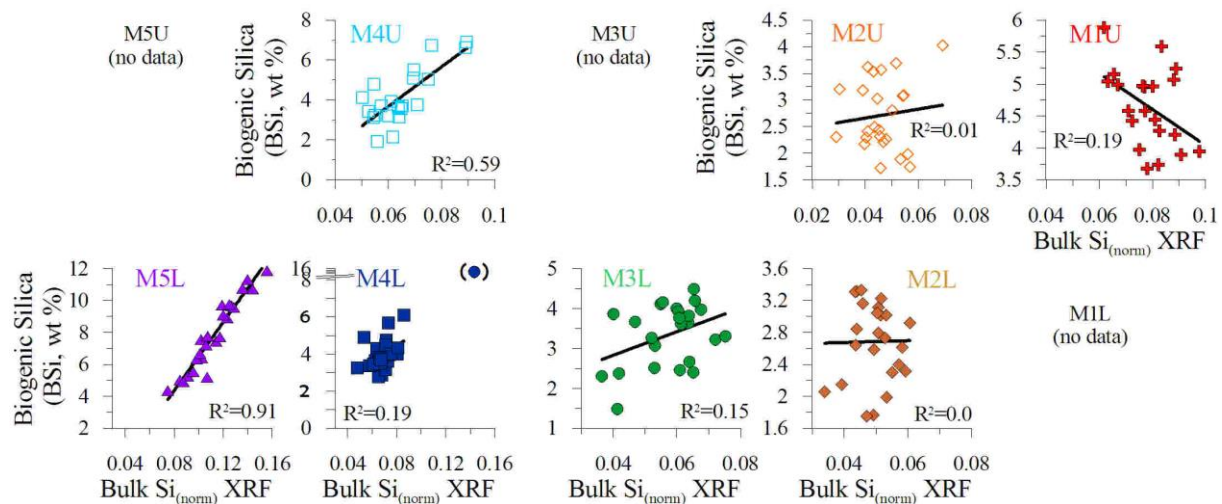


Fig 8. Biogenic silica versus total silica in particulate fluxes at five sites across the North Atlantic with sampling site M1 in the east (right) and M5 in the west (left).

Comment Referee #1:

P15L13: What does PSA mean?

Reply to comment:

PSA stands for potential source area, it was used in Scheuven et al. (2013), but was not yet is not clearly addressed in the manuscript.

Changes made in manuscript:

We added the missing term in the manuscript:

Page 19, line 7 – 9

“The land-based dust collectors in Iwik are located in the coastal region of western Mauritania, in potential source area 2 (PSA 2), which is one of the major source areas of dust that is transported across the Atlantic Ocean to the Americas (Scheuven et al., 2013).”

Comment Referee #1:

P15L18: How can the horizontal flux from the MWAC sampler can be compared to the vertical flux of the sediment traps? At least, information on wind direction is necessary here. Also, it seems to me rather improbable that horizontal surface flux on the Northafrican continent should be closely linked to ocean deposition near the Caribbean, as commonly a lot of dust is transported aloft in the Saharan air lay for longer distances (as detailed below in the manuscript).

Reply to comment:

The reviewer is correct; there is a difference between horizontal transport (Iwik) and downward deposition (marine traps) of dust. However, initially we thought that we might see an elevated dust flux in both the MWACs and the sediment traps (at least at site M1) in the same months due to high dust activity on the continent and a subsequent high dust flux in the atmosphere. Unfortunately, this simplification seems unjustified. We only see that the horizontal dust flux in the MWAC sampler is much higher due to its position on the continent. The wind speed is 3 to 7 m/s on monthly average, whereas we do not have information about the direction. Generally, the wind is blowing westwards from the Mauritanian coast which would transport the dust to the direction of site M1. The deposited residual flux at site M1 is due to mass loss much lower.

Changes made in manuscript:

We changed the following paragraph in the manuscript:

Page 19, line 13 – 15, page 20, line 1 – 4

“Overall, the horizontal Saharan dust flux from the land-based MWAC samplers in Iwik cannot be compared directly to the downward flux in residual mass in the sediment traps. However, they are compositionally the same, especially at the ocean site M1 as indicated by their similar Ti-Al slopes (Fig. 7). From site M1 we observe an overall decrease in both the residual mass and the marine biogenic matter fluxes westward to M2/M3 and M4.”

Comment Referee #1:

P15L20,25: By the notion ‘content’ in relation with XRF, reference is made to a linear contribution (mass, volume, moles). This seems in contradiction to the method section, where normalized intensities are referred to, which do not reflect directly a mass without proper calibration.

Reply to comment:

The wording is a contradiction; however, we show that the element counts are proportional to the element contents. Nevertheless, due to a missing quantitative calibration, which is beyond the scope of this paper.

Changes made in manuscript:

We changed the following sentences:

Page 20, line 4 – 11

“The much higher counts of lithogenic elements Ti, Al, Fe and K found in the pure dust at Iwik result from the absence of diluting biogenic matter produced in the marine realm. While the Ti-Al slopes are the same at Iwik and ocean sites M1 through M4U, and Al, Fe and K counts are most similar at Iwik and M1 as well, but become significantly higher towards the west (Fig. 7), it suggests a downwind change in mineralogical composition of Saharan dust. At ocean sites M4L and M5L, both the Ti/Al slope is lower and the Al counts are higher especially at M5, indicating that a second source in addition to Saharan dust is involved, which may be derived from admixture of re-suspended clay-sized sediments advected towards the deep sediment traps.”

Comment Referee #1:

P16L7: Particle size would play the primary role (over shape) in settling speed, so it should be termed like that instead of the ambiguous “lighter“. Moreover, the siliciumrich quartz and feldspar particles are usually found at larger particle sizes, which can be extracted from the referenced literature, e.g. the Kandler et al. (2009) paper. As result, this compositional change is consistent with the downwind fining.

Reply to comment:

Indeed, settling speed is dependent on particle size, size and density. The ‘lighter’ clay particles are, however, not unconditionally also finer. Moreover, they are kept in suspension more easily due to their flat shape.

Changes made in manuscript:

We exchanged ‘lighter’ with ‘finer’:

Page 20, line 16 – 20

“Such a downwind decrease in the relative amount of quartz may result from the increase of finer particles due to their slower settling speed, and a relative increase in platy clay minerals such as micas suspended in the atmosphere that can be transported over greater distances.”

Anonymous Referee #2

Korte et al. present an interesting account of the total mass and compositional fluxes of African dust across the Atlantic Ocean. This study is complimentary to the sizedependent transport study by van der Does et al. (2016). Generally, the authors report higher concentrations of mass closest to Africa and the Caribbean, yet the composition changes as dust is transported downwind, indicating different dust sources of the study area. Although the authors present a unique story for ACP, there are several issues that need to be addressed prior to publication in its final form.

Comment Referee #2:

General comments:

Previous studies have demonstrated atmospheric transport of North African dust is strongest in the summer (Jun – Aug), which is not consistent with the current work. Can the authors comment on why this inconsistency exists?

Chiapello, I., and C. Moulin (2002) TOMS and METEOSAT satellite records of the variability of Saharan dust transport over the Atlantic during the last two decades (1979– 1997), *Geophys. Res. Lett.*, 29(8), doi:10.1029/2001GL013767.

Perry, K. D., T. A. Cahill, R. A. Eldred, D. D. Dutcher, and T. E. Gill (1997) Longrange transport of North African dust to the eastern United States, *J. Geophys. Res.*, 102(D10), 11225–11238, doi:10.1029/97JD00260.

Reply to comment:

Apart from potential errors in satellite-derived quantifications of dust transport like for example the problems with clouds obscuring the visibility of dust, there are several possible reasons why we do not see such a pronounced seasonality in all our sediment traps or direct dust measurements on land as opposed to satellite imagery. First, we show the residual mass fluxes in which the dust is incorporated. This is the fraction which is calculated by subtraction of the biogenic particles from the total mass, and for now the best guess of dust fluxes when comparing to other sediment trap studies dealing with Saharan dust. At site M1, closest to the African continent, the residual mass fraction does show a clear seasonality with higher fluxes in summer and fall, confirming previous studies showing atmospheric Saharan dust transport strongest in summer (Chiapello et al. 2002). However, this pronounced seasonality at M1 becomes weaker with distance from the source, possibly attributed to changes in conversion factors in areas with less Saharan dust deposition. Another reason might be that there is a time lag between dust entering the surface ocean and dust-particle aggregates arriving at the sediment trap depths. The sediment traps collect all the aggregates (dust plus biological constituents) settling through the water column as soon as they are heavy and dense enough to sink. This may be in the order of a few weeks to a month. However, this would not change an entire season but makes it more challenging to compare atmospheric satellite data to deep ocean particle fluxes. For the biological part, seasonality in the equatorial Atlantic is generally weak due to the small changes in water temperature, sun light and nutrient availability.

Changes made in manuscript:

We added the following sentence to the results at site M1:

Page 10, line 11 – 17

“Consequently, the residual mass fraction shows the opposite, with a low relative abundance in winter and spring and a high in summer and fall. The variation of the residual mass flux is consistent with the long-term seasonal transport of Saharan dust above the eastern tropical North Atlantic (Chiapello and Moulin, 2002). The residual mass flux ranges from 32-71 mg m⁻² d⁻¹ with an average of 47.7 mg m⁻² d⁻¹ (17.4 g m⁻² a⁻¹) and slightly elevated values during summer and fall but with a peak in March (#9). A pronounced seasonality is also seen in the grain-size distributions of the same material with coarser grained Saharan dust in summer and finer grained dust in winter (Van der Does et al. 2016).”

Comment Referee #2:

The authors should more clearly link the spatiotemporal heterogeneity in the composition of the current work to the size-dependent depositional trends of van der Does et al. (2016), i.e., the smaller clays are transported farther while the larger quartz are deposited closer to Africa.

Reply to comment:

Yes, we can link the small clay particles in the west at site M5 to the aluminum and potassium, where we see a linear correlation.

Changes made in manuscript:

We added a sentence to the XRF ratios:

Page 16, line 22 – 26

“However, there are spatial gradients from east to west relating to a continuous enrichment of especially Al and Fe from M1 to M5, and to an offset of higher K at M5, while Ti stays constant. The modal particle sizes of the dust (Van der Does et al. 2016) do not show a relation to the lithogenic elements. Best relations found were in the west at site M5 where sizes and lithogenic elements are negatively correlated (Al, $R^2=0.54$ and K, $R^2=0.44$).”

Comment Referee #2:

The first paragraph of the introduction contains a nice summary of previous work, but it reads somewhat like a list of dust observation references with reporting their main findings. The first few sentences do contain this information, stating the quantities of dust measured, but what about the latter half of this paragraph? In order to demonstrate how this study is an advancement of and unique from previous work, discussing what these previous studies found is useful. Additionally, the introduction, in general, is lacking broader implications for evaluating Saharan dust transport. Without the broader impacts, the motivation behind this work is not evident.

Reply to comment:

We reorganized the introduction and broke it down to the main findings regarding Saharan dust sampling in the east on land, the atmosphere and in the ocean, as well as Saharan dust sampling at Barbados. The different Saharan dust transport mechanisms are introduced as well, about how they differ in altitude in winter and summer and have different path ways.

Changes made in manuscript:

Introduction, page 2 and 3

“Latest estimates of trans-Atlantic Saharan dust transport and deposition based on 3-D satellite imagery indicate that on a yearly basis (2007 – 2013, between 10° S and 30° N) an average amount of 182 Tg dust is blown from the northwest African coast at 15°W westward towards the Americas (Yu et al., 2015). Of this dust, about 132 Tg reaches 35°W and 43 Tg reaches 75°W (Yu et al., 2015). Due to the dust’s impact on global climate (e. g. Mahowald et al., 2014;Jickells et al., 2005;Griffin et al., 2001;Goudie and Middleton, 2001;Maher et al., 2010), Saharan dust has been examined extensively on either side of the equatorial North Atlantic Ocean, using ample different approaches. At the eastern side, northwest African *in-situ* measurements of dust events were performed on land (e. g. Kandler et al., 2009;Kandler et al., 2011;Marticorena et al., 2010;Kaly et al., 2015;Skonieczny et al., 2011;Skonieczny et al., 2013), in addition to ship board atmospheric dust sampling (e. g. Stuut et al., 2005;Baker et al., 2003) and deep ocean sediment fluxes (e. g. Bory et al., 2002;Ratmeyer et al., 1999b;Ratmeyer et al., 1999a;Fischer and Karakas, 2009). On the other side of the ocean, Prospero and colleagues have been sampling Saharan dust mainly on Barbados since the late 1960’s, resulting in the longest continuous time series of Saharan-dust sampling (Prospero et al., 1970;2014;Prospero and Lamb, 2003). All observations of Saharan dust showed a strong seasonality, with higher dust concentrations during the winter season close to the dust sources in the east, and higher dust concentrations during the summer season in the Caribbean. This seasonal pattern is related to the prevailing wind systems, which are influenced by the seasonal movement of the intertropical convergence zone (ITCZ). Saharan dust is transported year-round by the north-easterly trade winds at relatively low altitudes, carrying the dust to the proximal parts of the Atlantic Ocean (Stuut et al., 2005;Pye, 1987). In winter, easterly winds transport the dust in surface winds at altitudes below three km (Chiapello et al., 1995), when the ITCZ reaches its most southern position and the dust crosses the

Atlantic Ocean in the direction of South America (Prospero et al., 2014;1981). As the ITCZ migrates northward during summer, the dust is transported by the Saharan Air Layer (SAL) at higher altitudes up to five km (Tsamalis et al., 2013), and crosses the Atlantic Ocean above the trade-wind zone in the direction of North America and the island of Barbados (Prospero et al., 1970;2014).

The knowledge of the fate of Saharan dust in between the sources and sinks is, however, limited due to the vastness of the North Atlantic Ocean, though observed by remote sensing (e. g. Liu et al., 2008;Yu et al., 2015;Huang et al., 2010) and shipboard lidar measurements (Kanitz et al. 2014). From the latest approximation (Yu et al., 2015) it can be derived that around 50 Tg a⁻¹ of dust is deposited into the eastern equatorial North Atlantic Ocean between 15 and 35° W, and 140 Tg a⁻¹ of dust is deposited into the equatorial North Atlantic Ocean and the Caribbean Sea as well as onto parts of the Amazon rainforest. The dust deposited onto the ocean has a great influence on the vertical downward particle fluxes as well (Jickells et al., 2005;Ittekkot et al., 1992;Armstrong et al., 2009). Sediment trap studies within for example the Joint Global Ocean Flux Study (JGOFS) and Biogeochemical Ocean Flux Study (BOFS), dealing with deep ocean particle fluxes, show elevated total mass fluxes with a high contribution of lithogenic particle fluxes in the North Atlantic Ocean off Mauritania, (Jickells et al. 1996). In addition, Saharan dust particles are thought to contribute to the total particle flux in the Sargasso Sea (Deuser et al., 1988).

A 25-year time series of northwest African dust fluxes was established at the University of Bremen, Germany, using sediment traps moored on the Mauritanian continental slope. First results were presented by Wefer and Fischer (1993), Ratmeyer et al. (1999a; 1999b) and Nowald et al. (2015), followed by the 25-year record by Fischer et al. (2016). On average, lithogenic particles make up about a third of the total mass flux and up to 50 % during dust events (Nowald et al., 2015). Biogenic mass fluxes in this area are generally high as well, as the sediment traps are located in one of the four major Eastern Boundary Upwelling Ecosystems (EBUEs) (Fréon et al., 2009), bringing cold, nutrient-rich waters to the surface waters stimulating primary productivity. Fischer et al. (2016) demonstrate that the Cape-Blanc sediment-trap series show a weak relationship between dust input and productivity as reflected by the biogenic silica, mainly derived from diatoms as important primary producers. In addition, dust particles were found to strongly enhance the settling of organic matter through the water column by means of mineral ballasting, although no evidence was found for a relation between bulk fluxes and dust particle size.

In order to determine the simultaneous downward particle flux of the deposited Saharan dust across the entire Atlantic Ocean between the source in Africa and the sinks in the Atlantic Ocean and Caribbean Sea, we deployed an array of five moorings below the core of the dust plume, starting in 2012. Based on eight years of satellite observations, the Saharan dust plume is very consistently located between 7° and 17° N (Mulitza et al., 2008). Therefore, the array of moorings was positioned along the 12th northern parallel between 23° W and 57° W, each equipped with two time-series sediment traps. In addition, we positioned land-based dust collectors in Iwik on the Mauritanian coast nearest to the source (Fig. 1). Here, we present the initial set-up of the monitoring experiment and the first-year results of the mass fluxes and their composition as intercepted by the sediment traps in the Atlantic Ocean and the land-based samplers in Iwik. The results are compared to each other, and to satellite images of atmospheric Saharan dust transport, recorded by the Moderate Resolution Imaging Spectroradiometer (MODIS) carried by the Terra satellite. Complementary results of the size distributions of Saharan dust from the sediment traps are discussed by Van der Does et al. (2016), showing a strong seasonality with coarser grained dust in summer and finer grained dust in winter and spring, as well as a fining in particle size with increasing distance to the source.”

Comment Referee #2:

Clarification on some of the methods and calculations is needed, specifically pertaining to biogenic Si, the experimental setup with the mast and moored trap locations, and horizontal flux transport. See specific comments below:

Was BSiO₂ measured in the Iwik samples to serve as a control; to truly indicate that this Si was of a marine biogenic origin? How was this silica discernable from the mineral dust Si? What would be helpful is if the methods for extracting and detecting BSi contained more explanation, especially since this is not a technique one commonly encounters in an atmospheric journal. For instance, why the 660 nm laser; does it differentiate biogenic from mineral Si somehow, or is there something done in tandem to the diatom reference material? How does this differ from, say, XRF, in terms of measuring Si? More explanation would be helpful for someone who is not familiar with this technique.

Reply to comment:

The reviewer is correct, it would have been desirable to measure the biogenic silica content in the Iwik samples as well, if only to demonstrate there is no biogenic silica in these samples. However, we were not able to do so, due to insufficient material. For the used method around 100 mg of pure dust would be needed, is more than we have in the MWAC samples from Iwik.

The method to determine biogenic silica is well-accepted and much applied in marine sciences and is based on an alkaline (NaOH) leaching technique dissolving the amorphous silica (opal) in the presence of lithogenic silica minerals (Koning et al. 2002). Marine sediments surely consist of a number of silica fractions; of biogenic origin, clay minerals, aluminosilicates and quartz, from which the quartz and aluminosilicate minerals are virtually insoluble within the time span of a normal leaching experiment (60-90 min). For clay minerals it was shown that they dissolve at a constant, much slower rate and independently from the biogenic silica. Extrapolation of the linear 'clay dissolution line' to time zero would correct for the contribution of non-biogenic silica resulting in the BSiO_2 content of the sample (DeMaster 1981). The determination of dissolved silicon is based on the formation of a yellow silicomolybdate acid when an acid sample is treated with the molybdate solution (Grasshoff et al. 1983). The chemicals added for the used method are a sulphuric acid – molybdate solution, acidifying the 0.5 M NaOH to pH 2, allowing to form the molybdate complex, followed by oxalic acid, avoiding reduction of excess molybdate and ascorbic acid as a reductant to stabilize the blue complex. Eventually, the absorbance of the blue complex is measured at 660 nm in the spectrophotometer, which is the defined wavelength where no dilution is necessary (Grasshoff et al. 1983). As described above, the leaching technique is only measuring the biogenic silica while the XRF scanning determines the total silica fractions, including all the biogenic silica, clay minerals, aluminosilicates and quartz. The principle of the XRF analysis is based on excitation of electrons by incident X-radiation (Weltje & Tjallingii 2008). It is a semi-quantitative method which results are presented in form of element intensities (counts) and/or ratios of these element intensities. By contrasting both methods we showed the differences in composition with decreasing contribution of lithogenic silica towards the west to the extent that almost all silica is of biogenic origin in the west at site M5.

Changes made in manuscript:

We changed the method description as follows:

Page 7, line 19 – 26

“A standard amount of 25 – 30 mg of ground sample was placed in a 0.5 M NaOH solution at 85° C to dissolve the biogenic silica, which was subsequently reacting with a sulphuric acid-molybdate solution to form a blue molybdate complex. The complex was prevented from molybdate reduction and stabilized by adding oxalic and ascorbic acid, respectively. The solution was flushed through a photocell where the absorption of the blue complex was measured at the defined 660 nm (Grasshoff et al., 1983) and recorded every second. Each sample was run for 60 to 90 minutes. Results were evaluated with a weekly measured standard calibration curve ($R^2 > 0.99$) and calculated with the MS Excel data solver tool, extrapolating the dissolution curve to time zero to correct for contribution of non-biogenic silica (DeMaster, 1981).”

Comment Referee #2:

Horizontal flux transport indicates transport over a set distance, however, the authors present this factor as $\text{g m}^{-2} \text{d}^{-1}$, which indicates a time-dependent flux. I would assume both masts would be used to calculate the true horizontal flux of dust. Perhaps the wording is throwing me off here, but it seems as if this calculation is at one MWAC sampler as indicated on p8, 111, yet there were 20 samplers (2 masts with 5 pairs of samplers each). How far apart were these masts? Additionally, can the authors comment on how much of the dust flux is missed at higher altitudes, since the samplers extend up to 2.9 m but dust transport can happen up to 5 km?

Reply to comment:

The main interest of this study was to sample the local dust within the (accessible) source areas, which we can chemically compare to the dust we find in the sediment traps. The two dust masts are less than 30 m apart and hold 20 samplers in total. The given flux data is, however, only from the samplers at 290 cm height, in order to avoid sediment input by saltation. How much dust is missed on its way into the high atmosphere is difficult to estimate, and beyond the scope of our study. The amount is mainly dependent on the particle size and shape, keeping the dust particles in air suspension and allowing them to be uplifted to higher altitudes. We tried to

translate the amount of 182 Tg a^{-1} dust leaving the coast of North African given by Yu et al. (2015) to horizontal dust fluxes per m^2 . This simple conversion results in a dust flux of a few tens $\text{g m}^{-2} \text{ d}^{-1}$, assuming a homogeneous horizontal and vertical dust distribution (over 10° S to 30° N and 4 km or 7 km height) which is, however, impossible to compare with the horizontal mass flux in Iwik.

No changes made in manuscript.

Comment Referee #2:

For those of us not familiar with sediment traps, why were they located so deep and not closer to the surface where the dust would initially deposit and where the Northern Equatorial Current would carry dust from east to west? It seems like there would be a lot of room (literally) for different processes to remove the dust during sedimentation, like the deeper circulation from the conveyor belt in that region (N-to-S directionality). Since many readers will not have an oceanographic background, but are well versed in atmospheric transport mechanisms and how sensitive aerosol transport can be due to shifts in large and small scale circulation patterns, it would be helpful to provide a few sentences early on regarding Atlantic Ocean circulation and how this could potentially impact samples collected 2-3 km deep where the traps were located.

Reply to comment:

Sediment traps are a handy tool for direct and accurate time-series measurements of settling particles in the ocean when conditions are favorable (Knauer & Asper, 1989). Among these conditions are that current velocities are $< 12 \text{ cm/s}$, no deep eddy penetration and a stable vertical mooring cable, no biofouling or fisheries activities. These conditions are only met in the deep ocean. When sediment traps are too close to the ocean's surface, they suffer from surface currents suddenly changing the trap position, resulting in an unreliable particle flux. Around 99% of the material in the ocean is decomposed and recycled within the euphotic zone, only 1% of the produced particles escape to the deep ocean, which eventually account for the sequestered carbon. However, as the reviewer states, there is a lot of space for lateral transport affecting the total particle flux. Waniek et al. (2000) suggest that particles are moving with an angle of about 1° relative to the surface with a typical horizontal velocity in the range of $10\text{-}20 \text{ cm/s}$ and a particle sinking speed of $100\text{--}200 \text{ m/d}$. Therefore, it might be possible that the lower trap collects more or less material than the upper trap, depending on the pathway of the material. The best we can do is to obtain a thorough understanding of the mooring line motion using the current- and tilt meters attached to the moorings to evaluate the performance of the sediment traps. This is what we did and explained in the method section:

Changes made in manuscript:

[Page 4, line 12 – 14](#)

“Sediment traps are a common tool for direct and accurate time-series measurements of settling particles in the ocean when conditions are favourable, e.g. low currents ($< 12 \text{ m s}^{-1}$), no deep eddy penetration and a vertical mooring line (Knauer and Asper, 1989).”

Comment Referee #2:

For the calculations of carbonates and OM: Where does the 8.33 correction factor come from for CaCO_3 ? For OM, it seems like this calculation is likely underrepresenting total OM. Why isn't the TON included in OM? There is more than just C in organic matter (O, S, H, N to name a few; see references below as a couple examples), so if these aren't accounted for, the authors should clearly state the limitations and caveats with this calculation. And lastly, what is Corg and how was this measured/calculated (is it just TOC)?

Hazem S El-Zanan, Barbara Zielinska, Lynn R Mazzoleni & D. Alan Hansen (2009) Analytical Determination of the Aerosol Organic Mass-to-Organic Carbon Ratio, Journal of the Air & Waste Management Association, 59:1, 58-69, DOI: 10.3155/1047- 3289.59.1.58.

L. M. Russell (2003) Aerosol Organic-Mass-to-Organic-Carbon Ratio Measurements. Environmental Science & Technology 37, 2982-2987.

Reply to comment:

*The calculations used are the common way to measure and calculate deep ocean fluxes, which is a well-accepted and much applied technique in marine sciences, including for sediment trap samples. The total carbon (TC) and total organic carbon (TOC) were analyzed with an elemental analyzer. From these two analyses the total inorganic carbon (TIC) is calculated by subtracting the organic carbon from the total carbon ($TC - TOC = TIC$). TIC contains all calcium carbonate ($CaCO_3$), which is also the most concrete factor in this calculation. It is calculated as $CaCO_3 = TIC * 8.33$ based on the molecular weight of $CaCO_3$ ($Ca (40) + C (12) + O (16 * 3) = 100$), divided by the carbon measured ($C = 12$) resulting in $100/12 = 8.33$. For determination of the organic matter (OM), conversion factors vary since its molecular composition is highly heterogeneous and hence poorly constrained. Indeed, the reviewer names additional elements incorporated in organic matter besides organic carbon. Depending on which molecular structure one uses (OM simply as CH_2O , or the more sophisticated $(CH_2O)_{106} (NH_3)_{16} H_3PO_4$ derived from the global Redfield ratio), one ends up with different calculation factors. However, the most commonly used factor of 2 is chosen here for better comparison to other studies (Wefer & Fischer, 1993, Fischer et al. 2016, Neuer et al 2002) dealing with Saharan dust fluxes. C_{org} is the same as the TOC.*

Changes made in manuscript:

We adapted the method section on the carbon analysis as follows:

Page 7, lines 11 – 17

“Carbonates were calculated as $CaCO_3 = (TC - TOC) * 8.33$ and organic matter as $OM = 2 * TOC$. $CaCO_3$ is calculated as $(TC - TOC) * 8.33$, and organic matter as $OM = 2 * TOC$. The conversion factor for $CaCO_3$ is based on its stoichiometry, given 100 mol/g of $CaCO_3$ for 12 mol/g of carbon, resulting in a factor of $100/12 = 8.33$. The conversion factor for organic matter varies from 2.0 in the eastern Atlantic Ocean to 2.5 in upwelling areas (Jickells et al., 1998; Klaas and Archer, 2002; Thunell et al., 2007; Fischer and Karakas, 2009) due to poorly constrained composition of the actual organic matter. We chose to use the factor of 2 for better comparison to particle fluxes influenced by Saharan dust deposition off Cape Blanc (Fischer and Karakas, 2009; Fischer et al., 2007; Wefer and Fischer, 1993).”

Comment Referee #2:

Some clarification on the elemental components used for mineral classification is needed. For determining the elemental compositions of lithogenic and biogenic minerals, why is Ca not used? Wouldn't this corroborate the carbonates calculation from TOC and TC? For K, was biomass burning K removed? Central Africa is prone to fires and thus a large source of biomass burning aerosol (K is commonly used as a biomass burning marker). This could simply be resolved by calculating soil K, where $[non-soil\ K] = [K] - 0.6[Fe]$ and $[soil\ K] = [total\ K] - [non-soil\ K]$ from Kreidenweis et al. (2001).

Kreidenweis, S. M., Remer, L. A., Bruintjes, R., and Dubovik, O. (2001) Smoke aerosol from biomass burning in Mexico: Hygroscopic smoke optical model, J Geophys Res-Atmos, 106, 4831-4844.

Along these lines, what is the possible contribution from sea salt minerals (i.e., K) recrystallizing during sample processing to the elemental concentrations? Even though the samples were washed for salts, could the authors comment on how salts could potentially recrystallize onto insoluble particles during processing?

Reply to comment:

We used the XRF measurements to evaluate the lithogenic fraction in the sediment traps. We did not use Ca since in our samples it is almost exclusively biogenic in nature, formed by carbonate producing phyto- and zooplankton like coccolithophores and foraminifera. Consequently, also the Ca as measured by XRF analysis is positively correlated with the TIC measured in the sediment traps ($R^2=0.73$).

Indeed, K is used as a biomass burning marker. Our analysis does not distinguish between K-bearing lithogenic minerals (feldspars, clay minerals in the residual mass fraction) and K-bearing black carbon from biomass burning among the marine organic matter. All sediment trap samples are kept in their original liquid solution prior to processing and then washed three times with Milli-Q water prior to analysis, to remove any sea salts prior to drying. If there would be any sea salts left, it would be the same for every sample and would bias the samples equally. Given the high correlations between K, Ti and Al, the K would not be derived from any sea

salts since seawater concentrations of Al and Ti are in the nano-picomolar range, which also argues that any K contribution from black carbon associated with biomass burning is minor.

Changes in manuscript:

Page 16, lines 15 – 22

“We identify the residual mass fraction as lithogenic fraction by elements such as titanium (Ti), aluminium (Al), iron (Fe) and potassium (K). These elements are incorporated in mineral dust, in especially in aluminosilicates and feldspar, oxides and hydroxides and are incorporated in crystal lattice (Scheuven et al., 2013). Although K is also detected in biomass burning aerosols (Cachier et al. 1995), the high correlations between K and the other lithogenic elements show that the contribution of biomass burning K is minor in our samples. Normalized intensities of the lithogenic elements are highly correlated with the residual mass fraction and are therefore thought to represent the deposition flux of Saharan dust (Fig. 6).”

Comment Referee #2:

None of the figures show uncertainties/error bars. For instance, the methods section says the total mass fluxes (MWAC) have a 90% collection efficiency, so that remaining mass not collected should be accounted for in the measurement uncertainty. In order to clearly draw statistically significant relationships between the months, error bars should be provided.

Reply to comment:

Yes, the reviewer is right, errors should be accounted for. In general, the MWAC samplers are a good and widely-accepted tool to trap wind-blown sediments. However, the efficiency ranges have been demonstrated to vary. They are > 75 % and usually > 90 % according to Goossens and Offer (2000). For the revised manuscript we chose the minimum and maximum deviations, i. e. 75 and 90 %, and added those to the graph, in addition to wind speed data (Fig. 3 (now Fig. 2)). However, we would like to stress that the horizontal dust fluxes are only shown as a side information. The main purpose of these samples is the elemental composition, to which we want to compare to the sediment trap samples. For detailed information on the MWAC samples we would like to refer to Friese et al. (2017).

The errors for the marine particle fluxes are only given in the methods section. We are aware that they add up for the residual mass fraction since this is the fraction which is calculated from the remainder of the total to the biogenic particles. We do not show them in the figures, since it would make them unreadable and as this method is well-established (Wefer and Fischer, 1993; Neuer et al. 2002; Fischer et al. 2007), and most other literature only discusses average fluxes, we chose to leave them out from the figures as well.

Changes made in manuscript:

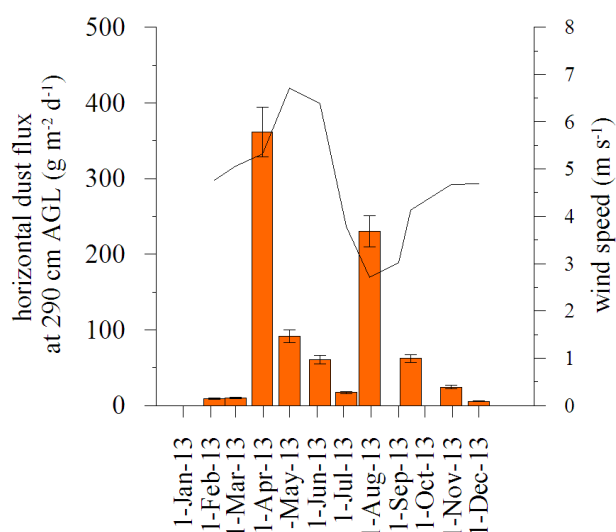


Figure. 2. Horizontal transport fluxes of Saharan dust from the land-based MWAC sampler in Iwik, Mauritania (19° N, 16° W) between January and December 2013 in orange bars. Error bars show the MWAC sampler efficiency of 75 to 90 %. Monthly averaged wind speed at 10 m above displacement height (MERRA model) around the Iwik location is indicated as a black line (data obtained from the Giovanni online data system, NASA GES DISC). AGL = above ground level

Comment Referee #2:

The seasonal maxima of the MWAC versus moored traps are offset. Can the authors comment on this in the discussion section?

Reply to comment:

The MWAC sampler and the sediment traps are two different kind of traps. The MWAC sampler is a really local aerosol sampler on land, collecting all indigenous transported dust events, while the marine sediment traps collect deposited dust. In addition, they do not sample at the same time intervals, which makes it even more difficult to compare. However, and that is what we would like to show here, the dust collected in both the MWAC aerosol samplers and the deep-ocean sediment traps have the same chemical composition, although they are so far apart from each other and sample a different process (transport versus deposition). A detailed description of the working of the MWAC samplers is beyond the scope of this paper and details are provided in Friese et al. (2017), ACPD.

Changes made in manuscript:

We adapted the following section in the discussion:

Page 19, line 13 – 15, page 20, line 1 – 4

“Overall, the horizontal Saharan dust flux from the land-based MWAC samplers in Iwik cannot be compared directly to the downward flux in residual mass in the sediment traps. However, they are compositionally the same, especially at the ocean site M1 as indicated by their similar Ti-Al slopes (Fig. 7). From site M1 we observe an overall decrease in both the residual mass and the marine biogenic matter fluxes westward to M2/M3 and M4.”

Comment Referee #2:

Specific comments:

P1, 117: The way this is written, it sounds like the authors are stating that the African coast is the source. Perhaps change to “African continent” or “North Africa”.

Reply to comment:

We agree on the comment.

Changes made in manuscript:

We changed the phrasing on page 1, line 19:

“Massive amounts of Saharan dust are blown from the coast of northern Africa across the Atlantic Ocean towards the Americas each year.”

Comment Referee #2:

P6, 18: To be clear up front, it would be helpful to provide a statement saying which analyses were conducted on the MWAC and which were conducted on the moored trap samples.

Reply to comment:

The reviewer is right, a statement on which analyses was applied to which samples is helpful and makes it easier for the reader to follow.

Changes made in manuscript:

We added the following paragraph at the beginning of the methods section:

Page 6, line 10 – 15

“The following paragraphs describe the different methods used to analyse particle fluxes and the element and biological composition of the specific samples. Due to the different nature and amount of material, i.e. the dry dust from the on-land dust collector in Iwik and the wet sediment trap samples from the deep ocean, not all methods were applicable to all samples. While for the marine sediments the total, biological and residual mass fluxes were determined, as well as their element composition, the Iwik samples were analysed exclusively for their total mass and horizontal dust flux at 290 cm as well as their element composition.”

Comment Referee #2:

P8, 12: State that lithogenic minerals are clays, quartz, and feldspars here. These are in fact listed but much later on.

Reply to comment:

We agree with the reviewer that the lithogenic minerals should be stated earlier.

Changes made in manuscript:

We rephrased the following sentence:

Page 8, line 20 – 24

“The elements Ti, Al, Fe and K were chosen and shown, since they are only present as main or minor elements in lithogenic minerals like clays, quartz, and feldspars, rather than in any biogenic mineral formed in the ocean, while Si represents both lithogenic Si minerals (clays, quartz, and feldspars) but also biogenic produced silica (BSiO₂), as found in phytoplankton diatoms or zooplankton radiolarians.”

Comment Referee #2:

P8, 16: Please provide the chemical compositions/components of these standards.

Reply to comment:

We recognized that the XRF-method needed more clarification. Therefore, we adopted the method section and provided the chemical composition of the XRF-standards in the appendix (S1).

Changes made in manuscript:

Page 8, line 1 – 12

“The elemental composition of each sediment trap sample was determined by X-ray fluorescence (XRF) using the Avaatech XRF core scanner (Richter et al., 2006). This analytical technique has the important advantage that it is non-destructive, allowing that very small-size samples – such as sediment trap samples – can be used for other analyses after measurement. XRF scanning results in semi-quantitative compositional data (Richter et al., 2006), being expressed as intensities (i.e. counts or counts per second), which we normalize to the total counts to take into account the closed sum of geochemical data. We analyse our data as normalized element intensities, using the advantage that XRF-scan measurements on homogenised dry sediment trap samples largely avoid physical properties biasing, e.g. wet down-core XRF measurements (Tjallingii et al., 2007; Weltje and Tjallingii, 2008). For dry-powder samples, Tjallingii et al. (2007) showed that element intensities are proportional to their chemical concentration, which we confirm by measuring 13 standards with various matrices, including marine sediments that have a similar matrix to sediment trap samples (Supplement S1).”

Comment Referee #2:

P8, 111-23: The addition of all the figures in parentheses in this paragraph place the figures out of order and cause confusion as they are not fully described (my eye was jumping back and forth between figures and text without enough clarification). It would be easier if references to these figures were eliminated here and remain discussed in the following sections. Additionally, this is a nice summary of what is to come, but perhaps providing a sentence at the end describing how all of these different observations will be used in tandem to develop the main picture “X” would clearly tie it all together.

Reply to comment:

We agree on the reviewer’s opinion that the reader have to jump back and forth between the figures and text.

Changes made in manuscript:

We accordingly adjusted the phrasing of the introductory paragraph of the results:

Page 8, line 34 – 41, page 9, line 1 – 8

“Horizontal transport fluxes from the land-based MWAC sampler are given in $\text{g m}^{-2} \text{d}^{-1}$. The vertical (downward) deposition fluxes from seven sediment traps deployed across the Atlantic Ocean are treated in downwind succession from east to west, starting at ocean site M1 closest to Africa, to ocean site M5 closest to the Caribbean. For each trap, the relative contribution of the biogenic components CaCO_3 , BSiO_2 , OM, and the residual mass fraction are given, in addition to the total mass flux, the flux of each biogenic component and that of the residual mass fraction, expressed both in $\text{mg m}^{-2} \text{d}^{-1}$ and $\text{g m}^{-2} \text{a}^{-1}$. The biogenic components are produced by autotrophic phytoplankton and heterotrophic zooplankton. In particular, phytoplankton CaCO_3 producers are mainly coccolithophores while zooplankton CaCO_3 is mainly from foraminifera and gastropod shells. The BSiO_2 is primarily produced by the phytoplankton diatoms and zooplankton radiolarians. Seasonal variations are shown for specific fluxes as deviation from their annual mean, together with the grain-size distribution of Saharan dust (Van der Does et al. 2016) for the residual mass fraction in the same material. To evaluate the residual mass fraction in the sediment traps, we compare XRF data from MWAC samplers with those from the sediment traps. The horizontal mass fluxes of the land-based dust collectors in Iwik are almost 100 % pure dust with negligible contribution of organic matter and fresh water diatoms from paleolakes and can therefore be used for chemical element comparison to the residual mass fraction from the sediment traps. Deviations between the chemical composition of the MWAC samplers and the sediment traps reveal compositional changes.”

Comment Referee #2:

P8, 126-27: Only Apr and Aug have the spikes (remove Mar and Jul). Fig 3.: Swap the y and x axes to align with how time series are shown in the rest of the figures.

Reply to comment:

We acknowledge the comment of the reviewer.

Changes made in manuscript:

We adjusted Figure 3 (now Figure 2) and added local wind data:

Page 9, line 10 – 16

“The total mass fluxes of the MWAC samples at 290 cm height show significant month-to-month differences (Fig. 2). Highest dust fluxes of around 360 and 230 $\text{g m}^{-2} \text{d}^{-1}$ were found during spring in April 2013 and during summer in August 2013, respectively. Local wind speeds at 10 m above the displacement height (MERRA model, NASA GES DISC) show monthly average velocities between 3 and 7 m s^{-1} with higher wind speeds in early summer and lower wind speeds in fall. Throughout the entire sampling period, the wind speeds are above desert pavement threshold velocities of 1.5 to 3 m s^{-1} (Pye, 1987), suggesting also dust contribution from adjoining areas.”

Comment Referee #2:

P9, 116: Why isn’t the lower trap shown? At the very least, provide it in the supporting information.

Reply to comment:

We are not sure what the reviewer means here. The text on mass fluxes at ocean site M2 (4.2.2) describes both the upper and lower trap, and so does the figure 4 (now 3) on page 11.

Comment Referee #2:

P13, 114-15: It can also contain Ca in the form of calcite (see Glaccum and Prospero (1980)), although at < 10%. A reference for this statement is needed.

Reply to comment:

Indeed, dust may contain Ca. However, the marine samples are dominated by biogenic Ca produced by calcifying plankton to the extent that any lithogenic Ca is heavily overprinted. Therefore, we used the lithogenic elements Ti, Al, Fe and K, rather than the Ca data for the evaluation of Saharan dust. XRD analysis show that lithogenic carbonates account for only 1.3 wt % in the Iwik samples (Friese et al. 2017). Nevertheless, we rephrased the sentence:

Changes made in manuscript:

[Page 16, lines 15 to 16](#)

“We identify the residual mass fraction as lithogenic fraction by elements such as titanium (Ti), aluminium (Al), iron (Fe) and potassium (K).”

Comment Referee #2:

P15, 111: First mention of DUSTTRAFFIC; should be introduced in the methods section.

Reply to comment:

We agree and removed it entirely.

Changes made in manuscript:

[Page 19, line 6 & 7](#)

“We provide the first comprehensive time series of biogenic particle fluxes and Saharan dust deposition from source to sink across the equatorial North Atlantic Ocean.”

Anonymous Referee #3

Comment Referee #3:

Thank you for your invitation to review the manuscript submitted to the ACP journal by Laura Korte et al. This paper reports on an ambitious sediment trap experiment carried out in the Tropical North Atlantic Ocean. This experiment, which was set up in the frame of the “DUST TRAFFIC” (ERC) project, aims at documenting Saharan dust inputs to the Atlantic Ocean from the West African coast all the way to the Caribbean, along the main path for Saharan dust atmospheric transport. Korte’s paper presents an impressive amount of data obtained during the first year of this experiment, including particle flux and composition temporal variability obtained from 7 sediment traps deployed at about 1100-1200m or 3400-3500m depths, at 5 mooring locations. The manuscript also includes data obtained on land in a dust source region in Mauritania. This study nicely complements earlier investigations carried out off West Africa and in the Sargasso Sea and provides, for the first time, simultaneous flux and compositional data across the entire Atlantic ocean. This is, I find, the main originality and interest of this report. In any case, this study represents a remarkable logistical and analytical effort, which is to be commended. One of the interesting outcome of this study is that there appears to be no marked seasonal variability in the downward particle fluxes across the entire Tropical Atlantic (a moderate seasonal variability is apparent on the edges of the transect though, near western and eastern margins, especially in the terrigenous fraction and the biogenic silica, respectively). This study also demonstrates the prominent influence of Saharan dust inputs in the “residual” (i.e., non-biogenic) material along most of the E-W transect (based on a few major element ratios (Al, Fe, K) that compare well with ratios measured in the dust they collected in Mauritania). “Dust” major element composition, still, displays a marked evolution across the ocean, enabling Laura Korte et al. to track the progressive mineralogical (and grain-size) sorting during transport (with the gradual reduction of coarse quartz particles contribution from East to West, as indicated for instance by the non-biogenic Si content). Korte et al. study finally reveals the influence of an additional terrigenous source in the Western Atlantic: major compositional differences are recorded in particles collected near the Caribbean -higher K contents for instance- suggesting a possible influence of fluvial discharges (Amazon and/or Orinoco) in the area. Overall, I find that the paper presents a large number of rare, high quality sediment trap data that will be of great interest to those interested in the Saharan dust transport (including modelers) and its impact on the ocean (and Amazonian) biogeochemistry. I therefore recommend publication after the authors have addressed the minor comments listed below.

Reply to comment:

This reviewer clearly has an oceanic background and is also interested in the oceanic consequences of dust deposition and is very aware of all the implications that are related to the marine organic fractions that are also collected by our sediment traps. However, given that the average ACP reader likely has an atmospheric background, in this paper we focus on the terrigenous sediment fraction rather than all the other marine organic fractions. We think that discussing all the marine organic fractions and their potential implications would make this manuscript unreadable and very long. Rather, we would like to draw the attention of the atmospheric community to our work, in an attempt to bridge the gaps that exist between the different disciplines. We believe that bringing them together will be of benefit for all disciplines. As a result, we try to highlight the Saharan-dust part of our studies and limit the oceanic part as much as possible, in order provide focus for the manuscript.

Comment Referee #3:

Detailed comments

page 2, line 24-25: Very few studies actually measured “fluxes” (in most cases, dust was collected by air filtration), so you may want to rephrase.

Reply to comment:

We reorganized the introduction in general and included your following suggestions.

Changes made in manuscript:

We changed the ‘fluxes’ into concentrations:

[Page 2, line 19 & 20](#)

“All observations of Saharan dust showed a strong seasonality with higher dust concentrations during the winter season close to the dust sources in the east, and higher dust concentrations during the summer season in the Caribbean.”

Comment Referee #3:

page 2, line 31: Sarthein et al. [1981] (which discusses paleo-dust records) might not be the best reference here – Chiapello et al., [1995] for example, which documents present-day seasonal Saharan dust transport in the trade wind levels, would be more suitable.

Reply to comment:

We thank the reviewer for the additional reference.

Changes made in manuscript:

We exchanged the reference regarding the Saharan dust transport:

Page 2, line 25 – 29

“In winter, easterly winds transport the dust in surface winds at altitudes below three km (Chiapello et al., 1995), when the ITCZ reaches its most southern position, and the dust crosses the Atlantic Ocean in the direction of South America (Prospero et al., 2014;1981).”

Comment Referee #3:

pages 2-3, bridging paragraph: Although there is no doubt that the Bremen group has built the longest sediment trap record in this region, providing one of the most valuable trap data time series ever collected –and should be praised for this!– other groups, in the UK and France for instance, have also contributed to ample sediment flux studies in the area (both off West Africa and in the Sargasso sea) within the 90’s BOFS and JGOFS programs among other frameworks. Some results obtained in these experiments are actually fully relevant to Korte’s study (see below). So, I think it would be useful to provide a more exhaustive list of earlier experiments (at least provide the references).

Reply to comment:

We reorganized the introduction and adapted the bridging paragraph.

Changes made in manuscript:

Page 2, line 32 – 40, page 3, line 1 & 2

“The knowledge of the fate of Saharan dust in between the sources and sinks is, however, limited due to the vastness of the North Atlantic Ocean, though observed by remote sensing (e. g. Liu et al., 2008; Yu et al., 2015; Huang et al., 2010) and shipboard lidar measurements (Kanitz et al. 2014). From the latest approximation (Yu et al., 2015) it can be derived that around 50 Tg a⁻¹ of dust is deposited into the eastern equatorial North Atlantic Ocean, and 140 Tg a⁻¹ of dust is deposited into the equatorial North Atlantic Ocean and the Caribbean Sea as well as onto parts of the Amazon rainforest. The deposited dust onto the ocean has a great influence on the particle fluxes as well (Jickells et al., 2005; Ittekkot et al., 1992; Armstrong et al., 2009). Sediment trap studies within for example the Joint Global Ocean Flux Study (JGOFS) and Biogeochemical Ocean Flux Study (BOFS), dealing with deep ocean particle fluxes, show elevated total mass fluxes with a high contribution of lithogenic particle fluxes in the North Atlantic Ocean off Mauritania (Jickells et al. 1996). In addition, Saharan dust particles are thought to contribute to the total flux in the Sargasso Sea (Deuser et al., 1988).”

Comment Referee #3:

page 3, line 12-14: I find this sentence somewhat unclear (the “other factors” have implication on grain size? on the dust deposition timing?) – please clarify.

Reply to comment:

Friese et al 2016 showed that grain size is not only dependent on precipitation (as suggested by Fischer et al., 2016) but also on wind speed, especially in winter, and on episodic dust storms which lead to anomalously coarse dust particles in the sediment traps off Cape Blanc.

This statement is however irrelevant in the reorganized introduction, since we do not mention it anymore.

Comment Referee #3:

Page 3, line 24: “Composition” is a little vague (you may want to specify here what has been characterized in your study: biogenic components (CaCO₃, BsiO₂, OM. . .), major elements in the residual (i.e., lithogenic) fraction (Si, Al, Fe. . .) etc.).

Reply to comment:

The ‘composition’ was referring to the dust sampled on Barbados and measured by ICP-MS (Trapp et al., 2010) and was not referring to our particle flux in this study.

However, in the reorganized introduction this observation does not occur anymore.

Comment Referee #3:

page 6, lines 20-24: You may want to indicate that sediment traps are supposedly collecting the downward (i.e., “vertical”) flux, especially since you just mentioned (line 10) that your land-based collector allows to estimate the “horizontal” dust flux.

Reply to comment:

Indeed, we should clarify that there is a difference in horizontal and vertical flux.

Changes made in manuscript:

We added the suggested vertical flux in the text:

Page 6, line 27 – 29

“For the vertical (downward) marine particle fluxes from the sediment traps, the samples were wet-sieved over a 1 mm mesh, wet-split in five aliquot subsamples using a rotary splitter (WSD-10, McLane Laboratories), washed to remove the HgCl₂ and salts, and centrifuged.”

Comment Referee #3:

page 7, line 34: Which “two” elements? Any? (in this case, you may want to remove “these” or rephrase).

Reply to comment:

We realized that the XRF method needed a bit more clarification and now spend a few more words on the XRF method, including standard reference material in the supplement:

Changes made in manuscript:

Page 8, line 1 – 12

“The elemental composition of each sediment trap sample was determined by X-ray fluorescence (XRF) using the Avaatech XRF core scanner (Richter et al., 2006). This analytical technique has the important advantage that it is non-destructive, allowing that very small-size samples – such as sediment trap samples – can be used for other analyses after measurement. XRF scanning results in semi-quantitative compositional data (Richter et al., 2006), being expressed as intensities (i.e. counts or counts per second), which we normalize to the total counts to

take into account the closed sum of geochemical data. We analyse our data as normalized element intensities, using the advantage that XRF-scan measurements on homogenised dry sediment trap samples largely avoid physical properties biasing, e.g. wet down-core XRF measurements (Tjallingii et al., 2007; Weltje and Tjallingii, 2008). For dry-powder samples, Tjallingii et al. (2007) showed that element intensities are proportional to their chemical concentration, which we confirm by measuring 13 standards with various matrices, including marine sediments that have a similar matrix to sediment trap samples (Supplement S1).”

Comment Referee #3:

page 8, line15: I find “daily averages” somewhat misleading (as it suggest that flux was sample with a daily or even sub-daily resolution); “daily mean” (and annual mean) might be less ambiguous; you may also simplify (e.g., “. . . each constituent, expressed both in $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and $\text{g}\cdot\text{m}^{-2}\cdot\text{a}^{-1}$ ”).

Reply to comment:

We thank the reviewer for the advice and simplified the expression accordingly:

Changes made in manuscript:

[Page 8, line 37 – 39](#)

“For each trap, the relative contribution of the biogenic components CaCO_3 , BSiO_2 , OM, and the residual mass fraction are given, in addition to the total mass flux, the flux of each biogenic component and that of the residual mass fraction, expressed both in $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and $\text{g}\cdot\text{m}^{-2}\cdot\text{a}^{-1}$.”

Comment Referee #3:

4.2.1 to 4.2.5 Mean annual fluxes (“annual averages”) for each component of the settling particulate matter (CaCO_3 , BSiO_2 , OM, and for the residual fraction) are of great interest too, and should also be provided for each trap (either in the text or in a table). I find such data are clearly missing in the manuscript. These would also allow discussing how they compare with results obtained at nearby locations in earlier studies (e.g., Bory and Newton, 2000; Bory et al., 2001), which may provide useful clues on the spatial (and/or interannual) variability in the Saharan dust deposition in the tropical Atlantic off West Africa.

Reply to comment:

Yes, we agree with the reviewer that these are of great interest, too. However, we wanted to focus on the residual mass fraction since this material includes all Saharan dust and is of most interest for this journal and audience. Providing the averages for all parameters for all traps would be too much detail. In addition, they will be the subject of a parallel manuscript. Still, we added a table with the yearly mass fluxes of the measured parameters showing the decreasing trend from east to west (M1 to M5) in numbers. In addition, these data set will be available from the Pangaea database after final publication.

Changes made in manuscript:

We added a table with yearly mass fluxes of all measured flux parameters.

[Page 11, line 28](#)

Table 3. Yearly mass fluxes of measured flux parameters.

Trap	Total Mass Flux	CaCO_3	BSiO_2	OM	Residual mass	CaCO_3	BSiO_2	OM	Residual mass
	$\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ($\text{g}\cdot\text{m}^{-2}\cdot\text{a}^{-1}$)	%				$\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ($\text{g}\cdot\text{m}^{-2}\cdot\text{a}^{-1}$)			
M1 U	111.6 (40.7)	33.9	9.7	13.6	42.7	37.8 (13.8)	10.9 (4.0)	15.2 (5.6)	47.7 (17.40)
M2 U	39.5 (14.4)	56.0	5.9	11.3	26.8	22.2 (8.1)	2.3 (0.8)	4.5 (1.6)	10.6 (3.9)

M2 L	54.6 (19.9)	59.2	5.7	7.2	27.9	32.3 (11.8)	3.1 (1.1)	3.9 (1.4)	15.3 (5.6)
M3 L	70.4 (25.7)	52.0	6.9	8.4	32.7	36.6 (13.4)	4.9 (1.8)	5.9 (2.2)	23.0 (8.4)
M4 U	84.8 (30.9)	48.3	10.8	12.6	28.3	41.0 (14.7)	9.2 (3.4)	10.7 (3.9)	24.0 (8.7)
M4 L	63.6 (23.2)	53.0	10.7	8.9	27.4	33.7 (12.3)	6.8 (2.5)	5.7 (2.1)	17.4 (6.4)
M5 L	115.6 (42.2)	32.5	16.5	5.7	45.3	37.6 (13.7)	19.1 (6.7)	6.6 (2.4)	52.3 (19.1)

Comment Referee #3:

4.4 Shouldn't the M5L (and M4L) compositional difference (K for instance) mentioned in this section?

Reply to comment:

We only mentioned the difference in Ti/Al for M5L and M4L, having a lower slope than M1U to M4U. For K the differences are not that obvious for M4L. Indeed, M5L shows differences, which we added in a clause:

Changes made in manuscript:

[Page 16, line 22 – 24](#)

“However, spatial differences from east to west indicate the continuous enrichment of especially Al and Fe from M1 to M5, and to an offset of higher K at M5, while Ti stays constant.”

Comment Referee #3:

section 5, page 5, lines 14-16: It seems as if you assume that dust collected at Iwik derives exclusively from local sources (during emission processes); couldn't distal sources also contribute (during deposition processes)? This should be discussed I think, especially as Iwik might be on the path of large outbreaks involving numerous source regions in West Africa.

Reply to comment:

In this manuscript, the main purpose of the Iwik samples was to compare the lithogenic elements with the element composition in the traps. This turned out to fit nicely, especially for site M1, which is closest to the African coast. We agree that the dust in the Iwik samples might be from distal sources as well. We looked at the backward trajectories which show different wind directions in summer and winter. The average accumulated dust, however, represents the dust we find back in the traps. For a detailed source identification, we propose to refer to Friese et al. (2017).

Changes made in manuscript:

We added the direction of the backward trajectories to the discussion:

[Page 19, line 10 – 15, page, line 1](#)

“The spring high can be related to the trade-wind intensities at lower altitudes, whereas the summer high point to sporadic dust storms, invisible on land by satellites. Backward trajectories reveal that the location in Iwik is a transit area for long-range transported Saharan dust (Friese et al. 2017). Overall, the horizontal Saharan dust flux from the land-based MWAC samplers in Iwik cannot be compared directly to the downward flux in residual mass to in the sediment traps. However, they are compositionally the same, especially at the ocean site M1 as indicated by their similar Ti-Al slopes (Fig. 7).”

Comment Referee #3:

Page, line 19: The fact there is a downwind decrease in the residual mass flux (and marine biogenic matter fluxes) is interesting but these data are not given in the text or in the table (unless I missed them), as I already pointed out above (4.2.1 to 4.2.5); so please provide numbers (for residual mass fluxes especially, but for the other components as well), especially as such decreases are clearly not obvious from the plots! (cf. fig. 4)

Reply to comment:

We added a table giving the annual fluxes of all components (Table 3). In addition, the entire data set will be available online on pangaea.de.

Comment Referee #3:

page 16, line 3: suggests

Reply to comment:

We corrected the typo.

Changes made in manuscript:

[Page 20, line 13](#)

“Overall, we observe a downwind increase in the Al and Fe content of the residual mass fraction, which suggests an increase in clay minerals relative to quartz.”

Comment Referee #3:

page 16, 5.1, line 17: Is it realistic that the event shown on fig. 10 (i.e., from 31 July) could be sampled in the 18 July- 3 August cups, considering the fact that it takes at least a week for the dust to reach the upper traps (as indicated page 18, lines 1-2)? Especially as dust travels at higher atmospheric levels at that time of the year, as you rightly point it out, and so that it may take longer for the dust to settle through the atmospheric column in the first place.

Reply to comment:

The reviewer is right, cup 19 at M1 would be probably more realistic. And indeed, cup 18 and 19 show higher fluxes in the residual mass fraction. Therefore, it does not change the interpretation as such. In addition to that, most of the dust events last for several days. The shown images show the days where the dust is best seen.

Changes made in manuscript:

We added cup #19 to the text:

[Page 20, line 27 – 29](#)

“Such enhanced fluxes in residual mass are found in intervals #16, #18 and #19 at site M1U, and M4U and #16 of M3L, corresponding to the period from 16 June to 2 July, from 18 July to 3 August, and 3 August to 19 August 2013, respectively.”

Comment Referee #3:

Section 5.1: Overall (for the reason just stated) I find this section should come after section 5.2, since indications on the settling speed in the water column is obviously essential to make the connection between satellite data and water column measurements (as this is actually acknowledged lines 32-33).

Reply to comment:

The current structure, satellite comparison before the downward transport, is from atmosphere to the ocean and is followed by the comparison of other deep ocean mass fluxes off northwest Africa. Therefore, we decided to keep the order of the sections as it is now.

No changes made in manuscript:

Comment Referee #3:

page 16, 5.1, lines 22-24: The point you want to make in this sentence (starting by “The diffuse. . .”) is unclear to me. Please clarify and a reference should be provided.

Reply to comment:

The diffuse dust cloud is referring to the wide-spread dust in the atmosphere in summer which is visible on satellite imagery. As satellite imagery is nadir, it integrates the amount of dust that is suspended across the whole atmosphere. For this reason, dust that is dispersed very thinly across a wide range of altitudes can be interpreted as high AOD, whereas a thin and dense layer could be interpreted as relatively low AOD.

No changes made in manuscript

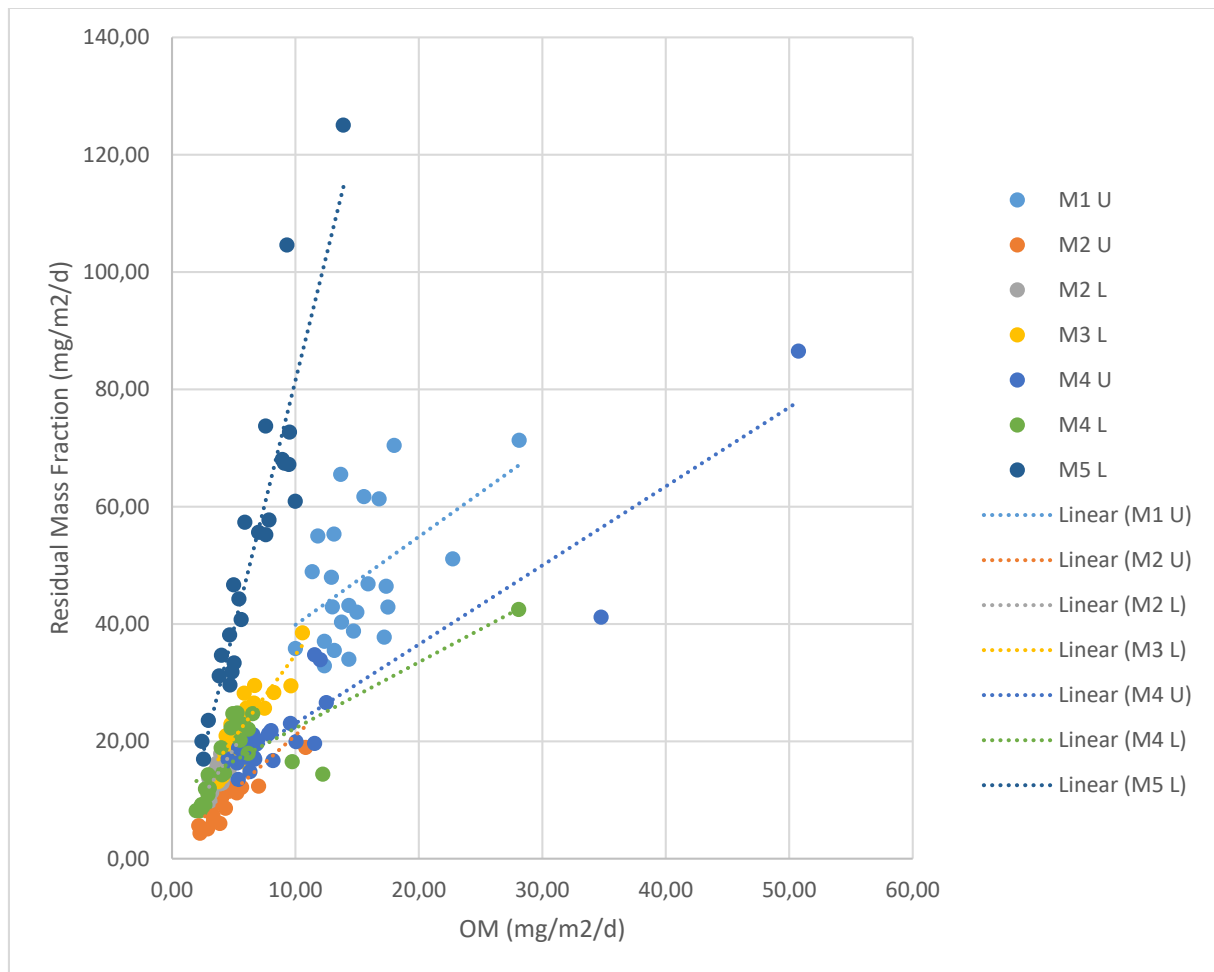
Comment Referee #3:

page 17, section 5.2, lines 7-8: This is an important point: as you rightly indicate, a significant part of the dust deposited in the ocean needs indeed to be incorporated with biogenic matter (OM in particular) in order to settle in the water column; for this reason, it would have been interesting to plot the residual flux vs the OM flux for all traps as this may provide useful clues on the interplay between the two in the particle settling processes in the different sectors of the Atlantic ocean (see for instance Deuser et al., 1983; Jickells et al., 1990; Bory et Newton, 2000. . . etc.).

Reply to comment:

We tried to avoid plotting the residual mass fraction vs the OM since one depends partially on the other, i.e. the residual mass fraction is the remaining part of the total mass after subtraction of the calcium carbonate, organic matter and biogenic silica. Therefore, it is always a closed sum, promoting artificial negative correlations. Also, given that the composition of the total mass is much less variable than its flux, regression of one constituent mass flux to the other, will produce positive correlations. Bory and Newton (2000) and Jickells et al. (1990), for example, used a different approach to quantify their lithogenic fraction in as much as they analysed the Al content independently and calculated the lithogenic fraction from that. This allows them to correlate the data to the organic matter. However, due to the reviewer’s curiosity, we provide the required plot and a table giving the linear regressions and the correlation coefficient R^2 below.

We did not incorporate the figure in the manuscript.



Trap	Linear regression	R ²
M1 U	$1.50x + 24.80$	0.25
M2 U	$1.89x + 2.17$	0.61
M2 L	$3.27x + 2.41$	0.57
M3 L	$2.86x + 6.16$	0.73
M4 U	$1.35x + 9.58$	0.89
M4 L	$1.12x + 11.01$	0.58
M5 L	$8.49x - 3.38$	0.86

Comment Referee #3:

page 18, section 5.3, first paragraph: As above, the discussion would benefit from the comparison with all existing data (comments on 4.2.1 to 4.2.5).

Reply to comment:

The yearly data of all fluxes are given in table 3 (page 11) and is discussed in sections 4.2.1 to 4.2.5. In addition, we added the following lines to the discussion:

Changes made in manuscript:

[Page 22, line 23 – 30](#)

“However, biogenic mass fluxes are about the same as found at M1, where no upwelling-stimulated productivity occurs. At site M1 all biogenic particle fluxes are highest, also in comparison to the other sampling sites. The only exception is the BSiO₂ at site M5, which is by far the highest contribution of BSiO₂. The low flux of biogenic particles in the mid-Atlantic Ocean (M2 to M4) reflect the limited availability of nutrients and low productivity in the oligotrophic ocean. The higher biogenic mass fluxes closest to either continent (M1 & M5)

may have been enhanced by the higher lithogenic input, especially from higher Saharan dust input at site M1, as mineral dust enhances the settling of organic matter through the water column (Ittekkot et al., 1992; Hamm, 2002).

Comment Referee #3:

page 18, section 5.3, lines 15-17: Additional data on ocean productivity (and its seasonal patterns) in the two regions should be provided in order to support such a statement; I would develop the discussion in order to strengthen this hypothesis, or remove this statement, which is too speculative as such.

Reply to comment:

With the addition above, dealing with the same issue, we changed the sentence:

Changes made in manuscript:

[Page 22, line 27 – 30](#)

“The higher biogenic mass fluxes closest to either continent (M1 & M5) may have been enhanced by the higher lithogenic input, especially from higher Saharan dust input at site M1, as mineral dust enhances the settling of organic matter through the water column (Ittekkot et al. 1992; Hamm, 2002).”

Comment Referee #3:

page 18, section 5.3, lines 28-31: I am not sure I follow the reasoning here: the contribution of an additional “fluvial” (and therefore fine) component on the western side of the transect seems perfectly consistent to me with westward decreasing trend in the grain size. A major fluvial contribution would also be fully consistent with the negative correlation between M1 and M5. So the statement that your grain size data implies that the residual fraction is dominated at all stations by Saharan dust seems to be at odd with the above (and with the last paragraph of page 18 actually).

Reply to comment:

We see the reviewer’s point. When you would only look at the grain size data with the decreasing trend from east to west, the fine particles at site M5 do fit in this trend perfectly. Therefore, from the grain size data alone, one would not expect to find fluvial input at first place. Now with the additional data on all marine particle fluxes, we see that site M5 is different from all other sites. With the fluvial input from the west, we can explain the high contribution in residual mass flux as well as the seasonal input of biogenic silica. Nevertheless, the Saharan dust does reach site M5 as well and contributes to the residual mass flux to some extent.

Changes made in manuscript:

We changed the following sentence in the manuscript:

[Page 23, line 2 – 5](#)

“Nevertheless, the particle-size distributions at M4 and M5 (Van der Does et al. 2016) alone do fit the general pattern of decreasing particle size with increasing distance from the Saharan dust source, not expecting fluvial input from the west.”

Comment Referee #3:

page 19, conclusions, line 4: “same” source may be little too affirmative and restrictive (replace with “. . . are similar to the dust collected on the African coast”?).

Reply to comment:

We accepted the suggested wording:

Changes made in manuscript:

[Page 23, line 16](#)

“We demonstrate that the lithogenic particles collected in the sediment traps are similar to the dust collected on the African coast.”

Comment Referee #3:

page 19, conclusions, line 10-11: As above, I see no contradiction here.

Reply to comment:

The reviewer is right. We deleted the clause arguing for the contradiction of the fluvial and aeolian source.

Comment Referee #3:

page 19, conclusions, lines 13-14: This is surprising, and therefore interesting, but the apparent correspondence should be considered with caution (see my comments on page 16, 5.1, line 17).

Reply to comment:

We suggest that the coarser particles settle out faster in both the atmosphere and the ocean water column. In summer, when more dust is in the atmosphere, which is also coarser than in winter, it is plausible that there is also a higher dust deposition onto the ocean. Still, the exact backtracking remains difficult due to assumptions on the dust's transport heights and settling speed in the atmosphere and the ocean. Note that we also find a higher residual mass flux in cup #19 (now added), which fits to this observation as well.

No changes made in manuscript**Comment Referee #3:**

page 19, conclusions, lines 15-17: It is unclear what [changing] “fluxes” [of Saharan mineral dust] you are referring to (and how it would be calculated). Also, is this underestimation discussed in the text?

Reply to comment:

The wording ‘changing’ is unclear, it is the changing chemical composition.

The complications with the calculation are stated in the method sections from which an underestimation of marine biogenic matter and a subsequently overestimation of the lithogenic mass flux is assumed. However, it is now stated more clearly with an additional sentence.

Changes made in manuscript:

We changed the wording in the conclusions.

[Page 23, line 27 – 30](#)

“While the temporal and spatial variability in residual mass fluxes corresponds to the changing chemical composition of Saharan mineral dust, they seem to overestimate the net fluxes due to underestimation of marine biogenic matter.”

Changes made in manuscript:

We added the following sentence to the method section:

[Page 7, line 38 & 39](#)

“Therefore, the residual mass fraction is most likely overestimated while the marine biogenic fraction is underestimated.”

Comment Referee #3:

Finally, I think the paper does not really document dust “deposition” as indicated in the title (this will surely be best documented by the buoys deployed within the DUSTTRAFFIC project). So you may want to reformulate (I would suggest something in the line of “Compositional changes of Saharan across the Atlantic as recorded in the water column downward particle flux”).

Reply to comment:

The floating buoys are active dust samplers, meaning that they suck in the dust-laden air from the atmosphere resulting in dust that is something between the transportation and deposition flux since they are only 2 m from the sea surface. However, we changed the title taking this reviewer’s suggestion into account.

Changes made in manuscript:

[Page 1, line 1 & 2](#)

“Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic”

Compositional changes of present-day transatlantic Saharan dust deposition

Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic

5

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Abstract. Massive amounts of Saharan dust are blown from the ~~African coast~~ of Northern Africa across the Atlantic Ocean towards the Americas each year. This dust has, depending on its chemistry, direct and indirect effects on global climate including reflection and absorption of solar radiation as well as transport and deposition of nutrients and metals fertilizing both ocean and land. To determine the temporal and spatial variability of Saharan dust transport and deposition and their marine environmental effects across the equatorial North Atlantic Ocean, we have set up a monitoring experiment using deep-ocean sediment traps as well as land-based dust collectors. The sediment traps were deployed at five ocean sites along a transatlantic transect between northwest Africa and the Caribbean along 12° N, in a down-wind extension of the land-based dust collectors placed at 19° N on the Mauritanian coast in Iwik. In this paper, we lay out the setup of the monitoring experiment and present the particle fluxes from sediment trap sampling over 24 continuous and synchronised intervals from October 2012 through to November 2013. We establish the temporal distribution of the particle fluxes deposited in the Atlantic and compare chemical compositions with the land-based dust collectors propagating to the down-wind sediment trap sites, and with satellite observations of Saharan dust outbreaks.

First-year results show that the total mass fluxes in the ocean are highest at the sampling sites in the east and west, closest to the African continent and the Caribbean, respectively. Element ratios reveal that the lithogenic particles deposited nearest to Africa are most similar in composition to the Saharan dust collected in Iwik. Down-wind increasing Al, Fe and K contents suggest a downwind change in the mineralogical composition of Saharan dust and indicate an increasing contribution of clay minerals towards the west. In the westernmost Atlantic Ocean, admixture of re-suspended clay-sized sediments advected towards the deep sediment trap cannot be excluded. Seasonality is most prominent near both continents but generally weak, with mass fluxes dominated by calcium carbonate and clear seasonal maxima of biogenic silica towards the west. The monitoring experiment is now extended with autonomous dust sampling buoys for better quantification of Saharan dust transport and deposition from source to sink and its impact on fertilization and carbon export to the deep ocean.

Keywords Saharan dust; North Atlantic Ocean; sediment traps; particle fluxes; XRF element ratios; transatlantic

1 Introduction

Latest estimates of transatlantic Saharan-dust transport and deposition based on 3-D satellite imagery indicate that on a yearly basis (2007 – 2013, between 10° S and 30° N) an average amount of 182 Tg dust is blown from the northwest African coast at 15°W ~~and transported~~ westward towards the Americas (Yu et al., 2015). Of this dust, about 132 Tg reaches 35° W and 43 Tg ~~a⁺~~ reaches 75° W (Yu et al., 2015). ~~meaning that around 50 Tg a⁺ of dust is deposited into the eastern equatorial North Atlantic Ocean between 10°S and 30°N and 140 Tg a⁺ of dust is deposited into the equatorial North Atlantic Ocean and the Caribbean Sea as well as onto parts of the Amazon rainforest.~~ Due to the dust's impact on global climate (e.g. Mahowald et al., 2014; Jickells et al., 2005; Griffin et al., 2001; Goudie and Middleton, 2001; Maher et al., 2010), Saharan dust has been examined extensively on either side of the equatorial North Atlantic Ocean, using ample different approaches. At the eastern side, northwest African *in-situ* measurements of dust events were performed on land (e. g. Kandler et al., 2009; Kandler et al., 2011; Marticorena et al., 2010; Kaly et al., 2015; Skonieczny et al., 2011; Skonieczny et al., 2013), in addition to ship board atmospheric dust sampling (e. g. Stuut et al., 2005; Baker et al., 2003) and deep ocean sediment fluxes (e. g. Bory et al., 2002; Ratmeyer et al., 1999b; Ratmeyer et al., 1999a; Fischer and Karakas, 2009). On the other side of the ocean, Prospero and colleagues have been sampling Saharan dust mainly on Barbados since the late 1960's, resulting in the longest continuous time series of Saharan-dust sampling ~~in the world~~ (Prospero et al., 1970; 2014; Prospero and Lamb, 2003). All observations of Saharan dust showed a strong seasonality with higher dust ~~fluxes concentrations~~ during the winter season close to the dust sources in the east, and higher dust ~~fluxes concentrations~~ during the summer season in the Caribbean. This seasonal pattern is related to the prevailing wind systems, which are influenced by the movement of the intertropical convergence zone (ITCZ). Saharan dust is transported ~~during any time of the year round~~ by the ~~northeasterly~~north-easterly trade winds at relatively low altitudes, carrying the dust to the proximal parts of the Atlantic Ocean (Stuut et al., 2005; Pye, 1987). In ~~winter addition~~, easterly winds transport the dust in surface winds at altitudes below three km (Chiapello et al., 1995), when the ITCZ reaches its most southern~~ly~~ position, ~~the Harmattan trade winds become more important. The Harmattan trade when and~~ the dust crosses the Atlantic Ocean in the direction of South America (Prospero et al., 2014; 1981). As the ITCZ migrates northward during summer, the dust is transported by the Saharan Air Layer (SAL) at higher altitudes up to five km (Tsamalis et al., 2013), and crosses the Atlantic Ocean above the trade-wind zone in the direction of North America and the island of Barbados (Prospero et al., 1970; 2014).

The knowledge of the fate of Saharan dust in between the sources and sinks is, however, limited due to the vastness of the North Atlantic Ocean, though observed by remote sensing (e. g. Liu et al., 2008; Yu et al., 2015; Huang et al., 2010) and shipboard lidar measurements (Kanitz et al. 2014). From the latest approximation (Yu et al., 2015) it can be derived that around 50 Tg a⁻¹ of dust is deposited into the eastern equatorial North Atlantic Ocean, and 140 Tg a⁻¹ of dust is deposited into the equatorial North Atlantic Ocean and the Caribbean Sea as well as onto parts of the Amazon rainforest. The deposited dust onto the ocean has a great influence on the particle fluxes as well (Jickells et al., 2005; Ittekkot et al., 1992; Armstrong et al., 2009). Sediment trap studies within for example the Joint Global Ocean Flux Study (JGOFS) and Biogeochemical Ocean Flux Study (BOFS), dealing with deep ocean particle fluxes, show elevated total mass fluxes with a high contribution of lithogenic particle fluxes in the North

Atlantic Ocean off Mauritania; (Jickells et al. 1996). In addition, Saharan dust particles are thought to contribute to the total flux in the Sargasso Sea (Deuser et al., 1988).

A 25-year time series of northwest African dust fluxes was established ~~by colleagues~~ at the University of Bremen, Germany, using sediment traps moored on the Mauritanian continental slope. First results were presented by Wefer and Fischer (1993), Ratmeyer et al. (1999a; 1999b) and Nowald et al. (2015), followed by the 25-year record by Fischer et al. (2016). On average, lithogenic particles make up about a third of the total mass flux and up to 50 % during dust events (Nowald et al., 2015). Biogenic mass fluxes in this area are generally high as well, as the sediment traps are located in one of the four major Eastern Boundary Upwelling Ecosystems (EBUESs) (Fréon et al., 2009), bringing cold, nutrient-rich waters to the surface waters stimulating primary productivity. ~~The biogenic matter consists of organic matter (OM) as well as marine biominerals of CaCO_3 and amorphous silica (BSiO_2) which are produced by autotrophic phytoplankton and heterotrophic zooplankton. In particular, phytoplankton CaCO_3 producers are mainly coccolithophores while zooplankton CaCO_3 is mainly from foraminifera and gastropods shells. The BSiO_2 is primarily produced by the phytoplankton diatoms and zooplankton radiolarians.~~ Fischer et al. (2016) demonstrate that the Cape-Blanc sediment-trap series ~~did show~~ a weak relationship between dust input and productivity as reflected by the biogenic silica, mainly derived from diatoms as important primary producers. In addition, dust particles were found to strongly enhance the settling of organic matter through the water column by means of mineral ballasting, although no evidence was found for a relation between bulk fluxes and dust particle size. ~~Throughout the last 25 years, the bulk fluxes off Cape Blanc varied strongly on seasonal and interannual timescales but without any significant relationship to known large scale climatic oscillations (Fischer et al., 2016). As their sediment traps are located relatively close to the Mauritanian continental shelf, likely also lateral input from re-suspended shelf sediments was collected, propagating down the slope in nepheloid layers (Fischer et al., 2009).~~

In order to determine the simultaneous downward particle flux transport and of the deposition of Saharan dust across the entire Atlantic Ocean between the source in Africa and the sinks in the Atlantic Ocean- and Caribbean SeaOceans, we deployed an array of five moorings below the core of the dust plume, starting in 2012. Based on eight years of satellite observations, the Saharan dust plume is very consistently located between 7° and 17°N (Mulitza et al., 2008). Therefore, the array of moorings was positioned along the 12th northern parallel between 23°W and 57°W, each equipped with two time-series sediment traps. In addition, we positioned ~~a~~ land-based dust collector~~s~~ in Iwik on the Mauritanian coast nearest to the source (Fig. 1). Here, we present the initial set-up of the monitoring experiment and the first-year results of the mass fluxes and their composition as intercepted by the sediment traps in the Atlantic and the land-based samplers in Iwik. The results are compared both with each other, and satellite images of atmospheric Saharan dust transport, recorded by the Moderate Resolution Imaging Spectroradiometer (MODIS) carried by the Terra satellite. Complementary results of the size distributions of Saharan dust from the sediment traps ~~are were~~ discussed by Van der Does et al. (2016), showing a strong seasonality with coarser grained dust in summer and finer grained dust in winter and spring, as well as a fining in particle size with increasing distance to the source.

2 Instrumentation and Performance

2.1 Land-based dust collectors

Saharan dust transport and deposition is traced from source to sink, starting on the Mauritanian coast with two masts with passive modified Wilson and Cooke samplers (MWAC dust collectors; Goossens and Offer (2000), located in Iwik, (19°53.11'N, 16°17.64'W), which have been sampling dust over monthly intervals since January

2013. Each mast contains a total of ten MWAC samplers, deployed in pairs at five heights, equally spaced at ± 90 to ± 290 cm from the ground. The masts contain a wind vane that directs the opening of the MWAC collectors into the wind.

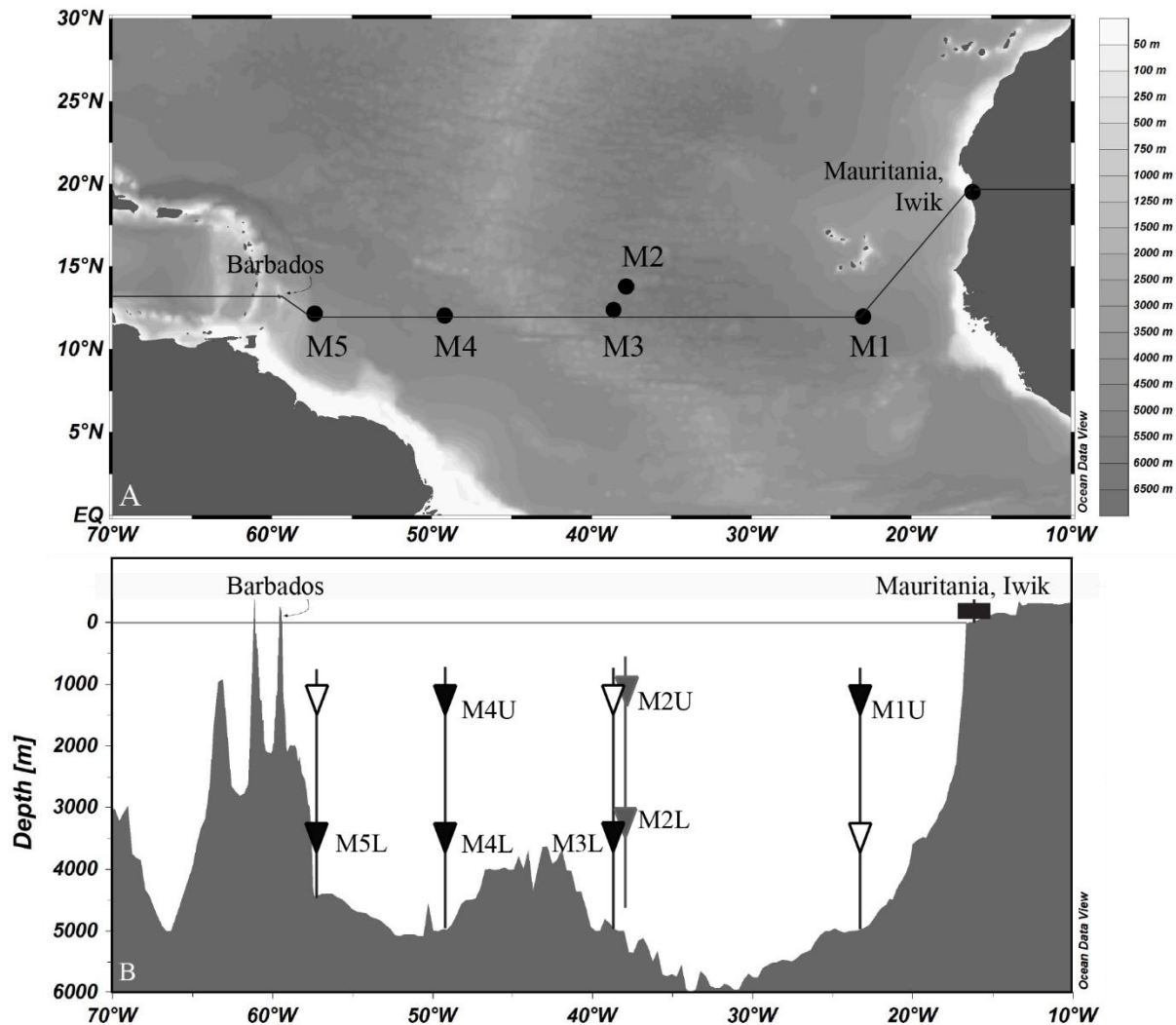


Figure 1. A: Location of land-based dust collectors on the Mauritanian coast in the village of Iwik and sediment trap moorings M1 – M5 between northwest Africa and Barbados. B: Bathymetric profile, on the line in A, through the equatorial North Atlantic between Mauritania (15° W) and Barbados (60° W) showing the five moorings with sediment traps (triangles) and the land-based MWAC dust collectors (rectangle). MxU and MxL indicate the upper and the lower traps on the five moorings at 1200m and 3500m nominal water depth, respectively. Open symbols refer to sediment traps that failed or were lost during recovery.

2.2 Ocean moorings

Sediment traps are a common tool for direct and accurate time-series measurements of settling particles in the ocean when conditions are favourable, e.g. low currents ($< 12\text{ m s}^{-1}$), no deep eddy penetration and a vertical mooring line (Knauer and Asper, 1989). Moorings were deployed at five sites (M1 – M5) along a transect in the equatorial North Atlantic during RV Meteor cruise M89 (Stuut et al., 2012) in October 2012 (Fig. 1). Four of the five moorings (M1, M3, M4 and M5) were deployed at 12°N and 23°W, 38°W, 49°W and 57°W, respectively, and one mooring (M2) was positioned to the north of station M3 and deployed at 13.5°N and 37.5°W to assess potential north-south movements of the Saharan dust plume. Each mooring was equipped with a number of oceanographic instruments (Table 1). These include two Technicap PPS 5/2 sediment traps provided with a tilt meter at a nominal water depth of 1200 m (upper) and 3500 m (lower), two SBE MicroCat CTDs for conductivity,

temperature and depth measurements, two Aanderaa RCM-11 current meters, and four float~~ation~~ ~~bodies~~ to keep the mooring upright. The uppermost float included a downward-looking Acoustic Doppler Current Profiler (ADCP, 75 Hz) for measuring current profiles (velocity and direction) and particle backscatter intensities, and a XEOS iridium beacon and flasher on top.

5

Table. 1. Layout of moorings' instruments as an example of sampling site M1

Instrument name	Description	Approx. W water depth (m)
Benthos floats	Floats	720
Smartie float 500	Float with beacon + flasher + downward looking ADCP	750
SBE MicroCat CTD	CTD – Conductivity Temperature & Depth sensor	1190
Technicap PPS 5/2	Sediment trap with 24 bottles + tilt meter	1200
Smartie float	Float with 500kg buoyancy	1250
Aanderaa RCM	Current meter	3480
SBE MicroCat CTD	CTD – Conductivity Temperature & Depth sensor	3490
Technicap PPS 5/2	Sediment trap with 24 bottles + tilt meter	3500
Benthos floats	Floats to retrieve mooring	40m above bottom
Aanderaa RCM	Current meter	20m above bottom
Releasers	To detach mooring from anchor	10m above bottom
Anchor	Steel weight 2000kg	bottom

Generally, current velocities did not exceed 12 cm s^{-1} , which is considered a threshold for unbiased collection of settling particles (Knauer and Asper, 1989). Current-meter measurements showed that the average velocities around each mooring were $< 6 \text{ cm s}^{-1}$ in the deep and bottom ocean at around 3500 and 4600 m water depth, respectively and $< 10 \text{ cm s}^{-1}$ ~~at around 1200 m water depth in the upper ocean~~ as measured by the ADCPs ~~at around 1200 m water depth~~. For only a few days during the entire sampling period current velocities exceeded the 12 cm s^{-1} for the upper sediment traps at 1200 m at the sites M2 and M3, while at M4 maximum velocities at 3350 m reached 19 cm s^{-1} in early August 2013. All other sensors showed that the sediment traps at sites M1 to M4 remained well within 5° from the vertical and at constant depths during the entire sampling period. Only during two periods at ~~sitetation~~ M5 current velocities exceeded 12 cm s^{-1} to the extent of biasing collection efficiency: in February 2013 and late March 2013. This caused a downward movement of the traps as recorded by increased pressure (depth) by both CTDs, as well as deviations of up to 14° and 12° from the vertical, respectively, as measured by the tilt meters, affecting three sample intervals at station M5 (intervals #8, 10 and 11). For these three intervals we adjusted the total mass flux using $F = F_t - (1 + 1.4 \sin \theta)$ with F being the vertical flux, F_t the flux in the tilted trap, and θ the degree of tilt from the vertical (Gardner, 1985).

Each sediment trap has a collecting area of 1 m^2 and is equipped with an automated rotary collector carrying 24 sampling cups. All traps collected the settling particles simultaneously and synchronously in 16-day intervals, starting on October 19, 2012 and ending on November 7, 2013. Prior to deployment in 2012, sampling cups had been filled with filtered seawater from the respective sampling depths at 1200 m and 3500 m, using HgCl_2 as a biocide (to an end concentration of 1.3 g/L) and Borax ($\text{Na}_2[\text{B}_4\text{O}_5(\text{OH})_4] \cdot 10\text{H}_2\text{O}$; end concentration 1.3 g/L) as a buffer at pH 8.5. Both additives increased the density ~~to slightly higher than seawater~~ to limit exchange with ambient seawater. Seven sediment traps were recovered successfully during RV Pelagia cruise 64PE-378 in November 2013 (Stuut et al. 2013). These include three upper (1200 m) sediment traps at sites M1, M2 and M4 and four lower (3500 m) sediment traps at sites M2, M3, M4 and M5 (Fig. 1, Table. 2). On board, the pH of the

supernatant solution was measured and an aliquot of supernatant liquid was analysed for nutrient concentration (SiO_4^{4-} , NH_4^+ , PO_4^{3-}) for shipboard quality control. When necessary, samples were post-poisoned and brought to an appropriate pH when lower than 8 to prevent CaCO_3 dissolution. Samples were kept dark and cool at 4°C until further processing.

5

Table. 2. Sediment trap details. Sampling was performed simultaneously and synchronously from October 19, 2012 until November 7, 2013 (384 days). (mbsl = meter below sea level, MAR = Mid-Atlantic Ridge)

Trap	Position	Bottom depth (m)	Trap depth (mbsl)	Distance to African coast (km)	Closest horizontal distance to seafloor at trap depth (km)
M1U	12°00'N, 23°00'W	5000	1150	700	610
M2U	13°81'N,	4790	1235	2300	2260
M2L	37°82'W		3490		520 (MAR)
M3L	12°39'N, 38°63'W	4640	3540	2400	500 (MAR)
M4U	12°06'N,	4670	1130	3500	640
M4L	49°19'W		3370		580 (MAR)
M5L	12°02'N, 57°04'W	4400	3520	4400	63

3 Methods

10 The following paragraphs describe the different methods used to analyse particle fluxes and the element and biological composition of the specific samples. Due to the different nature and amount of material, i.e. the dry dust from the on-land dust collector in Iwik and the wet sediment trap samples from the deep ocean, not all methods were applicable to all samples. While for the marine sediments the total, biological and residual mass fluxes were determined, as well as their element composition, the Iwik samples were analysed exclusively for their total mass
15 and horizontal dust flux at 290 cm as well as their element composition.

3.1 Particle Mass Fluxes

From the land-based dust collectors, all dust was removed from each sample bottles by loosening and shaking the dust out of the bottles. The removed dust was and weighed on a micro-balance ~~for every bottle height~~. Dust fluxes represent the horizontal transport fluxes of Saharan dust in the source. It is estimated by equation (1)

20

$$F_{MWAC} = \frac{MAR}{A} * \frac{1}{n} \quad (1)$$

where F is the dust flux ($\text{g m}^{-2} \text{d}^{-1}$), MAR is the mass accumulation rate (g d^{-1}), A is the cross-sectional area of the inlet tube of the MWAC sampler (m^2) and n is the estimated sampling efficiency of the MWAC bottles. The sampling efficiency of the MWAC samplers is between 75 % and 90-90 % for 30 μm dust (Goossens and Offer, 2000), which is within a similar size fraction of the Iwik dust (Friese et al. 2017).

25

For the -vertical (downward) marine particle fluxes from the sediment traps, the samples were wet-sieved over a 1 mm mesh, wet-split in five aliquot subsamples using a rotary splitter (WSD-10, McLane Laboratories), washed to remove the HgCl_2 and salts, and centrifuged. Afterwards samples were freeze-dried and ground. Total mass
30 fluxes refer to the < 1 mm size fraction and were determined by weighing two freeze dried 1/5 aliquots for every

sample. Average weight ~~difference~~ between replicate aliquots were within 2.4% (SD = 2.2) and less than 12%, with 87% of all samples differing < 5% between splits. Total mass fluxes were determined by equation (2)

$$F = MAR * A^{-1} * d^{-1} \quad (2)$$

where F is the total mass flux (mg m⁻² d⁻¹), MAR is the mass accumulation rate (mg), A is the sediment trap funnel opening (m²), and d the sampling time interval.

Total nitrogen (TN), total carbon (TC) and organic carbon (TOC) content were determined with a Thermo Scientific Flash 2000 Elemental Analyser. Samples for TOC measurements were decalcified by acid fuming with a subsequent addition of 2 N HCl and dried in an oven at 60⁰ C. Samples for TN and TC measurements remained untreated. Carbonates were calculated as CaCO₃ = (TC – TOC) * 8.33 and organic matter as OM = 2*~~€_{org}~~TOC. ~~The €~~conversion factors for ~~CaCO₃~~ CaCO₃ is based on its stoichiometry, given 100 mol/g of CaCO₃ for 12 mol/g of carbon, resulting in a factor of 100/12 = 8.33. The calculation factor for organic matter ~~varies~~ from 2.0 in the eastern Atlantic Ocean to 2.5 in upwelling areas (Fischer and Karakas, 2009;Jickells et al., 1998;Klaas and Archer, 2002;Thunell et al., 2007); ~~due to poorly constraints~~constrained composition of the actual organic matter molecule. We chose to use the factor of 2 for better comparison ~~to of~~ particle fluxes influenced by Saharan dust deposition off Cape Blanc (Fischer and Karakas, 2009;Fischer et al., 2007;Wefer and Fischer, 1993).

Biogenic silica (BSi) was analysed by sequential alkaline leaching on a HITACHI U-1100 spectrophotometer after Koning et al. (2002). ~~Briefly, A standard amount of~~ 25 – 30 mg of ground sample was ~~placed~~ leached in a 0.5 M NaOH solution at 85° C, to dissolve the biogenic silica, which subsequently reacted with a sulphuric acid—molybdate solution to form a blue molybdate complex. The complex was prevented from molybdate reduction and stabilized by adding oxalic and ascorbic acid, respectively. ~~The~~ is solution was flushed through a photocell where the absorption of the blue complex was measured at the defined 660 nm (Grasshoff et al. 1983) ~~and was~~ recorded every second. Each sample was run for 60 to 90 minutes. Results were evaluated with a weekly measured standard calibration curve (R² > 0.99) and calculated with the MS Excel data solver tool, extrapolating the dissolution curve to time zero to correct ~~ing~~ for contribution of non-biogenic silica (DeMaster, 1981). The diatom reference material (pure *Thalassiosira punctigera* from the North Sea) is measured with a reproducibility of ±0.46 % and sample reproducibility is ±0.36%. For estimating mass fluxes, BSi is expressed as BSiO₂ (BSi*2.139), although this conversion systematically underestimates the actual mass by 10-20 % given the crystal water associated with the opaline silica of which the siliceous plankton consists (Mortlock and Froelich, 1989).

The remaining, residual mass fraction is often referred to as the lithogenic fraction (Fischer and Wefer, 1996;Wefer and Fischer, 1993;Fischer et al., 2016;Fischer and Karakas, 2009;Neuer et al., 2002); since it contains all the refractory lithogenic particles (quartz, clay minerals, feldspars). The residual mass is defined by subtracting the biogenic CaCO₃, BSiO₂ and OM from the total mass (Eq. 3) and includes all Saharan dust:

$$\text{Residual mass} = \text{Total mass} - \text{CaCO}_3 - \text{BSiO}_2 - \text{OM} \quad (3)$$

However, this residual fraction potentially also includes biogenic phosphates and sulphates, as well as particles of volcanogenic, cosmogenic and anthropogenic origin. In addition, crystal water associated with the opaline silica and the O-H content of organic matter and clay minerals also contribute to the residual mass. Therefore, the residual mass fraction is most likely overestimated while the marine biogenic fraction is underestimated.

3.2 XRF element analysis

The elemental composition of each sediment trap sample was determined by X-ray fluorescence (XRF) using the Avaatech XRF core scanner (Richter et al., 2006). This analytical technique has the important advantage that it is non-destructive, allowing that very small-size samples – such as sediment trap samples – can be used for other analyses after measurement. XRF scanning results in semi-quantitative compositional data (Richter et al., 2006), being expressed as intensities (i.e. counts or counts per second), which we normalize to the total counts to take into account the closed sum of geochemical data. ~~Similar to XRF scanning of down-core sediment samples (Bloemsmma et al., 2012), we~~ We analyse our data ~~qualitatively~~ as normalized element intensities, ~~using the advantage that XRF-scan measurements on homogenised dry sediment trap samples largely avoid physical properties biasing, e.g. wet down-core XRF measurements (Tjallingii et al., 2007; Weltje and Tjallingii, 2008).~~ For dry-powder samples, Tjallingii et al. (2007) showed that element intensities are proportional to their chemical concentration, which we confirm by measuring 13 standards with various matrices, including marine sediments that have a similar matrix to sediment trap samples (Supplement S1). ~~which relate to the relative concentrations of these two elements (Weltje and Tjallingii, 2008).~~

Ground sediment trap samples (~25 mg) were pressed in polyethylene cylinders with a circular recess of 6 mm and 1.5 mm depth and covered with SPEXCerti Ultralene® foil. ~~This way we are largely avoiding common XRF-scan complications relating to measurement geometry (e.g. interstitial water content and large grain sizes) (Weltje and Tjallingii, 2008).~~ All samples were measured with a 4x4 mm slit size at a voltage of 10, 30 and 50 kV (elements: Al, Si, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Sr, Zr and Ba), with an electric current of 1.5, 1.3 and 0.8 mA, ~~respectively~~, and a measurement time of 20, 40 and 80 seconds, respectively. All measurements were performed five times, and ~~we show the~~ average values for these measurements are shown. The elements Ti, Al, Fe and K were chosen and shown, ~~since they are only present as main or trace-minor elements in lithogenic minerals like clays, quartz, and feldspars, rather than in any biogenic mineral formed in the ocean,~~ while Si represents both lithogenic Si minerals (clays, quartz, and feldspars) but also ~~and~~ biogenic produced silica (BSiO_2), as found in phytoplankton diatoms or zooplankton radiolarians. Pearson correlation was applied to the whole dataset of which we show the five elements Ti, Al, Fe, Si and K in table ~~S4~~S2. Five of the sediment trap samples (M1U #9, 12 and 24; M4U #12 and 24) could not be analysed due to insufficient material.

Two certified external standards (powdered marine sediment MESS-3 and PACS2) were used for quality control of the XRF measurements (Supplement S1), and were both analysed within each run of ~25 samples in exactly the same way as the sediment trap samples. These standards showed a precision, expressed as relative standard deviation ($\text{RSD} = \text{standard deviation} / \text{average of replicates} \times 100\%$), $< 7\%$ for all normalized element intensities. Moreover, the normalized intensities of these two standards closely follow the calibration line of all 13 standards (Supplement S1).

4 Results

Horizontal transport fluxes from the land-based MWAC sampler are given in $\text{g m}^{-2} \text{d}^{-1}$. The vertical (downward) deposition fluxes from seven sediment traps deployed across the Atlantic Ocean are treated in downwind succession from east to west, starting at ocean site M1 closest to Africa, to ocean site M5 closest to the Caribbean. For each trap, the relative contribution of the biogenic components CaCO_3 , BSiO_2 , OM, and the residual mass fraction are given, in addition to the total mass flux, the flux of each biogenic component and that of the residual mass fraction, expressed both in $\text{mg m}^{-2} \text{d}^{-1}$ and $\text{g m}^{-2} \text{a}^{-1}$. The biogenic components are produced by autotrophic phytoplankton and heterotrophic zooplankton. In particular, phytoplankton CaCO_3 producers are mainly coccolithophores while zooplankton CaCO_3 is mainly from foraminifera and gastropod shells. The BSiO_2 is

primarily produced by the phytoplankton diatoms and zooplankton radiolarians. Seasonal variations are shown for specific fluxes as deviation from their annual mean, together with the grain-size distribution of Saharan dust (Van der Does et al. 2016) for the residual mass fraction in the same material. To evaluate the residual mass fraction in the sediment traps, we compare XRF data from MWAC samplers with those from the sediment traps. The horizontal mass fluxes of the land-based dust collectors in Iwik are almost 100 % pure dust with negligible contribution of organic matter and fresh water diatoms from paleolakes and can therefore be used for chemical element comparison to the residual mass fraction from the sediment traps. Deviations between the chemical composition of the MWAC samplers and the sediment traps reveal compositional changes.

4.2.1 Horizontal land-based transport fluxes at Iwik

The total mass fluxes of the MWAC samplers at 290 cm height show significant month-to-month differences (Fig. 32). Highest dust fluxes of around 400-360 and 2350 g m⁻² d⁻¹ were found during spring in March and April 2013 and during summer in July and August 2013, respectively. Local wind speeds at 10 m above the displacement height (MERRA model, NASA GES DISC) show monthly average velocities between 3 and 7 m s⁻¹ with higher wind speeds in early summer and lower wind speeds in fall. Throughout the entire sampling period, the wind speeds are above desert pavement threshold velocities of 1.5 to 3 m s⁻¹ (Pye, 1987), suggesting also dust contribution from adjoining areas.

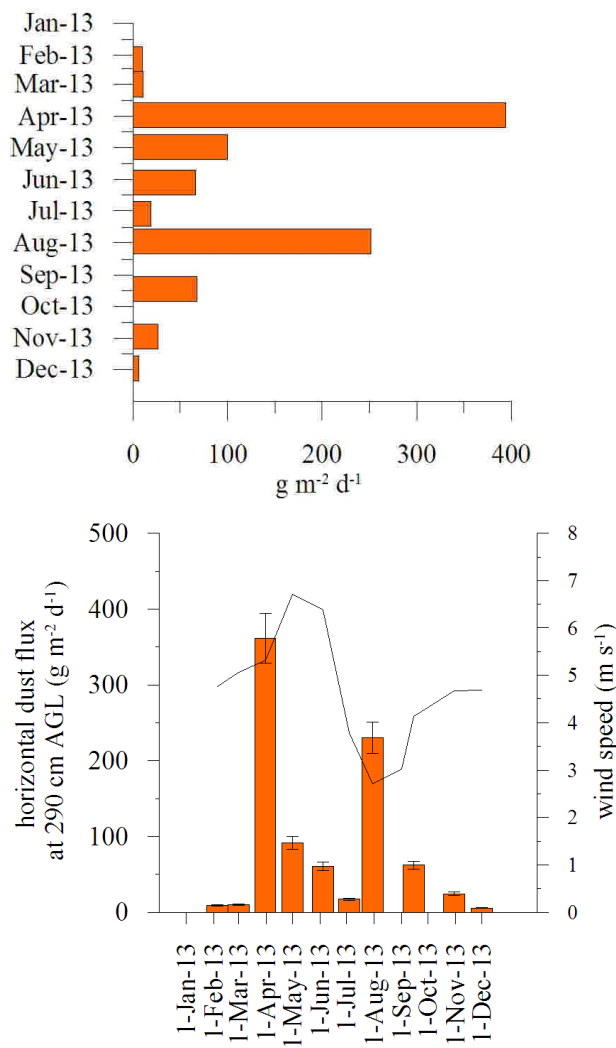


Figure. 32. Monthly resolved horizontal transport fluxes of Saharan dust from the land-based MWAC sampler in Iwik, Mauretania (19° N, 16° W) between January and December 2013 in orange bars. Error bars show the MWAC sampler efficiency of 75 to 90 %. Monthly averaged wind speed at 10 m above

displacement height (MERRA model) around the Iwik location is indicated as a black line (data obtained from the Giovanni online data system, NASA GES DISC). AGL = above ground level

4.2.1 Mass fluxes at ocean site M1

The shallow sediment trap at 1150 m water depth at site M1 collected a fairly steady total mass flux year round (Fig. 34) with a daily average of approximately $100 \text{ mg m}^{-2} \text{ d}^{-1}$ which equals an annual export of $40.7 \text{ g m}^{-2} \text{ a}^{-1}$ (Table 3). The mass fluxes of the marine biogenic constituents (CaCO_3 , BSiO_2 and OM) follow those of the total mass and are in close relation to each other. The composition of the trapped material is dominated by CaCO_3 (21-45 %) and the residual mass fraction (30-58 %), with minor OM (14 %) and BSiO_2 (10 %). A clear seasonal pattern in the total mass flux is absent, ~~whereas-although~~ a peak flux is recorded in March (interval #9). Relative to the annual mean, biogenic matter fluxes are high during winter and spring, and low during fall and especially summer (Fig. 45). Consequently, the residual mass fraction shows the opposite, with a low relative abundance in winter and spring and a high in summer and fall. The variation of the residual mass flux is consistent with the long-term seasonal transport of Saharan dust above the eastern tropical North Atlantic (Chiapello and Moulin, 2002). The residual mass flux ranges from $32\text{-}71 \text{ mg m}^{-2} \text{ d}^{-1}$ with an average of $47.7 \text{ mg m}^{-2} \text{ d}^{-1}$ ($17.4 \text{ g m}^{-2} \text{ a}^{-1}$) and slightly elevated values during summer and fall (#16, 18, 19 and 22) but with a peak in March (#9). A pronounced seasonality is also seen in the grain-size distributions of the same material with coarser grained Saharan dust in summer and finer grained dust in winter (~~van-Van~~ der Does et al. 2016).

4.2.2 Mass fluxes at ocean site M2

Total mass fluxes at site M2 average $40 \text{ mg m}^{-2} \text{ d}^{-1}$ ($14.4 \text{ g m}^{-2} \text{ a}^{-1}$) in the upper trap at 1235 m and $55 \text{ mg m}^{-2} \text{ d}^{-1}$ ($19.9 \text{ g m}^{-2} \text{ a}^{-1}$) in the lower trap at 3490 m (Fig. 34, Table 3). Peak fluxes are recorded ~~during spring and summer, especially~~ in March and late April (intervals #9 and 12). The composition of the material is rather similar in the upper and lower trap. In general, the trapped material is dominated by CaCO_3 (56 % and 59 %, respectively), followed by the residual fraction (27 % and 28 %, respectively), OM (11 % and 7 %, respectively) and BSiO_2 (both about 6 %). No clear seasonal pattern is seen in either traps in the total mass fluxes despite some peaks which occur in the upper and the lower trap during the same sampling intervals (Fig. 45). No clear seasonal pattern appears in the biogenic fractions, ~~whereby-although~~ CaCO_3 and BSiO_2 tend to be relatively enhanced during spring and summer, and OM in fall. Therefore, the resulting residual fraction is relatively enriched during summer and fall and suppressed during winter and spring. The small contribution of the residual fraction in spring is also expressed in its mass flux, yet is more variable in the upper trap than in the lower trap. It ranges between $4\text{-}32 \text{ mg m}^{-2} \text{ d}^{-1}$ with an average of $10.6 \text{ mg m}^{-2} \text{ d}^{-1}$ ($3.9 \text{ g m}^{-2} \text{ a}^{-1}$) in the upper trap and varies between $10\text{-}22 \text{ mg m}^{-2} \text{ d}^{-1}$ with an average of $15.3 \text{ mg m}^{-2} \text{ d}^{-1}$ ($5.6 \text{ g m}^{-2} \text{ a}^{-1}$) in the lower trap. This also holds for the grain-size distributions, while the dust particles in the upper trap show a distinct seasonality with coarser grains in summer and fall, the grain sizes of the dust particles in the lower trap fluctuate with no clear seasonality (~~V~~an der Does et al. 2016).

4.2.3 Mass fluxes at ocean site M3

At site M3 about 220 km south of site M2, the total mass flux at 3540 m water depth amounts to $70 \text{ mg m}^{-2} \text{ d}^{-1}$ ($14.4 \text{ g m}^{-2} \text{ a}^{-1}$) (Table 3). A double peak flux occurs during summer (intervals #15 & #16). The peak fluxes are mainly CaCO_3 , which forms more than half of the total mass (52 %) followed by OM (8 %) and BSiO_2 (7 %). Around one third (33 %) of the total mass resides in the residual fraction, with maxima during fall and minima during spring. ~~Although the S~~seasonality is weak ~~but several~~ small peaks appear, especially in June ~~but also during spring (March and late April) and during winter in January~~ (Fig. 34). The residual mass fraction varies between

13-38 mg m⁻² d⁻¹ with an average of 23.0 mg m⁻² d⁻¹ (8.4 g m⁻² a⁻¹). The differences from the mean contribution of the biogenic components show enhanced CaCO₃ in winter and spring and BSiO₂ in summer and fall 2013 (Fig. 45). The OM shows little seasonal variation, albeit slightly suppressed during spring.

4.2.45 Mass fluxes at ocean site M4

5 Average total mass flux amounts to 85 mg m⁻² d⁻¹ (30.9 g m⁻² a⁻¹) in the upper trap and 64 mg m⁻² d⁻¹ (23.2 g m⁻² a⁻¹) in the lower trap (Table 3). Both traps intercepted two striking peak flux events, the first during spring (late April) and the second during fall 2013 (late October/early November), when total mass fluxes exceeded 200 mg m⁻² d⁻¹ in the upper and 100 mg m⁻² d⁻¹ in the lower trap (Fig. 34). The peak fluxes differ in composition; while the peak in spring is CaCO₃ dominated (60 % in the upper trap and 73 % in the lower trap), the peak in fall is marked by BSiO₂ (25 % and 28 % in the upper and the lower trap, respectively). In addition, both peaks show elevated OM fluxes in both traps. Residual fluxes are elevated in the upper trap for the peak fluxes as well, but in the lower trap only in fall. Overall, the residual mass fraction ranges between 14-87 mg m⁻² d⁻¹ with an average of 24.0 mg m⁻² d⁻¹ (8.7 g m⁻² a⁻¹) in the upper trap and varies between 8-42 mg m⁻² d⁻¹ with an average of 17.4 mg m⁻² d⁻¹ (6.4 g m⁻² a⁻¹) in the lower trap. The seasonal variability of the grain-size distributions become smaller but are still visible in the upper trap with coarser grained dust during summer and fall (Van der Does et al. 2016). Seasonal variability of BSiO₂ is dominated by the exceptionally high peak in fall (Fig. 45).

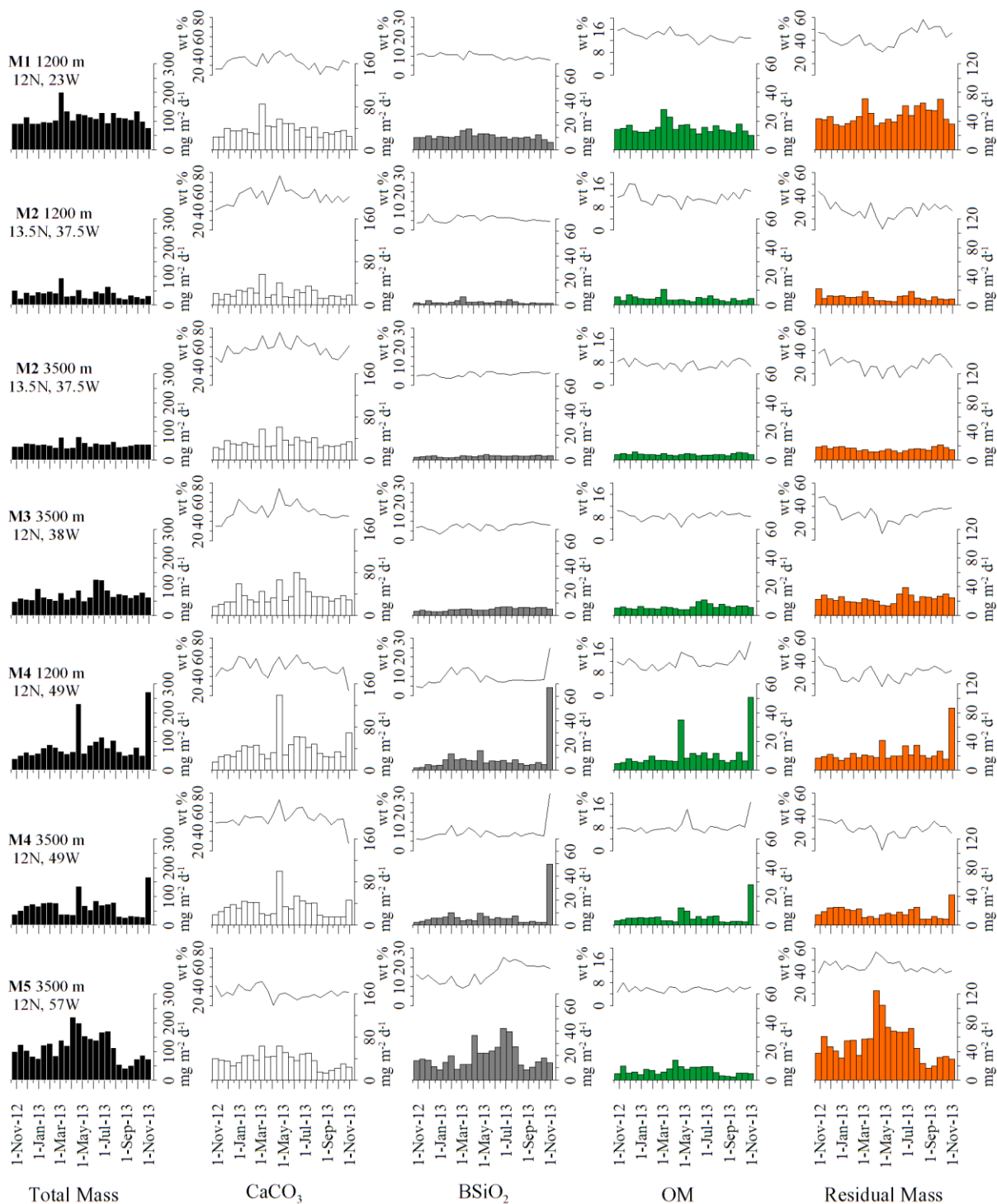
4.2.56 Mass fluxes at ocean site M5

The sediment trap at 3520 m water depth at the western most ocean site M5, intercepted an average total mass flux of 116 mg m⁻² d⁻¹, which equals an annual mass flux of 42.2 g m⁻² a⁻¹ (Fig. 34, Table 3). Most of the trapped material resides in the residual mass fraction (45 %), followed by CaCO₃ (33 %), BSiO₂ (17 %) and OM (6 %). Seasonal variability is clear in the total mass flux with maxima during spring and halfway through summer, followed by a changeover in August to lower values during the second half of summer and fall 2013 (Fig. 45). Clearest seasonality is shown by BSiO₂ with a pronounced maximum in summer to late fall and minima in winter and spring. Seasonality is less clear in CaCO₃ and absent in OM, while the residual mass shows maxima in spring and minima in summer to late fall. It ranges between 17-125 mg m⁻² d⁻¹ with an average of 52.3 mg m⁻² d⁻¹ (19.1 g m⁻² a⁻¹). The grain sizes of the residual fraction become smallest at this western most station and varies around the clay size fraction (van der Does et al. 2016).

Table 3. Yearly mass fluxes of measured flux parameters.

Trap	Total Mass Flux	CaCO ₃	BSiO ₂	OM	Residual mass	CaCO ₃	BSiO ₂	OM	Residual mass
	mg m ⁻² d ⁻¹ (g m ⁻² a ⁻¹)	%				mg m ⁻² d ⁻¹ (g m ⁻² a ⁻¹)			
M1 U	111.6 (40.7)	33.9	9.7	13.6	42.7	37.8 (13.8)	10.9 (4.0)	15.2 (5.6)	47.7 (17.40)
M2 U	39.5 (14.4)	56.0	5.9	11.3	26.8	22.2 (8.1)	2.3 (0.8)	4.5 (1.6)	10.6 (3.9)
M2 L	54.6 (19.9)	59.2	5.7	7.2	27.9	32.3 (11.8)	3.1 (1.1)	3.9 (1.4)	15.3 (5.6)
M3 L	70.4 (25.7)	52.0	6.9	8.4	32.7	36.6 (13.4)	4.9 (1.8)	5.9 (2.2)	23.0 (8.4)
M4 U	84.8 (30.9)	48.3	10.8	12.6	28.3	41.0 (14.7)	9.2 (3.4)	10.7 (3.9)	24.0 (8.7)
M4 L	63.6 (23.2)	53.0	10.7	8.9	27.4	33.7 (12.3)	6.8 (2.5)	5.7 (2.1)	17.4 (6.4)

M5 L	115.6 (42.2)	32.5	16.5	5.7	45.3	37.6 (13.7)	19.1 (6.7)	6.6 (2.4)	52.3 (19.1)
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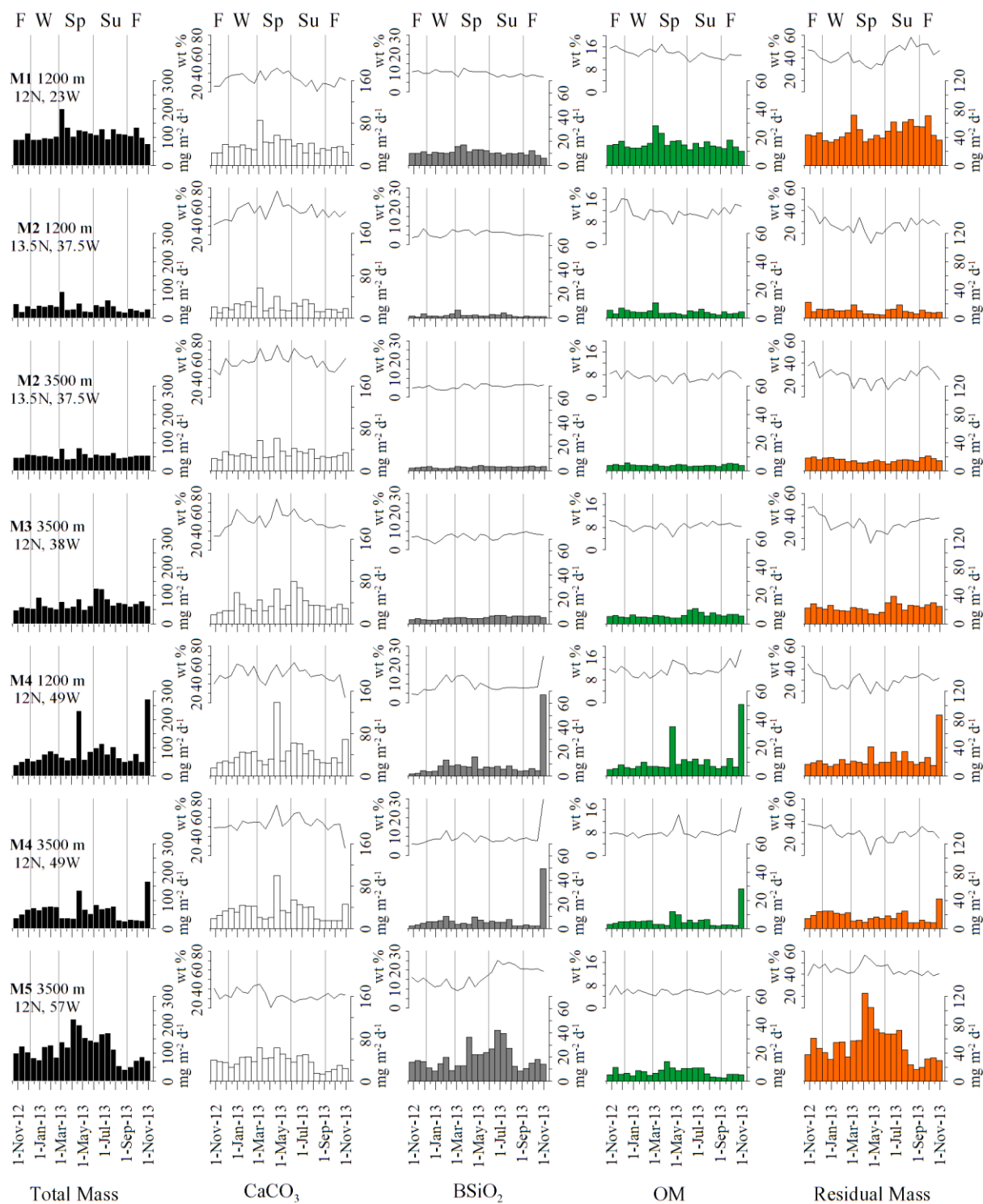


Figure 34. Mass fluxes at ocean site M1U to M5L in mg m⁻² d⁻¹ (bars) for total mass (black), CaCO₃ (white), BSiO₂ (grey), organic matter (green) and the residual mass fraction (orange); black lines represent their relative contributions (wt %). **F = Fall, W = Winter, Sp = Spring, and Su = Summer.**

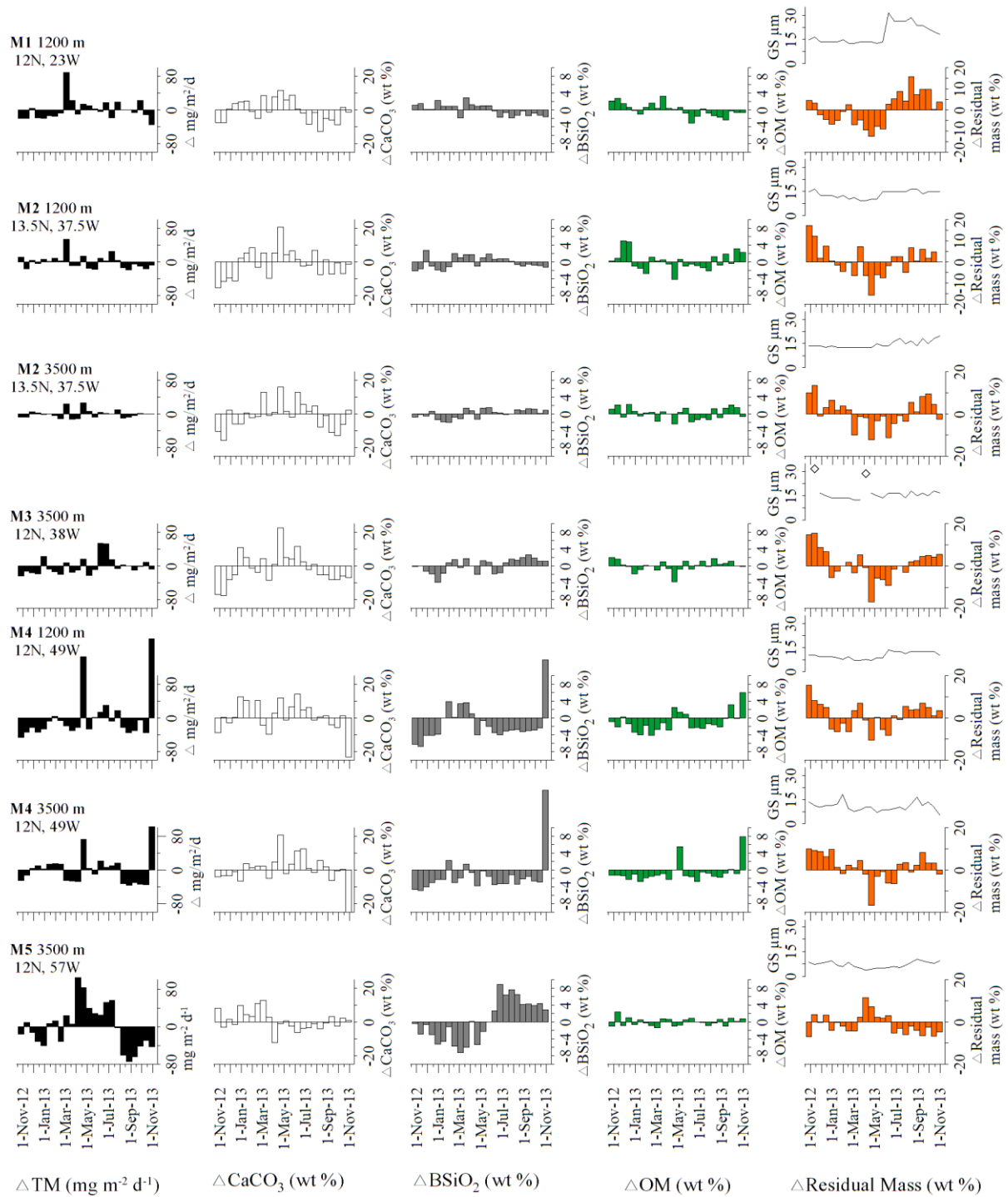


Fig. 5. Deviation from the flux-weighted annual mean for total mass (black), $CaCO_3$ (white), $BSiO_2$ (grey), organic-matter (green) and the residual mass fraction (orange) at ocean sites M1U to M5L. Black line represents modal grain size of Saharan dust of the same sample (data van der Does, et al. 2016)

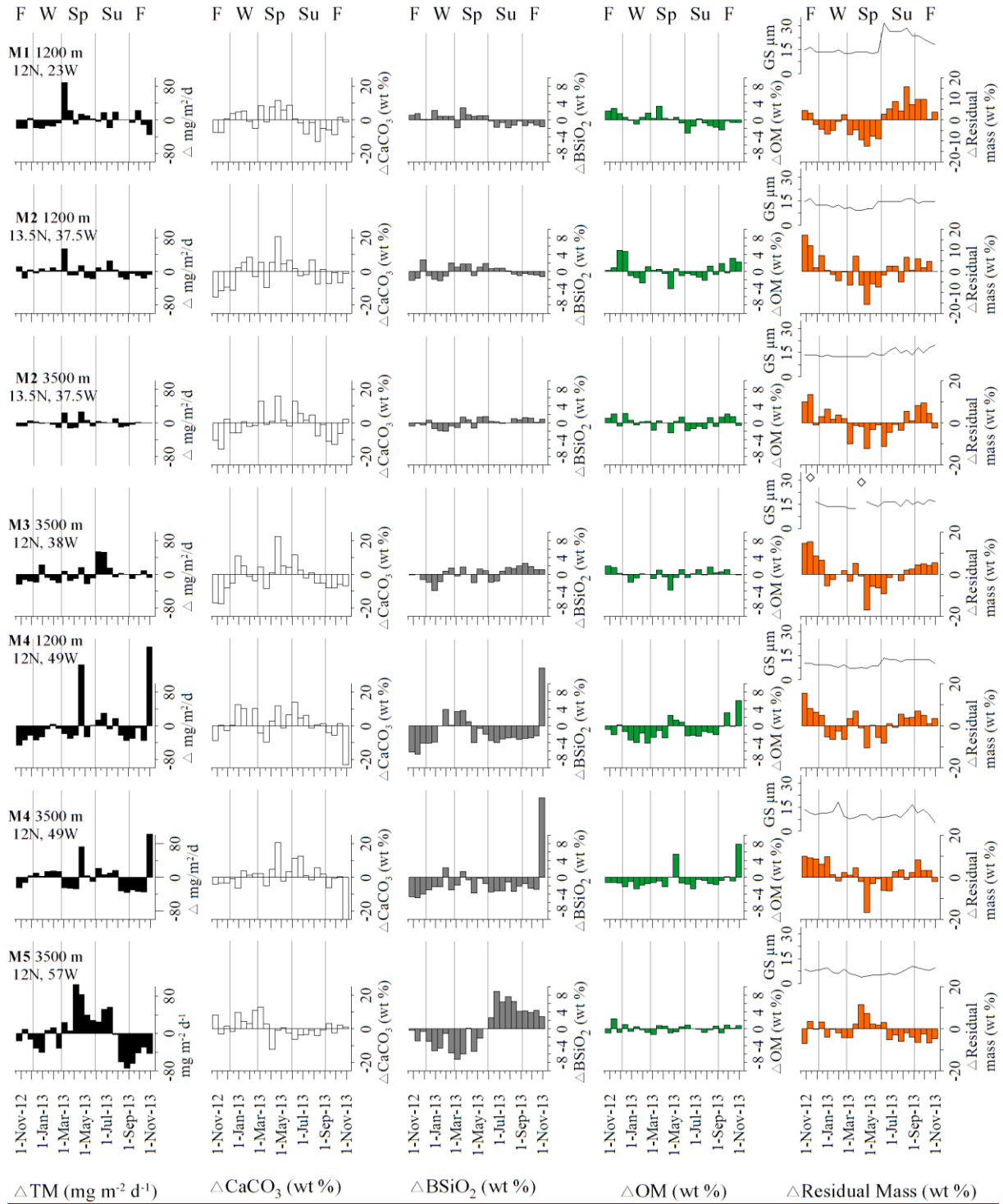


Figure 45. Deviation from the flux-weighted annual mean for total mass (black), CaCO_3 (white), BSiO_2 (grey), organic matter (green) and the residual mass fraction (orange) at ocean sites M1U to M5L. Black line represents modal grain sizes of Saharan dust of the same sample, diamonds in M3 are outliers (data ~~van~~ Van der Does et al. 2016). **F = Fall, W = Winter, Sp = Spring, and Su = Summer.**

4.2.67. Molar C : N ratios

Total organic carbon (TOC) and nitrogen (TN) are highly correlated at both trap depths (Fig. 56). The upper traps of the moorings M1, M2 and M4 show higher TOC and TN contents than the lower traps of the moorings M2, M3, M4 and M5. However, two fluxes (interval #13 and 24) at site M4L contain as much TOC and TN as found in the upper traps. The average molar C : N ratio of all traps is on average 9.17 with a standard deviation of 0.95. Overall, the ratios in the lower traps are slightly higher than in the upper traps, but without significant changes. The highest value (10.26) is reached at site M3L and the most uniform ratios between 8.46 and 9.65 are observed at ocean site M1U. The molar C : N ratios are in the typical range of sinking detritus collected in deep sediment traps and are comparable to the material collected off Mauritania but without seasonal differences (Fischer et al. 2016).

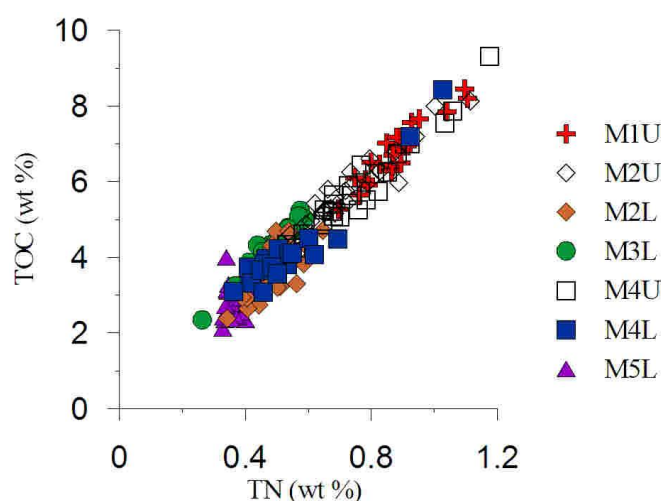


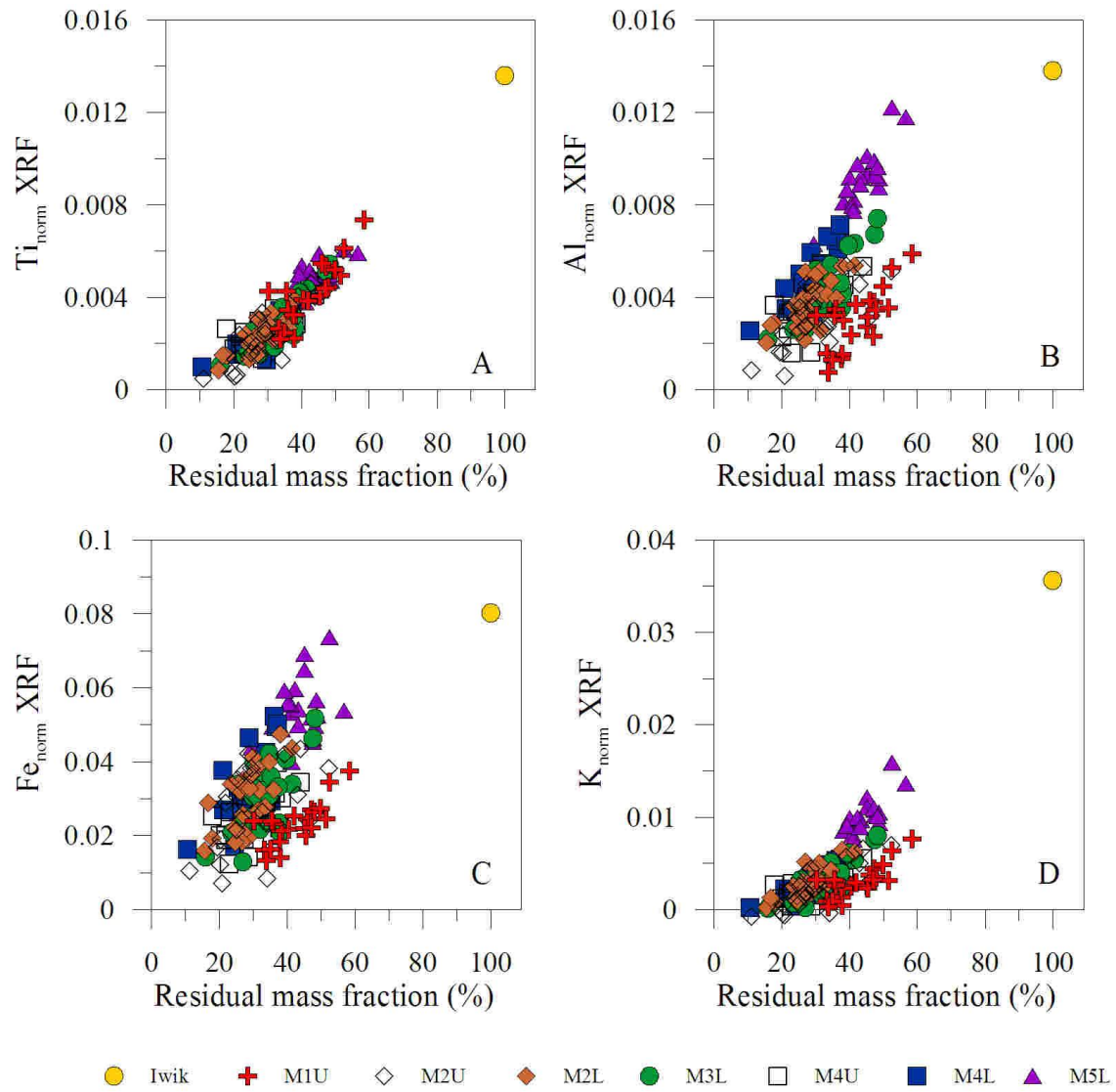
Fig. 56. Total organic carbon (TOC) versus total nitrogen (TN) content for all ocean sites M1-M5.

4.4.8 XRF element ratios

We identify the residual mass fraction as lithogenic fraction. Saharan dust by elements is characterized by exclusively lithogenic elements such as titanium (Ti), aluminium (Al), iron (Fe) and potassium (K), that were analysed by powder XRF. These elements are incorporated in mineral dust, in especially in aluminosilicates and feldspar, oxides and hydroxides and are incorporated in crystal lattice (Scheuven et al., 2013). Although K is also detected in biomass burning aerosols (Cachier et al., 1995), the high correlations between K and the other lithogenic elements show that the contribution of biomass burning K is minor in our samples. Normalized intensities of the lithogenic elements are highly correlated with the residual mass fraction and thus are therefore thought to represent the deposition flux of Saharan dust (Fig. 67). However, spatial differences from east to west indicate the continuous enrichment of especially Al and Fe from M1 to M5, and to an offset of higher K at M5, while Ti stays constant. The modal particle sizes of the dust (Van der Does et al. 2016) do not show a relation to the lithogenic elements. Best relations found were in the west at site M5 where sizes and lithogenic elements are negatively correlated (Al, $R^2=0.54$ and K, $R^2=0.44$).

Normalized intensities of Al and Ti are highly correlated as well, both within the time series in Iwik and throughout all seven sediment traps deployed in the deep ocean (Fig. 78; Table S4S2, S3S3). The pure-dust sample of from Iwik and the samples at the proximal ocean site M1 are most similar in element composition, having the same slope and intercept for Ti and Al normalized intensities (Table S2S3). Further downwind, Ti/Al intensity ratios

are lower but very similar at M2, M3 and M4U, while those at M4L and M5L have very similar but lower slopes (Fig. 78).



5 Fig. 67. Comparison of the lithogenic elements Ti (A), Al (B), Fe (C) and K (D) normalized to total XRF counts versus the residual mass fraction in all sediment traps and the average of the land-based dust collectors in Iwik.

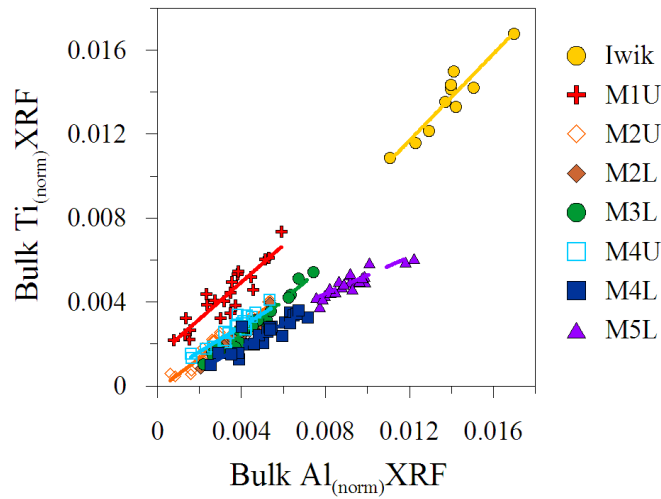


Fig 78. Comparison of bulk Ti and Al, normalised to total XRF counts for all seven sediment traps and the on-land dust collectors in Iwik, Mauritania.

- 5 Saharan dust mostly consists of lithogenic silicates such as quartz, clay minerals and feldspars (Caquineau et al., 2002) that contribute to the total silica content in addition to the biogenic silica found in the sediment traps derived from skeletal plankton such as diatoms and radiolarian. Regression of biogenic silica determined by sequential leaching to the total silica determined by powder XRF show a clear downwind increase from M1 to M5 (Fig. 89). Starting at ocean site M1, closest to the Saharan dust source, correlation is weakly negative, then vanishes at M2 and becomes positive and progressively stronger from M3 to M5.
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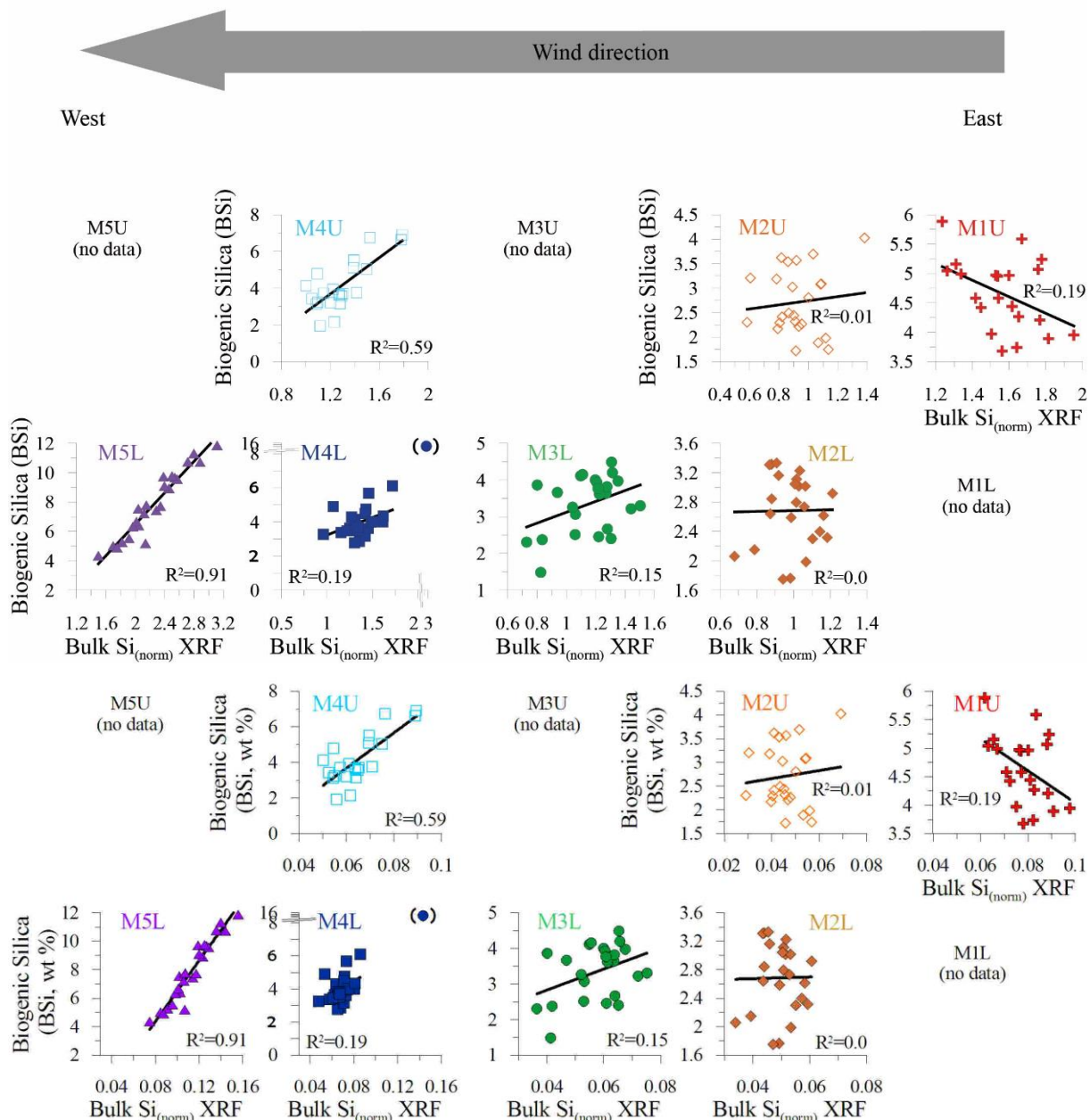


Fig 89. Biogenic silica versus total silica in particulate fluxes at five sites across the North Atlantic with sampling site M1 in the east (right) and M5 in the west (left).

5 Discussion

Within the DUSTTRAFFIC program we provide the first comprehensive time series of biogenic particle fluxes and Saharan dust deposition from source to sink across the equatorial North Atlantic Ocean. The land-based dust collector in Iwik areis located in the coastal region of western Mauritania, in the potential source area 2 (PSA 2), which is one of the major source areas of dust that is transported across the Atlantic Ocean to the Americas (Scheuven et al., 2013). The transport fluxes are highest in spring and summer (Fig. 23). The winter-spring high can be related to the trade-wind intensities at lower altitudes, whereas the summer high point to sporadic dust storms, invisible on land by satellites. Backward trajectories reveal that the location in Iwik is a transit area for long-range transported Saharan dust (Friese et al. 2017). Overall, However, the horizontal Saharan dust flux from the land-based MWAC samplers in Iwik cannot be compared directly to the downward flux in residual mass flux into in the sediment traps. However, they are are still not only compositionally the same, especially as in the

sediment traps at the ocean sites M1, ~~which is indicated as indicated by in terms of~~ their similar Ti-Al slopes (Fig. 78), ~~but also testify to a downwind decrease in flux with distance from the source of North African dust. From site M1 This trend continues downwind as~~ we observe an overall decrease in both the residual mass and the marine biogenic matter fluxes ~~from ocean sites M1 westward to M2/M3 and M4~~. The much higher ~~content counts~~ of lithogenic elements Ti, Al, Fe and K found in the ~~pure~~ dust at Iwik result from the absence of dilution ~~by~~ biogenic matter produced in the marine realm. While the Ti-Al slopes are the same at Iwik and ocean sites M1 through M4U, and Al, Fe and K ~~contents counts~~ are most similar at Iwik and M1 as well, but become significantly higher towards the west (Fig. 67), it suggests a downwind change in mineralogical composition of Saharan dust. At ocean sites M4L and M5L, both the Ti/Al slope is lower and the Al ~~content counts are higher especially at M5 is the same~~, indicating that a second source in addition to Saharan dust is involved, which may be derived from admixture of re-suspended clay-sized sediments advected towards the deep sediment traps. This is especially indicated by the offset of K at M5L. Overall, we observe a downwind increase in the Al and Fe content of the residual mass fraction, which suggests an increase in clay minerals relative to quartz. A similar change in mineralogy was observed by Glaccum and Prospero (1980) who compared the mineralogy at the eastern and western Atlantic Islands. Our observation is also supported by the clear downwind trend in the correlation between biogenic silica, as determined by sequential alkaline leaching, and total silica measured by XRF (Fig. 89). Such a downwind decrease in the relative amount of quartz may result from the increase of finer particles due to their slower settling speed, and a relative increase in platy clay minerals such as micas suspended in the atmosphere that can be transported over greater distances. This is consistent with the downwind decrease in grain size as observed by Van der Does et al (2016) on the same material.

5.1 Comparison with satellite observations

Saharan dust needs about a week (5 – 6 days) to cross the Atlantic Ocean from east to west as observed in satellite images (Prospero et al. 1970, 2014). Since the sampling interval of all sediment traps is 16 days, it is difficult to identify individual dust outbreaks as individual peaks in mass fluxes that decrease in amplitude from east to west. However, this seems to be the case for the peak fluxes occurring in summer, when there is an enhanced contribution by the residual fraction, especially closest to the African coast at the ocean site M1. Such enhanced fluxes in residual mass are found in intervals #16, ~~and~~ #18 and #19 at site M1U, and M4U and #16 of M3L, corresponding to the period from 16 June to 2 July, ~~and~~ from 18 July to 3 August ~~2013, and 3 August to 19 August 2013,~~ respectively. During these summer intervals, dust outbreaks leave the African coast at high altitudes and propagates westwards across the Atlantic (Fig. 94). Despite this apparently good correspondence between the satellite data and the sediment trap record, there are also other cases where the sediment traps did not seem to record specific dust outbreaks observed in satellite images. This may relate to the altitude at which the dust is travelling; in summer, dust travels through the high atmosphere, as opposed to winter-dust that travels in the low-level Trade-wind zone. The diffuse cloud that is observed across a relatively wide band throughout the atmosphere in summer is interpreted from nadir-looking satellites as higher aerosol optical thickness than the relatively narrow and dense dust cloud that crosses the atmosphere in winter. Conversely, some sediment trap intervals recorded a residual mass peak in spring, e. g. interval #9 which could be followed in M1U, M2U, and M3L, (Fig. 34), when no evidence is found for the occurrence of dust events during that period (24 February – 12 March 2013) in satellite imagery. This could be reflecting the timing of satellite overpasses, which could potentially lead to missing short-term events. However, most dust events that are recorded by satellite images are visible as multiple-day events. The presence of clouds in virtually all satellite images is likely to at least partially obscure low-level winter dust

outbreaks. Although satellite products are a great help to follow huge dust outbreaks crossing the Atlantic, our results illustrate that there is still a high degree of difficulty in matching specific dust outbreaks observed by satellites and the fluxes we measure in the deep sediment traps. This may also be enhanced by time lag between the deposition of Saharan dust on the ocean surface and their arrival at depth.

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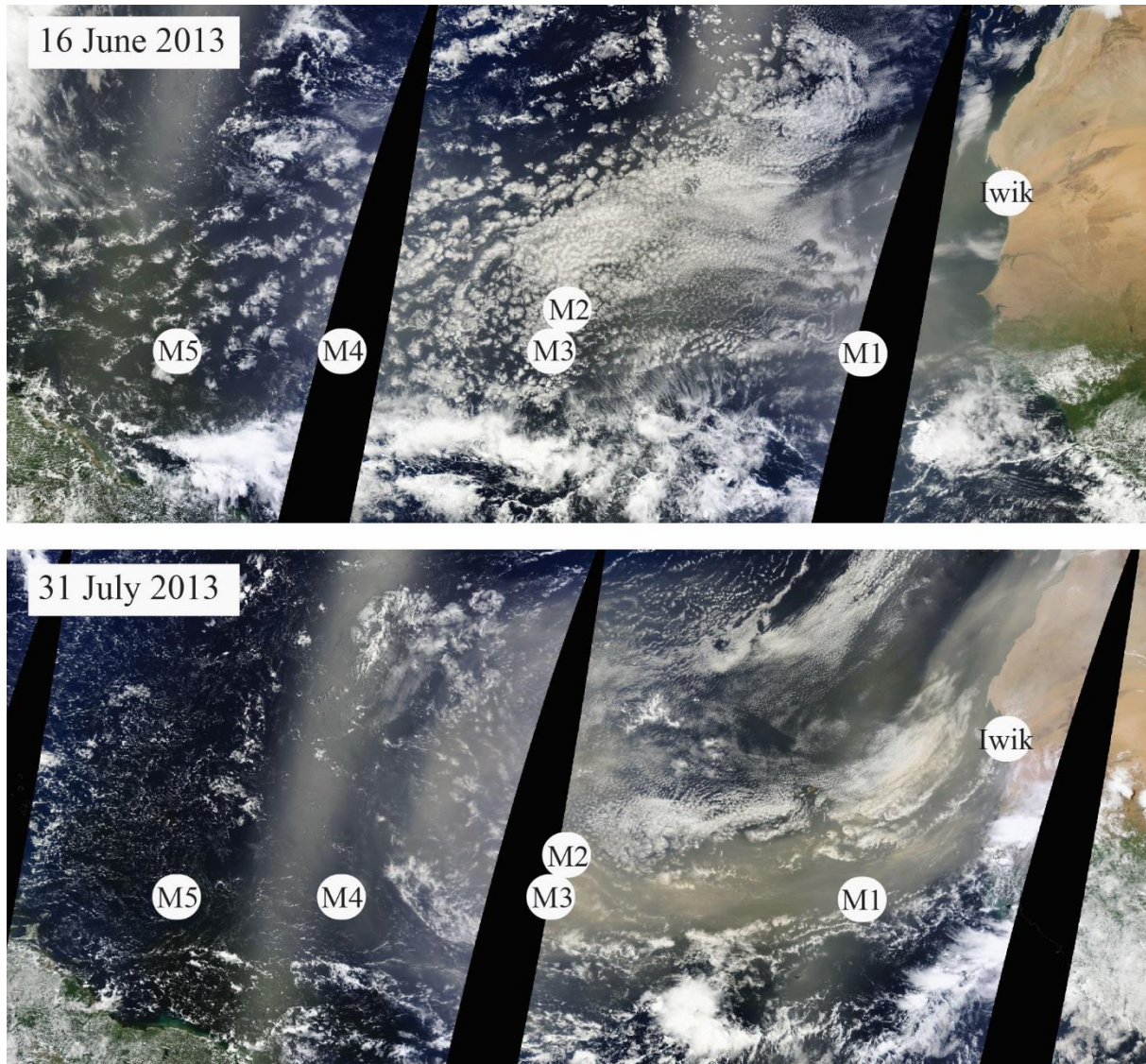


Fig 910. Satellite images (~~EOSDIS Worldview~~; <https://worldview.earthdata.nasa.gov> Images from NASA Worldview, MODIS Terra satellite) showing dust outbreaks propagating westwards from the African coast across the Atlantic Ocean, the location of the land-based dust samplers at Iwik and the five ocean moorings M1 through M5.

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5.2 Gravitational settling and downward transport

Saharan dust deposited on the surface ocean is generally too fine to settle out as individual particles, except perhaps for larger and massive quartz particles. Rather, they remain effectively suspended until incorporated by organic matter aggregates such as “marine snow” and fecal pellets that accelerate settling to velocities in the order of 200 m d⁻¹ (Knappertsbusch and Brummer, 1995; Berelson, 2002; Fischer and Karakas, 2009). Indeed, at site M4, both the traps at 1200 m and 3500 m intercepted two peak fluxes during the same 16-day interval, one in April and one in October (Fig. 34). Since these traps are 2300 m apart, this results in a settling velocity of at least 140 m d⁻¹. Higher settling velocities are conceivable for the fall peak fluxes at station M4, since the TOC and TN content (Fig. 56) of this peak flux in the lower trap is equally high as in all the upper traps (M1, M2 and M4), indicating

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rapid settling and little degradation. The same is observed at ocean site M2 for the flux peaks in March and in May (Fig. 34). However, the settling pathway might be different, as indicated by the higher total mass fluxes in the lower trap than the upper trap at site M2. Over the 2300 m distance of the upper and lower trap, the sinking particles are potentially subject to a variety of processes including remineralisation, disaggregation, or repacking, as well as horizontal movement of the particles, resulting in a greater catchment area (Waniek et al. 2000). Therefore, the enrichment in the lower trap at site M2. ~~Such enrichment~~ might result from the greater catchment area of the deeper trap (Siegel and Deuser, 1997; Waniek et al., 2000). The same holds for the residual mass, which is more enriched in the lower trap compared to the upper trap. Consequently, time lags between Saharan dust outbreaks and transport in the higher atmosphere as observed in satellite images and their arrival in the upper sediment traps are at least a week and might take another path until arrival in the lower trap two weeks later. In this respect, ~~the~~ buoys deployed now for time-series sampling of atmospheric dust just above the ocean surface at the same sites will serve as an intermediate between satellite observations of dust outbreaks and the actual deposition of dust on the surface ocean, which can be then followed down by the fluxes in the ocean.

5.3 Comparison with deep ocean mass fluxes off northwest Africa

The mass fluxes that we observe in the traps along the ~~DUSTTRAFFIC~~ transect are ~~much~~ in line with observations from sediment traps further north, off Cape Blanc on the Mauritanian continental slope at ~21°N, 20°W. For that site, Fischer et al., (2016) report total mass fluxes of 40.23 g m⁻² per year averaged over a period of 25 years with much interannual but little seasonal variability. This compares well with our sampling site M1, where we measured a total mass flux of 40.7 g m⁻² a⁻¹. Cape Blanc residual mass fluxes vary about 5 to 26 g m⁻² a⁻¹ (Fischer et al., 2016), which agrees very well with the average residual flux at site M1 of 17.4 g m⁻² a⁻¹. At both sites the residual mass fluxes make up about a third of the total mass flux and up to 50% during dust events (Nowald et al., 2015). Biogenic mass fluxes in the Cape Blanc area are generally also high, also owing to the upwelling of cold, nutrient-rich waters that cause high primary productivity in the surface waters. However, biogenic mass fluxes are about the same as found at M1, where no upwelling-stimulated productivity occurs. At site M1 all biogenic particle fluxes are highest in comparison to the other sampling sites. The only exception is the BSiO₂ at site M5, which is by far the highest contribution of BSiO₂. The low flux of biogenic particles in the mid-Atlantic Ocean (M2 to M4) reflect the limited availability of nutrients and low productivity in the oligotrophic ocean. The higher biogenic mass fluxes closest to either continent (M1 & M5) may have been enhanced by the higher lithogenic input, especially from higher Saharan dust input at site M1, as mineral dust enhances the settling of organic matter through the water column (Ittekkot et al., 1992; Hamm, 2002). ~~suggesting Saharan dust may be the common factor fuelling enhanced (export) production.~~

Moreover, M mass fluxes collected by sediment traps off Cape Blanc, for example, are affected by lateral input from re-suspended sediments that are advected from the Mauritanian continental slope in nepheloid or bottom layers (Fischer et al., 2009). Similar lateral transport of resuspended sediments (Van Raaphorst et al., 2001; Bonnin et al., 2006) may also have come from the nearby Barbados continental slope and contributed to the high fluxes of the residual mass fraction at ocean site M5 (Fig. 34), which is relatively close to the Barbados margin, around 63 km to the trap depth at 3500 m (Fig. 1B, Table 2). This admixture of resuspended sediments is also suggested by the lower Ti/Al ratios and high K contents at M5, and to a far lesser extent also in the deep trap at M4 although residual mass fluxes at this site are significantly lower (Fig. 34). The enrichment in Al, Fe and K indicates the addition of Al-Fe-K-rich clay minerals from a second source next to the Saharan dust deposited at these sites. This second source of sediments in the deep traps potentially also causes the negative correlation between the residual

masses at M5 and M1, while those between M1 to M4 are all positively correlated (Table S3S4). ~~However~~Nevertheless, the particle-size distributions at M4 and M5 (Van der Does et al. 2016) ~~alone,~~ do fit the general pattern of decreasing particle size with increasing distance from the Saharan dust source, ~~thus indicating that the sediments collected by all stations are dominated by Saharan dust not expecting fluvial input from the west.~~

The particle fluxes at ocean site M5 deviate from the general pattern in terms of enhanced total and residual mass fluxes and seasonal biogenic-silica contribution. Due to its westernmost position, the deep sediment trap may have received considerable amounts of lithogenic or biogenic material originating from either the Amazon- or Orinoco River, or both. The freshwater Amazon outflow disperses kilometres from the river mouth and affects the oceans' biogeochemistry (Yeung et al., 2012). Due to nutrient input, the occurrence of diatom-diazotroph associations is stimulated (Subramaniam et al., 2008). The fact that the biogenic silica correlates perfectly with the bulk silica (Fig. 89) possibly relates to the appearance of diatom phytoplankton and less contribution of quartz minerals.

6 Conclusions

The first-year results of our monitoring experiment yield valuable insights into the transport and deposition fluxes of Saharan dust between the African continent and the Americas. We demonstrate that the lithogenic particles collected in the sediment traps are ~~from the same sources similar to the~~ as dust collected on the African coast. With increasing distance from the source, lithogenic elements associated with clay minerals become more important relative to quartz settling out closer to the source. The total silica analyses show that the contribution by biogenic silica, produced by marine biota, increases significantly from east to west to the extent that virtually all silica consists of biogenic silica and lithogenic quartz is ~~insignificant~~minor in the west furthest away from the African source(s). At the westernmost ocean site, enhanced residual mass fluxes collected by the deep sediment trap suggest admixing with a second source from resuspended ocean floor sediments, ~~although the fining in lithogenic particle size with increasing distance from the source argues against this.~~ Tracing back individual dust outbreaks from satellite images to the arrival in the deep ocean sediment traps is still demanding given sampling resolution and the time lags involved. Best accordance ~~of satellite images and the residual mass fraction~~ was found ~~in for~~ summer, ~~when higher~~ ~~The relative contribution of the residual mass fraction fluxes are~~ is found ~~to be that are~~ highly co-variant with typically lithogenic elements such as Al and Ti from Saharan mineral dust. While the temporal and spatial variability in residual mass fluxes ~~does correspond~~s ~~with to~~ the changing ~~fluxes-chemical composition~~ of Saharan mineral dust, they seem to overestimate the net fluxes due to underestimation of marine biogenic matter. To better approximate the deposition of Saharan dust, ~~the~~ buoys that were deployed in 2013 should give information on precise fluxes, which can then be followed in the underlying sediment traps.

7 Data availability

Supplementary data are available at <https://doi.pangaea.de/10.1594/PANGAEA.872093>.

Acknowledgements

The Project is funded by ERC (project no. 311152) and NWO (project no. 822.01.008). We thank the crew of Meteor Cruise M89, Pelagia Cruise 64PE378 and NIOZ technicians for their contributions. Camara from the Parc National de Banc d'Arguin (PNBA) is thanked for assistance with the dust sampling in Iwik, Mauritania. Jort Ossebaar is thanked for helping with the EA analysis and Sharyn Ossebaar for assisting with the biogenic silica

measurements. XRF-analysis was supported through the SCAN2 program (NWO project no. 834.11.003) and assistance by Rineke Gieles. Furu Mienis is thanked for helping with interpreting the current-meter data and assistance in the sediment-trap lab. The authors also acknowledge the MODIS mission scientists and associated NASA personnel for the production of the data used in this research effort. We thank three anonymous reviewers for their helpful contribution.

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