

Dear Dr. Schwarz,

Thank you for the time you spent assessing the manuscript. We would also like to thank the three anonymous reviewers for their comments. We considered every comment carefully and made changes to the revised manuscript accordingly. For reviewer 1, the changes were made in red, for reviewer 2 in green, and for reviewer 3 in blue.

We have found similar comments on the manuscript from different reviewers, and we are of the understanding that this is mainly due to the different “languages” researchers from different disciplines speak. One of the reviewers’ main concerns deals with the representation of the lithogenic particles found in the sediment traps as aeolian dust. As can be seen in the answers to the comments of the reviewers below, there are many reasons for this assumption. First, the sediment traps are located far away from any other possible source, e.g. riverine sediments, resuspension from shelf or bottom sediments, etc. Second, we compared grain size of the lithogenic particles collected by the sediment traps to dust collected from the atmosphere, directly above the sediment traps, which showed to be almost identical. Third, when comparing the upper (1200 m) and lower (3500 m) sediment traps from the same station, there are many indications of the nearly vertical settling of particles, indicating their atmospheric origin. And finally, when considering the large amounts of Saharan dust that are transported across the Atlantic Ocean every year, about 182 Tg (Yu et al., 2015), it will be fair to assume this will be a large contribution to the particle flux in the sediment traps.

Another question raised by the reviewers concerns the particle size of the dust we find in the traps, and if this is affected by some sort of processing or aggregation of particles. Aggregation of particles during or after deposition is possible, however all aggregates are destroyed by the three-step pretreatment processes to remove organic material in the sediments prior to grain-size analysis, including aggregates of dust particles that already existed during transportation in the atmosphere. Grain-size analysis is performed on the lithogenic fraction of the samples only. Therefore, the particle-size distributions of the dust measured in this study is at the very least an underestimation of the size of dust particles and aggregates as transported across the Atlantic Ocean.

With this paper we would like to bring the different disciplines together, which is also why we have chosen this journal for publishing. We greatly value the reviewers’ comments and hope we have answered them adequately. Please find attached the revised manuscript, with tracked-changes with respect to the original manuscript.

Anonymous Referee #1 (Referee comments in black, our reply and changes in the manuscript in red)

There is a lot of potential in this paper, but as written there are some serious issues with relating the data presented here with the conclusions. Even for big picture ideas (size gets smaller as you go across the Atlantic), it’s not clear to me how to interpret particles from sediment traps, so I think this needs to be discussed much more explicitly in the paper. The data itself, with the size changes across the ocean, should be publishable, but it’s the interpretation that is really an issue in this paper.

The big issue is: what is the aerosol in the trap and how is it related to what comes in at the top? Previous studies have shown that there are at least seasonally modulated relationships between the two (Bory et al., 2002), but here the authors are trying to interpret these seasonal changes as occurring in the atmosphere, which could be true. But the fact that the aerosol size is systematically different at the deeper cores suggests there is something else going, and the assertion that this must be from changes in the dust sources (agriculture!) is too speculative to be convincing. I am not even sure I believe the sizes they are getting represent aerosols, and definitely the time and space lag issues related to aerosol transport and processing in the oceans is too important, and almost completely neglected here, will heavily modify the signal they are trying to interpret!

1. size: “This resulted in particle-size distributions consisting of 92 logarithmic size classes ranging from 0.375 to 2000 μm . Grain-size statistics were calculated geometrically using the graphical method of Folk and Ward (1957) using GRADISTAT (Blott and Pye, 2001).” It sounds like you are assuming that the size of the particles you are measuring in the sediment trap is the size of the particles in the atmosphere? There is a lot that goes

into that kind of set of assumptions, so please spend at least a paragraph in the methods describing why you think this will work, previous papers which showed a relationship (or not) and what kind of assumptions it requires. Wetting an atmospheric aerosol during deposition, could either make particles coagulate or break the bonds of particles. On the other hand, material can coagulate onto the particles in the ocean, and change or process them. Already your evidence that the sizes are different at the trap and deeper down suggests changes in size or processing (or advection: see next point). Please be explicit about the assumptions you are making and justify them here in the results, and then in summary and conclusions discuss the implications of your assumptions for your work. Are these aerodynamic or geometric measurements, as a basis? Size of aerosols is tricky to measure (e.g. Reid et al., 2003) and different measurement methods get quite different results: how do your methods compare? Is there any way you can use previous measurements of size in aerosols (e.g. Reid et al., 2003; Skonieczny et al. (2013)) to help you with this problem? How do you know that these particles are from eolian deposition and not some other process?

First, the data described here are measurements of actual dust particles, and not a proxy for these dust particles. The cited paper by Bory et al. (2002) uses a proxy (Al) for lithogenic particles, but in our study we isolated the lithogenic fraction using chemicals, and performed grain-size analyses on this fraction only. As an example, Stuut et al. (2005) demonstrated the similarity between aerosol samples of sediment traps and dust found in sediment traps and in seafloor sediments. In this paper, we discuss data of deposited dust, and not of transported dust. The transportation of dust is, however, used for interpretation of the data. Seasonal changes observed in the traps are not only due to changes occurring in the atmosphere, but also in the sources of the dust: summer months are characterized by more convection due to bigger temperature differences, resulting in the uplifting of coarser particles. Also the different dust-transporting winds during the different seasons (the trade winds and Saharan Air Layer – SAL) that blow in different directions, at different altitudes and with different wind speeds cause the seasonal differences in particle size of transported dust.

We added the following lines to the revised manuscript:

Page 16, lines 3-5:

“In addition, increased convection in the source areas in summer, related to larger differences in temperature, can result in the uplift of coarser dust particles (Heinold et al., 2013).”

The assumption is made that the lithogenic particles we find in the sediment traps are of aeolian origin, more specifically dust originating from Africa. The sediment traps are far from the continental shelf, so riverine input of sediments can be excluded. Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll A or salinity, available from Giovanni of NASA GES DISC (<http://giovanni.sci.gsfc.nasa.gov/giovanni/>). Also, the lower sediment traps are positioned 880-1300 m above the seafloor (M2-M5), so resuspension of bottom sediments will not affect the sediment trap samples. When considering the large amounts of Saharan dust being transported every year, about 182 million tons (Yu et al., 2015), it is fair to consider the lithogenic fraction in the sediment traps to be of aeolian origin.

We added the following lines to the revised manuscript:

Page 5, lines 9-17:

“In this paper we argue that the lithogenic particles found in the sediment traps are of aeolian origin. The sediment traps are located far from the continental shelf, so riverine input of sediments is not affecting the samples. Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll or salinity (not shown; available from Giovanni NASA GES DISC: <http://giovanni.sci.gsfc.nasa.gov/giovanni/>). In addition, the lower sediment traps are positioned 880-1300 m above the seafloor, so resuspension of bottom sediments will not affect the sediment trap samples. When considering the large amounts of Saharan dust being transported across the Atlantic Ocean every year, about 182 Tg (Yu et al., 2015), any other external input is

assumed to be negligible. Stuu et al. (2005) also demonstrated the similarity between aerosol samples of Saharan dust collected off west Africa and the lithogenic fraction in sediment traps and seafloor sediments.”

During the grain-size analysis, individual particles are analyzed. First, the lithogenic fraction of the sediment-trap samples (or sea-floor sediments) is isolated, and the aggregates destroyed by the three-step pretreatment procedure of boiling with chemicals to remove all organic-produced components. So any post-transport or post-depositional aggregation is also removed. It is possible that dust particles are transported as part of aggregates in the atmosphere, and also these aggregates are destroyed during the sample preparations, resulting in the measurement of smaller particles. Additional processing of the particles in the ocean would not result in larger particles, only possibly in smaller particles. But since the processing before analysis is identical for every sample, the results can be directly compared to each other. In total, the particle-size distribution that is measured is at the very least an underestimation of the actual size of ‘particles’ (including aggregates) that are being transported. Grain-size analysis of 1-year accumulative samples from floating dust-collectors at our transect (unpublished results) have shown to be almost identical to the underlying sediment-trap samples, confirming again that the dust we find in the sediment traps is representative of the atmospheric dust.

Difference in particle size between the upper and lower traps could be the result of advection, but this is thought to be minimal. More likely is the larger catchment area for the deeper traps, resulting in the sampling of slightly more particles and particles of different sizes. A new figure added to the revised manuscript (Fig. 2) illustrates the similarity between the upper and lower traps at station M4, see also the next comment.

The measurements described here are geometric measurements, and were performed with a Coulter laser diffraction particle sizer, as described in the Material and Methods section of the manuscript. These measurements are the same as Skonieczny et al. (2013) performed, using a Malvern Mastersizer 2000 laser diffractometer. So these data can be compared almost directly (Konert and Vandenberghe, 1997). The geometric method may pose a potential problem for ‘lightweight’ mica particles: due to their platy shape they have a smaller aerodynamic size than geometric size, and as described in the manuscript [in Introduction and Discussion] as well as in Stuu et al. (2005), this is expressed as a secondary mode in the grain-size distribution. Therefore, it was decided to describe the grain-size data with the modal value of the distribution, which accounts less for these coarser particles with smaller aerodynamic size.

2. advection and sedimentation rates: The second issue is the relationship of what enters the top of the ocean and what is deposited in the sediment traps. Previous studies (Bory et al., 2002) have suggested that productivity could modulate the transfer rate between the top of the ocean, and the cores. We know that the dust has to be carried with the current as it floats downward: how far downward? (Han et al., 2008; Siegel and Deuser, 1997) show that it really can be quite far. If it also seasonally being modulated, that would really mess up your signal!

Since we have recovered both sediment traps, the upper (1200 m) and lower (3500 m), at two of the stations (M2 and M4), we can compare the data between these two traps. Although sediment fluxes are not the scope of this paper, a new Figure has been added to the revised manuscript (Fig. 2), showing photos of the sediment-trap bottles after recovery, with high levels of sediments in two samples (sample 12 and 24, collected during spring and fall, respectively). These high fluxes are present in both the upper and the lower traps, in the same sampling cup. This demonstrates the similarity in sediments received for both sediment traps, and that lateral advection is minimal. Since the sampling interval is only 16 days, it means that the downward transport velocity of these sediments is at least 140 m day^{-1} . However, it seems that the high flux in sample 12 of the upper trap is distributed over sample 12 and 13 in the lower trap. This demonstrates that there is a small time-lag between the two traps, of no more than a few days, due to the time it takes for the particles to settle. This could also be true for sample 24, however there is no sample directly after this last sample.

We added the following lines to the manuscript, in addition to Figure 2:

Page 4, lines 8-10 and page 5, lines 1-8:

“Since both the upper and lower traps are recovered for two of the five stations (M2 and M4), this allows for a direct comparison between the two depths. The upper and lower sediment traps are in very good accordance with

each other, demonstrated by images of the sediment-trap bottles after recovery (Fig. 2). Two samples, sample 12 and 24, have a much higher flux than the other samples, and these high-flux samples are present in both the upper and lower trap. Since the sampling interval is only 16 days, it means that the downward transport velocity of the sediments between the traps is at least 140 m day^{-1} and most likely much higher. It also shows that the sediments are deposited in a vertical way down to both sediment traps. It seems however that the higher flux observed in sample 12 of the upper trap is distributed over sample 12 and 13 of the lower trap. This demonstrates that there is a small time-lag between the two traps, of no more than a few days, due to the time it takes for the particles to settle. This could also be true for sample 24, however there is no sample directly after the last sample of the sediment trap.”

3. deposition rates. What are the deposition rates you are getting? Are they consistent with your assumptions? Are the deposition rates reasonable? Are they the same in the sediment traps as the sediment below? Please describe this a bit more.

Sediment fluxes of the sediment traps are beyond the scope of this paper, and a paper showing these results is in preparation at the moment. What we can conclude from the two events that were registered in the same bottles of traps that are positioned at a vertical distance of more than 2 km, is that settling rates through the water column are at least 140 m day^{-1} and probably higher. In any case they are in the order of days, not seasons. For the seafloor sediments, however, it is difficult to give a sedimentation rate for the upper 1 cm, since it is difficult to date these samples. Given the fact that they could be hundreds to thousands of years old, it is to be expected that they cannot be compared with our present-day samples directly. Mulitza et al. (2010) have shown that since the arrival of Portuguese colonists in Africa about 300 years before today, the change in land-use changed the emissions of northwest African dust dramatically. As the seafloor sediments presented in our manuscript could easily be older than 300 years, we present this line of reasoning as an explanation for the observed difference.

More details

“However, grain sizes in the seafloor sediments are substantially finer than found in the sediment-trap samples, and the downwind decrease in grain size is also less steep for the seafloor sediments.” What does this mean for interpretation? The more processed the cores, the finer they look? Or that they are being dissolved? Or that they are advected from farther upstream? I find this observation very difficult to understand, and makes me doubt your methodology.

As stated above, the main difference between the sediment traps and the seafloor sediments is the timing of the samples: the sediment traps have a very fixed time-resolution of 16 days. The seafloor sediments, however, are the result of accumulation of hundreds of years in the top centimeter alone. The grain-size distribution for the seafloor sediments is therefore an average of all these years, and the fact that the particle size is smaller means that over this long time period the dust was finer-grained than it is today (as found in the sediment traps). Dissolution of the particles is not likely, since most particles are quartz particles which are very resistant to this kind of processing.

“Since the seafloor sediments represent a longer time average of Saharan dust deposition than the sediment-trap samples, it implies that the downwind fining is a long-lived trend.” How long is the time average for the sediments on the seafloor compared to the traps?

See the comments above. The seafloor sediments are an accumulation of hundreds of years of sediments, and it is difficult to date the top centimeter. Typical accumulation rates for deep-sea sediments, however, are $1\text{-}5 \text{ cm kyr}^{-1}$ (Anderson, 2007), in this area possibly even lower, indicating that the top centimeter alone represents several hundreds to thousands of years.

“Mahowald et al. (2014) hypothesize that dust in the high atmosphere is finer grained than in the lower atmosphere, which is in turn finer than the deposited dust, due to the preferential settling of coarse particles. However, we observed giant particles ($\geq 100 \text{ m}$) as far as station M4 (49° W ; approximately 3500 km from African coast) (Fig. 4).” On the surface of it, these two statements have nothing to do with each other, since one is talking about vertical height in the atmosphere and the other is talking about horizontal distance from

Africa. You seem to be implying that they are somehow contradicting each other, but it doesn't seem possible to infer from distance downwind anything about vertical structure of the atmosphere?.

These two statements were meant to illustrate the preferential settling of coarse particles, however still giant particles are observed at great distances from the source. We removed this sentence in the revised version of the manuscript, since it is also stated in the previous sentence, and rephrased the two paragraphs:

Page 13, lines 5-17, and Page 14, lines 1-4:

“The grain size of dust decreases with increased distance from the source (Glaccum and Prospero, 1980;Goudie and Middleton, 2006;Mahowald et al., 2014;Stuut et al., 2005): coarse particles have a higher settling velocity and smaller particles can be transported over greater distances (Gillette, 1979;Tsoar and Pye, 1987). This mechanism accounts for the downwind fining observed in both the sediment traps and the seafloor sediments along the trans-Atlantic transect (Fig. 3). However, we observed giant particles ($\geq 100 \mu\text{m}$) at station M3 (38°W ; approximately 2400 km from the African coast) (Fig. 5), and also mica particles, whose platy shape allows for aerial transportation over greater distances (Stuut et al., 2005). Such coarse particles are generally not incorporated into climate models (Kok, 2011). Only a handful of these coarse particles are found in the samples, however when considering these are 1/25 splits of the original samples, collecting sediments over only 1 m^2 of ocean, over a time period of only 16 days, this means that the amount of giant particles being transported over the Atlantic Ocean is substantial. This underestimation of the coarse size fraction may have its origin in the sampling of dust of specific size classes, e.g. PM_{10} and $\text{PM}_{2.5}$, which form the basis of the guidelines from the World Health Organization (WHO, 2006) on fine-grained particles.

Since the seafloor sediments represent a longer time average of Saharan dust deposition than the sediment-trap samples, it implies that the downwind fining is a long-lived trend. However, the modal particle size of the sediment-trap samples is substantially coarser than that of the seafloor sediments at the same stations along the transect. [...]”

“The particle-size distribution found in the sediment-trap samples closely resemble Saharan dust sampled directly from the atmosphere, which has modal grain sizes varying between 8 and $42 \mu\text{m}$ (Stuut et al., 2005).” This is really important, but you don't say where this observation is made? Size is varying along the transect in the atmosphere also: : .. what type of observation is this? What kind of uncertainties are in that method (i.e. look at (Reid et al., 2003))

For more information on the scientific expedition during which these samples were collected and their exact sampling locations we refer to Figure 1 from Stuut et al. (2005).

We modified the following lines in the revised manuscript:

Page 14, lines 4-7:

“The particle-size distributions found in the sediment-trap samples closely resemble Saharan dust sampled directly from the atmosphere, by shipboard dust samplers along a transect off the West African coast, which has modal grain sizes varying between 8 and $42 \mu\text{m}$ (Stuut et al., 2005). This is in close resemblance with the observed modal grain size of 4 – $32 \mu\text{m}$ in the sediment traps.”

“By contrast, modal grain sizes in the underlying seafloor sediments range between 4 and $6 \mu\text{m}$. Since the seafloor sediments represent a longer time period, this suggests that Saharan dust was significantly finer in the recent past than it is today.” And in the conclusions: “Coarser dust found in the sediment traps opposed to the

seafloor sediments could result from emission of coarser dust due to the onset of commercial agriculture in the 19th century.” This is a huge jump, which seems incredibly unlikely. Most likely there is ocean processing: : .

The proposed mechanism is one of many possible causes for an increase in particle size over the past few hundred years. It is not meant to be conclusive or the sole mechanism behind this change in particle size over the last few centuries.

We modified the following lines in the revised manuscript:

Page 14, lines 7-14:

“By contrast, modal grain sizes in the underlying seafloor sediments range between 4 and 5.5 μm . Since the seafloor sediments represent a longer time period, this suggests that Saharan dust was significantly finer in the recent past and increased over the last centuries. Deposition of coarser dust is in line with increased emission as a result of human activity since the nineteenth century due to commercial agriculture (Mulitza et al., 2010). Not only does increased human activity in the source region increase dust emissions, it also enables larger particles to be emitted (McTainsh et al., 1997), which could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we see now in the sediment traps.”

Page 19, lines 7-9:

“Coarser dust is found in the sediment traps opposed to the seafloor sediments, in line with increased emission and coarser dust due to the onset of commercial agriculture in the 19th century.”

“The lower (3500 m) traps show less seasonality and are generally slightly coarser than the upper (1200 m) traps. This may be due to the disaggregation of marine snow, releasing the individual dust particles and thus decreasing their settling velocity. Therefore, it would take longer for particles to reach the lower traps at 3500 m, especially very fine particles, and as a result the particle-size distributions lose their seasonal characteristics. This would also explain why the dust in the lower traps (at M2 and M4) is slightly coarser than their upper counterparts, since these coarse particles settle more quickly, and the very fine particles may not reach the lower traps.” This is really important, and should be talked about first: you need to convince us that you can say anything about seasonality in the dust size from sediment trap data, especially with the observed bias between sediment trap and core sizes. So I would start from this and really convince us that any of the signal is actually from the atmosphere first.

See also previous comments about the difference between the sediment trap samples and seafloor sediments, and why we think the dust found in the sediment traps is a good representation of dust in the atmosphere.

We added the following paragraph to the revised manuscript:

Page 4, lines 8-10 and page 5, lines 1-17:

“Since both the upper and lower traps are recovered for two of the five stations (M2 and M4), this allows for a direct comparison between the two depths. The upper and lower sediment traps are in very good accordance with each other, demonstrated by images of the sediment-trap bottles after recovery (Fig. 2). Two samples, sample 12 and 24, have a much higher flux than the other samples, and these high-flux samples are present in both the upper and lower trap. Since the sampling interval is only 16 days, it means that the downward transport velocity of the sediments between the traps is at least 140 m day^{-1} and most likely much higher. It also shows that the sediments are deposited in a vertical way down to both sediment traps. It seems however that the higher flux observed in sample 12 of the upper trap is distributed over sample 12 and 13 of the lower trap. This demonstrates

that there is a small time-lag between the two traps, of no more than a few days, due to the time it takes for the particles to settle. This could also be true for sample 24, however there is no sample directly after the last sample of the sediment trap.

In this paper we argue that the lithogenic particles found in the sediment traps are of aeolian origin. The sediment traps are located far from the continental shelf, so riverine input of sediments is not affecting the samples. Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll or salinity (not shown; available from Giovanni NASA GES DISC: <http://giovanni.sci.gsfc.nasa.gov/giovanni/>). In addition, the lower sediment traps are positioned 880-1300 m above the seafloor, so resuspension of bottom sediments will not affect the sediment trap samples. When considering the large amounts of Saharan dust being transported across the Atlantic Ocean every year, about 182 Tg (Yu et al., 2015), any other external input is assumed to be negligible. Stuut et al. (2005) also demonstrated the similarity between aerosol samples of Saharan dust collected off west Africa and the lithogenic fraction in sediment traps and seafloor sediments.”

“We have shown seasonal and spatial changes in Saharan mineral dust transport and deposition across the Atlantic Ocean by means of sediment-trap sampling between October 2012 and November 2013, and seafloor sediments at the same stations.” So at this point, this statement has not been proven: you have only shown sea floor sediment changes in size. It is interesting that you see these trends, but anything about the atmospheric aerosols is speculation.

Indeed no seasonality can be seen for the seafloor sediments, unlike the sediment traps that sample at very high resolution (16 days).

We rephrased the following lines in the revised manuscript:

Page 19, lines 3-7:

“We have shown seasonal and spatial changes in Saharan mineral dust transport and deposition across the Atlantic Ocean by means of sediment-trap sampling between October 2012 and November 2013, and spatial changes in the seafloor sediments at the same stations. Our results show strong seasonal variations and significant fining in particle size with increasing distance from the source in the sediment trap samples, with modal particle diameters ranging from 4 to 32 μm .”

Han, Q., Moore, J.K., Zender, C., Measures, C., Hydes, D., 2008. Constraining oceanic dust deposition using surface ocean dissolved Al. *Global Biogeochemical Cycles* 22, doi:10.1029/2007GB002975.

Reid, J.S., Jonson, H., Maring, H., Smirnov, A., Savoie, D., Cliff, S., Reid, E., Livingston, J., Meier, M., Dubovik, O., Tsay, S.-C., 2003. Comparison of size and morphological measurements of dust particles from Africa. *Journal of Geophysical Research* 108, 8593: doi:1029/2002JD002485.

Siegel, D.A., Deuser, W.G., 1997. Trajectories of sinking particles in the Sargasso Sea: modeling of statistical funnels above deep-ocean sediment traps. *Deep-Sea Research* 44, 1519-1541.

Anonymous Referee #2 (Referee comments in black, our reply and changes in the manuscript in green)

Van der Does and colleagues present their preliminary findings for one year of data from a multi-year sampling campaign, aimed at retrieving samples of dust deposited to the equatorial Atlantic Ocean (at latitudes $\sim 12^\circ$ N), utilizing a transect of moored sediments traps at different water depths, downwind from the North African dust sources. The aim of this ambitious project to better constrain the evolution of the North African dust plume is of certain interest, and it is positive for the atmospheric and dust communities to be informed about these preliminary findings. This is a very interesting study, and the manuscript is in general well organized and quite clear. Nonetheless I do revise three major aspects that would deserve some revision, in order to clarify the work and perhaps improve the possible interpretation of the data.

General major comments

The particle size distributions are central in the manuscript. Nonetheless the descriptive metrics that are used for some of the diagnostic plots are only briefly mentioned. I think it would be very important to clearly show a validation of the metric used (e.g. mode of distributions fitted using GRADISTAT) against the specific observational data, before following with the discussion. This is particularly relevant since many of the samples show an apparent bi-modal distribution.

In this case we chose to represent the grain-size distributions with the mode instead of the median grain size, since the mode shows the most-occurring value. The median would also account for any secondary modes. However, these secondary modes, as also described in the manuscript, most likely represent platy mica particles, that due to their shape have a larger geometric size than their aerodynamic size. Therefore, we chose the mode to represent the grain-size distributions of our samples to better represent the geometric grain sizes.

In the discussion of the data, the coarseness of grain size distributions is assumed to mimic the behavior of dust deposition flux, despite the fact that such information is not reported. In addition, a full comparison among different samples is hampered by the lack of this piece of information.

In the discussion we speak only of increased dust emission (as seen from AOD and satellite data) in line with an increase in particle size of the dust found in the sediment traps which could be related. Specific flux data for the sediment traps should give more insight in this statement, but that is beyond the scope of this paper.

Some of the interpretations of the data provided in the discussion remain rather speculative. Please see the specific comments below.

Specific comments

2 / 8-9. Which is the mechanism that would explain this statement, in relation to the previous sentence?

As discussed in the discussion section of the paper, increased wind velocities in the SAL, combined with the higher elevation of the dust particles, results in the transportation of coarse particles over greater distances. Also convection within the SAL keeps coarse particles suspended. When high up in the atmosphere it takes more time for particles to settle down, and when also transported in a lateral sense these particles can reach further across the Atlantic Ocean.

3 / 26-32. I would argue that the point here is that we need to quantify how far a significant number of those particles can travel, to both (a) constrain the inputs to the ocean and (b) be able to estimate if, because of their actual amount, they are in fact relevant in terms of direct radiative effects or rather they could actually be ignored from this point of view, as typically done so far in models. That is why it would be important to have dust deposition fluxes associated to the size distribution data. If this piece of information is not available, the discussion should take into account this fact, and the interpretation of similarities / differences in the samples should be pondered accordingly.

Dust fluxes of the sediment traps are beyond the scope of this paper. With this information, however, it would be difficult to give an accurate number of the amount of particles larger than a certain diameter. The grain-size

distributions only show relative volumes or the particles, and not actual particle counts. However, given that about 182 Tg of dust are transported over the Atlantic Ocean every year (Yu et al., 2015), the samples collected for our study have a temporal resolution of only 16 days, the collection area of the sediment traps is 1 m², the analyzed split of these samples is 1/25, and that we can see at least a handful of these giant particles per sample illustrates that the amount of these coarse particles transported in the atmosphere must be substantial.

4 / 30. Briefly, why did you discard some of the traps?

See caption of Figure 1; three of the ten traps could not be recovered due to material failure and rough weather during recovery.

We modified the following lines in the manuscript:

Page 3, lines 37-39, and page 4, lines 1-2:

“This paper presents the results of successful sampling by seven sediment traps on the five moorings from 19 October 2012 to 7 November 2013 (Stuut et al., 2013). These include three of the upper (1200 m) sediment traps located at mooring stations M1, M2 and M4, and four lower (3500 m) sediment traps at stations M2, M3, M4 and M5 (Fig. 1, Table 1). Three of the ten sediment traps could not be recovered.”

5 / 19-20. Please clarify if you refer to radius or diameter, here and throughout the manuscript.

All particle sizes referred to in the manuscript are described as equivalent-sphere diameter of the particle.

We modified the following lines in the manuscript:

Page 6, lines 6-8:

“This resulted in particle-size distributions consisting of 92 size classes ranging from 0.375 to 2000 μm describing the equivalent-sphere diameter of the particle. Modal particle size is also expressed as particle diameter.”

5 / 20-21. As indicated above, please discuss much more extensively this aspect. For instance, describe how the method works, and show the comparison of the full distribution and the metric (mode) for two-three representative cases, e.g. a typical sample for each Winter, Summer, Spring from Figure 5. This should highlight how the metric vary according to the distribution's shape, thus help better understanding/constraining the following interpretations. This should also highlight whether the choice of the metric is the best option or better ones could be adopted in this case.

For a discussion why the mode should be used for describing the particle size of the dust, please refer to the first comment (page 8 of this letter). A note on why the method of Folk and Ward (1957) was used: initially we chose the median diameter to describe the particle size, and in this case there is a difference in what the Coulter Laser Diffraction Particle Sizer (LS13 320) calculates. This method also allows for better comparison to other methods (Blott and Pye, 2001). For the modal diameter, however, the GRADISTAT program recalculates the particle-size classes, resulting in different modal diameters than based on the size classes of the Coulter. We decided to change these modal diameter to the original mode given by the Coulter (the raw data), and not refer to the Folk and Ward method. We changed all the graphs accordingly. This does not change the trends we see in particle size, but alters the absolute values of the modal diameter slightly.

We modified the following lines in the manuscript:

Page 6, lines 6-8:

“This resulted in particle-size distributions consisting of 92 logarithmic size classes ranging from 0.375 to 2000 μm describing the equivalent-sphere diameter of the particle. Modal particle size is also expressed as particle diameter.”

This also resulted in small changes regarding the modal particle size shown in Figures 2, 6, 7, 8 and 11 (Figures 3, 7, 8, 9 and 12 in revised manuscript).

5 / 30. Did you only simulate four days? Later in the manuscript (9 / 31-32) it sounds like you may have selected four days out of a larger ensemble? Is that the case? If so, it would be interesting to see those. If not, how did you exactly determine that those would be representative days?

For the backward trajectories, only four days were simulated. This is a balance between a good insight of where the starting point of the air parcel is, and modelling uncertainties with simulating longer time periods. It is also used more frequently in literature (e.g. Stuut et al. (2005)). Only a basic overview was made for backward trajectories starting at station M1 for the entire sampling period, to see if there would be seasonal differences. This resulted in a very wide range of trajectories, showing no clear seasonal trend, and which can't be visualized in a clear way. The cases shown in the manuscript were chosen as a clear example for summer and winter dust transportation.

6 / 1. Do you have dust deposition flux data? I believe that all the comparisons among the samples in this study and the derived interpretations are subject to the limitation of not being associated to dust deposition fluxes. Therefore only partial information is available to derive conclusions.

Dust fluxes of the sediment traps are beyond the scope of this paper. We realise that they are of large scientific interest for many different scientific disciplines. Therefore, they are presently being determined, however not for this particular publication.

7 / 14-17. As already mentioned, I think that the point is not whether a handful of giant particles make it a great distance, but rather how many and how far. If they appear to be quantitatively important, then this suggest that models should account for that, and they will need data to constrain their results. Hopefully your study will help addressing this issue!

As already mentioned in one of the previous comments (on page 8-9 of this letter), it is difficult to give an accurate number of the amount of particles larger than a certain diameter. However, given that about 182 Tg of dust are transported over the Atlantic Ocean every year (Yu et al., 2015), the samples collected for our study have a temporal resolution of only 16 days, the collection area of the sediment traps is 1 m^2 , the analyzed split of these samples is 1/25, and that we can see at least a handful of these giant particles per sample illustrates that the amount of these coarse particles transported in the atmosphere must be substantial.

7 / 17. “Preferentially” vs what? Please clarify this sentence.

The platy particles are more easily transported over greater distances than deposited close to the source, opposed to larger spherical particles. We acknowledge this sentence is not clear, and therefore removed it from the revised manuscript. We left the following statement in:

Page 13, lines 9-11:

“However, we observed giant particles ($\geq 100 \mu\text{m}$) at station M3 (38° W ; approximately 2400 km from the African coast) (Fig. 5), and also mica particles, whose platy shape allows for aerial transportation over greater distances (Stuut et al., 2005).”

7 / 25-30. This paragraph seems very speculative: there is no support to it in the discussion, and no time control is reported about the age of those seafloor sediments.

The proposed mechanism is one of many possible causes for an increase in particle size over the past few hundred years. It is not meant to be conclusive or the sole mechanism behind this change in particle size over the last few centuries.

We modified the following lines in the revised manuscript:

Page 14, lines 8-14:

“Since the seafloor sediments represent a longer time period, this suggests that Saharan dust was significantly finer in the recent past and increased over the last centuries. Deposition of coarser dust is in line with increased emission as a result of human activity since the nineteenth century due to commercial agriculture (Mulitza et al., 2010). Not only does increased human activity in the source region increase dust emissions, it also enables larger particles to be emitted (McTainsh et al., 1997), which could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we see now in the sediment traps.”

8 / 11-15. Here you seem to suggest a direct relation between coarse grain size and high dust load (or AOD), and for extension to a high deposition flux? The reported study of Skonieczny et al. (2013) on the other hand shows coarser dust deposition at M’Bour, Senegal, associated with the season of low dust deposition flux. How would you justify your assumption in light of that? I think that absolute magnitudes of size distributions could help here in two different ways, most importantly with reference to Figures 6, 7, 8, 11. First, absolute values of particles concentrations (i.e. counting statistics on the direct output from the particle counter) may help to understand if the “shoulders” associated to the larger particles are actually statistically significant in all cases. One can see “tail effects” associated to sometimes individual large particles in low concentration samples such as from ice cores (e.g. Albani et al., 2012). This piece of information should be considered together with the choice of the mode as a metric to compare those samples. Second, even when samples are screened against possibly noisy signals, any interpretation on the actual quantitative transport potential (whether with season, or distance, or depth) of giant particles remains speculative without deposition flux data. The same way, in order to trace the spatial evolution of the North African dust plume, size distributions are necessary but not sufficient. Comparing sediment records from the Atlantic on different size ranges in fact yield surprising results, demonstrating the importance of considering both size distributions and fluxes (Albani et al., 2015). If this piece of information is missing, then the discussion should be extended to discuss the possible limitations of the derived interpretations.

In the discussion we speak only of increased dust emission (as seen from AOD and satellite data) in line with an increase in particle size of the dust found in the sediment traps which could be related. The study by Skonieczny et al. (2013) shows a similar trend as what we find in our study, with coarser dust deposition during summer. The fact that we see increased dust transportation (as reflected by the AOD) does not have to correspond with increased dust deposition. Dust fluxes for the sediment trap samples will give more insight in these mechanisms, but they are beyond the scope of this paper. However, these data will not provide number distributions of the particle size, as the method for particle size analysis describes the results as relative volumes of the particles, and not actual particle counts.

Our results are different from dust collected in ice cores, since the sediment trap samples have much more dust in them. For the ice core samples, these “tail effects” are much more significant since there is much less dust in each sample. In addition, the Antarctic ice sheet is 2315 m above sea level, decreasing the probability of coarse particles being uplifted to these heights and deposited on the ice sheets.

8 / 24-28. Interesting approach!
Thank you!

8 / 31-32. Quite the opposite. I cite: “On balance, the measurements (Fig. 4) indicate that dust PSD is independent of the wind speed at emission. This conclusion is supported ...”

Cited from Mahowald et al. (2014): (p. 64) “The results suggest that an increase in wind speed can be associated with a small (0.15 μm) increase in dust particle size downwind of the sources (Fig. 11). This is consistent with the paleoclimate interpretation that stronger winds will carry larger particles. “

And (p. 67): “The size of individual particles is to a large extent set at emission, [...]”

We modified the following lines in the revised manuscript:

Page 15, line 16 and page 16, lines 1-5:

“Mahowald et al. (2014) argue that the dust particle size does not depend on wind speeds at emission. However, high wind velocities in the SAL of $>7 \text{ ms}^{-1}$ (Tsamalis et al., 2013) enables coarser dust particles to remain in suspension in summer, and due to the high altitude these coarse particles are transported over great distances. In addition, increased convection in the source areas in summer, related to larger differences in temperature, can result in the uplift of coarser dust particles (Heinold et al., 2013).”

9 / 3. I would suggest changing “these air layers” with something like “the starting points for back-trajectories calculations”.

We agree. We modified the following lines in the revised manuscript:

Page 16, lines 8-11:

“The altitudes of the starting points of these backward trajectories were chosen in accordance with the hypothesized heights of the dust-carrying air layers, as demonstrated in Figures 11A and -B, with the lowest (500 m) elevation representing winter dust transport and the highest (3500 m) elevation representing summer dust.”

9 / 8-9. This sentence is not very clear, please rephrase.

We agree. We modified the following lines in the revised manuscript:

Page 16, lines 11-22:

“In winter (Fig. 11C), the higher trajectory is not originating from the African continent, and therefore the winds at these altitude are unlikely to transport dust to the sample location. The lower trajectory has a more eastern origin, and air layers at this altitude could be transporting dust (Fig. 11A), picked up from the surface and brought to higher altitudes. By contrast, in summer (Fig. 11D) this situation is reversed: the higher trajectory has a more continental origin and is the most likely dust-carrying air layer over the lower trajectory. The elevation profile shows that this high-elevation trajectory started at lower altitudes, but upon reaching the coastline it was uplifted to about 3500 m AGL (Fig. 11D, bottom panel). This is in accordance with how the Saharan Air Layer (SAL) is described, when dust-carrying air from the continent is uplifted by a cool marine inversion layer (Carlson and Prospero, 1972; Prospero and Carlson, 1972). This inverted air layer is visible in the 500 m air layer, moving in an opposite direction, from west to east. After this sharp increase in altitude, the trajectory decreases in altitude, which persists across the Atlantic Ocean (Tsamalis et al., 2013).”

9 / 12-14. It seems that here “air-layer” is used to indicate “air parcel trajectory”?

This comment is dealt with by the change in the manuscript discussed above.

9 / 15-18. You are not showing this. Please at least provide some reference.

We modified the following lines in the revised manuscript:

Page 17, lines 8-10:

“The summer season is also characterized by an increased number of more intense dust storms (e.g. Adams et al. (2012)). From May to September, dust is almost continuously emitted from the African continent, as shown by satellite images (MODIS Terra and Aqua satellites; NASA Worldview).”

9 / 18. “Increased deposition”: where?

What is meant here is “increased deposition of coarse particles”, relative to the other particles, so coarser dust deposition.

9 / 21-22. How do you know? You do not show any information about the atmospheric column above.

We can only note the apparent coincidence of increased particle size and increased precipitation. It can be one of many mechanisms related to deposition of coarser particles.

We modified the following lines in the revised manuscript:

Page 17, lines 12-16:

“Increased deposition of coarse particles can also be caused by increased precipitation in summer and fall, as opposed to almost no precipitation in winter and spring (Fig. 12). This was also noted off northwest Africa related to wet deposition was also noted by Friese et al. (In press). Increased precipitation at station M1 seems to coincide with increased modal grain sizes, and this relation commences with lowest precipitation early June 2013.”

9 / 23-24. Again, it is not clear whether the mode is a good metric to compare bi-modal distributions.

It describes the trends that are most clear in seasonality of the dust particle size. The main mode that occurs in all samples is thought to be made up of quartz particles, the second mode consists of mostly coarse, platy mica particles, and Fig. 12 is used to better demonstrate this second coarser mode (see also previous comments). Most samples however do not show a bi-modal grain-size distribution, hence the choice for comparing the mode of the different samples.

9 / 25-30. How does a laser particle counter sees a flat particle? Overestimate it’s spherical equivalent diameter? See e.g. Reid et al. (2003). How do you interpret this in your data, and according to the evolution of size distribution with distance from the source?

The laser particle sizer measures the diameter of the particle as it is oriented towards the laser beam. As the sample is being constantly homogenized by a magnetic stirrer, there is no preferred orientation for particles and thus random. As a result, the flat particles will be measured (in theory) in an infinite number of ways, and hence detected as a smaller or larger particle, depending on its orientation. The result would be an average of the smallest and largest diameter of equivalent spherical particle, but since it is described as volume percentage, the larger would have more influence on the grain-size distributions. Therefore it was chosen to describe the data with the modal diameter, and separately address the second mode of the distributions (see also previous comments).

9 / 30-32. As already mentioned, if more back-trajectories calculations were performed, it would be interesting to see them.

See answer to a previous comment (page 10 of this letter).

10 / 1-6. Also in this respect, absolute values of concentration and most importantly dust deposition fluxes might shed some light on the issue. In addition, a little more discussion on the fate of particles throughout the water column and the expected relation to the corresponding surface water and atmosphere could be added here.

Since we have recovered both sediment traps, the upper (1200 m) and lower (3500 m), at two of the stations (M2 and M4), we can compare the data between these two traps. A very clear example is visible for the samples at M4, where two samples (sample 12 and 24, collected during spring and fall, respectively) with very high fluxes are present at both the upper and the lower trap, in the same sampling cup. Since the sampling interval is only 16 days, it means that the downward transport velocity of these sediments is at least 140 m day^{-1} . Sediment fluxes are beyond the scope of this paper, however a new Figure has been added to the present manuscript showing sediment-trap bottles after recovery, with high levels of sediments in the aforementioned samples (Fig. 2 of revised manuscript). This demonstrates the similarity in sediments received for both sediment traps, and that lateral advection is minimal.

10 / 11. Please add also here in the conclusions whether you refer to particle radius or diameter.

We modified the following lines in the revised manuscript:

Page 19, lines 3-7:

“We have shown changes in Saharan mineral dust transport and deposition across the Atlantic Ocean by means of sediment-trap sampling between October 2012 and November 2013, and seafloor sediments at the same stations. Our results show strong seasonal variations and significant fining in particle size with increasing distance from the source in the sediment trap samples, with modal particle diameters ranging from 4 to $32 \mu\text{m}$.”

10 / 11-12. As indicated earlier, this statement is so far very speculative.

See also previous comments on page 10-11 of this letter. The proposed mechanism is one of many possible causes for an increase in particle size over the past few hundred years. It is not meant to be conclusive or the sole mechanism behind this change in particle size over the last few centuries.

We modified the following lines in the revised manuscript:

Page 19, lines 7-9:

“Coarser dust is found in the sediment traps opposed to the seafloor sediments, in line with increased emission and coarser dust due to the onset of commercial agriculture in the 19th century.”

10 / 22-23. From your study, one would expect to learn how many.

See also previous comments on page 8-9 and 10.

Dust fluxes of the sediment traps are beyond the scope of this paper, and it is difficult to give an accurate number of the amount of particles larger than a certain diameter. However, given that about 182 Tg of dust are transported over the Atlantic Ocean every year (Yu et al., 2015), the samples collected for our study have a temporal resolution of only 16 days, the collection area of the sediment traps is 1 m^2 , the analyzed split of these samples is 1/25, and that we can see at least a handful of these giant particles per sample illustrates that the amount of these coarse particles transported in the atmosphere must be substantial.

Figure 2. Please differentiate the markers based on the depth for M2 and M4.

We modified the indicated figure in the revised manuscript (Figure 3 in revised manuscript) and the figure caption.

Figure 7. Could you provide a brief explanation about those outliers?

The measurements for these two samples are out of the entire range of particle size, for all stations, and seem very unlikely. This may be related to analytical or processing errors, since there are many steps involved before the particle size is analyzed. Out of 168 samples analyzed for this paper, only two unrealistic outliers appear, which we consider reasonable. For this reason, we chose to not elaborate on this in the manuscript.

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Anonymous Referee #3 (Referee comments in black, our reply and changes in the manuscript in blue)

van der Does et al. present observations of what is thought to be Saharan dust along the trans-Atlantic transport pathway over the course of roughly a year. Their observations cover a wide lateral range across the Atlantic Ocean and demonstrate the decrease in particle size with increasing distance from the source. Additionally the paper is well written. Although these observations are interesting and worthy of representation in the literature, there are a few major concerns I express in the below review that should be addressed prior to final publication.

Major concerns

The assumption that the particles collected in the traps are all mineral dust from the Sahara seems like an over-interpretation of the results. First, there could still be interference from biological particles. The authors did carryout chemical degradation and deactivation techniques to denature biological constituents. However, these types of methods do not remove all of the viable cells; they simply kill them off while leaving behind a particle. They do not completely disintegrate under these methods. How did the authors account for leftover, dead cells or biological particles such as pollen or marine microorganisms, which can easily fall within the size range of what was measured? Second, there could also be contribution from sea salt particles or non-viable organic material from the ocean surface microlayer. How did the authors eliminate these other types of particles as potential candidates for what was sized? Third, the dearth of chemical or mineralogical analysis also forces me to question the conclusion that most of what was observed was dust. This could easily alleviate the issue by imaging and/or determining the composition of the particles in the samples. The authors do show one image of a dust particle, but was this conducted for all samples and multiple particles per sample? Maybe SEM/EDX, XRF, and/or XRD were conducted? Surely it may be too late to conduct such analyses, and if the authors decide to proceed with publication with the current methods only, should very clearly state the assumptions made regarding what the particles are and perhaps provide more background from previous work demonstrating dust observations in the Atlantic Ocean to support their assumptions. As a suggestion, it might be beneficial to look at salinity and surface chlorophyll concentrations of the domain over which the particles were transported to show possible sources of particles other than dust (or partially eliminate these as contributing sources).

1. As described in the methods section of the manuscript, all biogenic particles are removed prior to particle-size analysis.

Page 5, lines 32-33 and page 6, lines 1-3:

“Biogenic constituents were removed in three steps to isolate the insoluble or lithogenic dust fraction from all samples prior to grain-size analysis, following the procedure described by McGregor et al. (2009). Shortly, organic matter was oxidized using H₂O₂, followed by dissolving the biogenic carbonates using HCl, and removing biogenic silica by adding NaOH. Immediately prior to the grain-size measurements sodium pyrophosphate (Na₄P₂O₇·10H₂O) was added to ensure complete disaggregation of the particles.”

2. Sea-salt particles would dissolve in water, and in addition the salt is removed from the sample during the wet-splitting of the samples, after which they are washed and centrifuged, as described in the methods section of the manuscript.

Page 5, lines 27-30:

“For grain-size analysis, one of these aliquots was split into another 5 subsamples (1/25 of the original sample), that were washed and centrifuged repeatedly at approximately 1800 x g with Milli-Q water to remove the HgCl₂, borax, and sea-salts.”

3. In addition, more images of dust particles have been added to the revised manuscript (Fig. 5). No substantial SEM/EDX or XRD measurements have been performed on the sediment trap samples. XRF measurements have been performed (on the bulk sample, however), and will be published at a later stage. Indeed the assumption is made that the lithogenic fraction of the sediment-trap samples are dust particles. The sediment traps are far from the continental shelf, so riverine input of sediments is thought to be none. Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll A or salinity, available from Giovanni of NASA GES DISC (<http://giovanni.sci.gsfc.nasa.gov/giovanni/>). Also, the lower sediment traps are 880-1300 m from the seafloor (M2-M5), so resuspension of bottom sediments will not affect the sediment trap samples. When considering the great amounts of Saharan dust being transported every year, about 182 million tons (Yu et al., 2015), it is fair to consider the lithogenic fraction in the sediment traps to be of aeolian origin.

How representative are the lower and sea floor traps of the observations of particle deposition and sedimentation during the study time period? Especially the sea floor, could these particles result from years of sedimentation and ocean circulation/currents introducing particles from all over the ocean system? It seems as if the ocean floor would be even more of a hodgepodge of all types of particles; this is where some sort of compositional information on the particles in the samples would be useful. Along these lines, I am not convinced that the smaller particles observed on the sea floor are simply due to the fact that larger particle emission has occurred over time based on the methodology and observations presented.

At two of the stations, M2 and M4, we recovered both the upper (1200 m) and lower (3500 m) sediment traps. Here we can compare both traps and see differences and similarities between the two. In the revised manuscript we added a new figure (Fig. 2), highlighting two high-flux samples at station M4; samples 12 and 24. The fact that this high flux is visible at both depths, and in the same sampling cup, illustrates the high settling velocity of the dust particles. Since the sampling interval is only 16 days, it means that the downward transport velocity is at least 140 m day^{-1} . This also demonstrates the similarity in sediment received for both sediment traps, and that lateral advection is minimal.

The seafloor sediments are almost undisturbed. At these locations and at these great depths, biological activity at the seafloor is very minimal. Indeed, these are the result of years of sedimentation, and it is not easy to give a date to this top centimeter of sediments from the seafloor. Typical accumulation rates for deep-sea sediments, however, are $1\text{-}5 \text{ cm kyr}^{-1}$ (Anderson, 2007), indicating that the top centimeter alone represents several hundreds of years.

We hypothesize that the increased emission and emission of coarser particles in the recent past due to commercial agriculture is a possible explanation of the coarsening of dust in the samples, but there is no direct evidence for this.

We modified the following lines in the revised manuscript:

Page 14, lines 7-14:

“By contrast, modal grain sizes in the underlying seafloor sediments range between 4 and 6 μm . Since the seafloor sediments represent a longer time period, this suggests that Saharan dust was significantly finer in the recent past and increased over the last centuries. Deposition of coarser dust is in line with increased emission as a result of human activity since the nineteenth century due to commercial agriculture (Mulitza et al., 2010). Not only does increased human activity in the source region increase dust emissions, it also enables larger particles to be emitted (McTainsh et al., 1997), which could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we see now in the sediment traps.”

Page 19, lines 7-9:

“Coarser dust is found in the sediment traps opposed to the seafloor sediments, in line with increased emission and coarser dust due to the onset of commercial agriculture in the 19th century.”

Although it is generally understood that the SAL is transported westward over the Atlantic, the authors draw many conclusions of the seasonal altitude dependence of air mass transport and at only one trap location (M1). What would strengthen the argument regarding the impact of transport conditions and seasonal climate patterns on particle deposition/size is an ensemble or cluster analysis of HYSPLIT trajectories. The authors do state, “However, backward trajectories calculated over the entire sampling period do not suggest this: : :” which indicates that more trajectories were simulated. It would be helpful to show these to clearly show the seasonal variability. It would also be useful to conduct HYSPLIT analyses at all of the trap locations to better connect the sites and perhaps show that transport over the trap farthest from Africa does not experience as much transport as the trap closest.

More focus is on sampling station M1 since this is closest to the source, and the differences between seasons are greatest here. However as can be seen from the grain-size data, seasonality is present for all the five stations.

Only a basic overview was made for backward trajectories starting at station M1 for the entire sampling period, to see if there would be seasonal differences. This resulted in a very wide range of trajectories, showing no clear seasonal trend, and which can't be visualized in a clear way. The two cases shown in the manuscript were chosen as a clear example for summer and winter dust transportation. Again station M1 was chosen for this, as it is located closest to the source. Backward trajectories for the other stations could be useful, but since the distance to the source is greater there are more uncertainties, and it will be a futile task to attempt to illustrate air-layer trajectories for the entire sampling period for all stations. From satellite images it becomes very clear that dust is transported from the African continent over the Atlantic Ocean and the sampling stations, and the backward trajectories were intended to show typical summer and winter transport of dust, and to illustrate the seasonal differences.

General comments

The figures present data from a number of sources (i.e., MODIS and particle imaging). Although the captions to these figures briefly describe these data sources, they should be more comprehensively described in the methods section. As an example, what instrument was used to image the particles? How many images were acquired? Was this conducted for all samples? With respect to MODIS, provide at the very least a brief description of the satellite and how the data were acquired. For the precipitation, was this acquired from TRMM? Over what domain?

The microscope images were performed with a normal light microscope, and the ones shown were chosen to act as an example for the coarse particles found in the samples. This was not done for all 168 samples, however from the grain-size distributions it is clear that these coarse particles are present in most samples. The image acts as an aid to illustrate that these coarse particles are also solid quartz particles, and not only platy mica particles, and that the coarse particles measured are not simply aggregates of smaller particles.

In the revised manuscript, we added more microscope images of large particles (*Fig. 5*), and modified the figure caption:

“Figure 5. Light-microscope images of large dust particles from the lower (3500 m) traps at station M2 (13° N, 37° W; A and B) and at station M3 (12° N, 38° W; C and D). Both stations are situated at more than 2000 km from the African source. A: Large quartz particle (diameter approximately 180 μm over long axis) from sample 1 (October 19 – November 4, 2012). B: Large quartz particle (diameter approximately 290 μm over long axis) from sample 1. C: Large quartz particle (diameter approximately 200 μm over long axis) from sample 4 (December 6 – 22, 2012). D: Large mica particle (diameter approximately 86 μm over long axis) from sample 4.”

We also added more information about the MODIS images and precipitation data in the respective figure captions.

We added the following paragraph to the Materials and methods section of the revised manuscript:

Page 6, lines 22-25:

“Data for Aerosol Optical Depth (AOD) and daily precipitation were obtained from the Giovanni online data system, developed and maintained by the NASA GES DISC. AOD data was obtained from MODIS Terra, at monthly resolution and averaged over the respective seasons. Daily precipitation data from TRMM was used, averaged over the area between 11 - 13° N and 22 - 24° W (station M1).”

Specific comments

Page 2, line 19: Most people know what CALIPSO is, but do define the acronym.

We modified the following lines in the revised manuscript:

Page 1, lines 30-33:

“CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) lidar measurements between 2007 and 2013 show that annually 182 Tg of African dust leaves the African continent towards the Atlantic Ocean, 132 Tg reaches 35° W, and 43 Tg reaches as far west as 75° W (Yu et al., 2015).”

Page 9, line 23: Only sand can be this size? What about large minerals? This seems like a vague definition without any measurements of the mineralogy.

In sedimentology, the term “sand” is used as a classification of particle size: any mineral particles between 63 and 2000 μm . Hence, the term sand does not imply anything about its properties; any material in this size range may be called sand.

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Particle size traces modern Saharan dust transport and deposition across the equatorial North Atlantic

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Abstract. Mineral dust has a large impact on regional and global climate, depending on its particle size. Especially in the Atlantic Ocean downwind of the Sahara, the largest dust source on earth, the effects can be substantial but are poorly understood. This study focuses on seasonal and spatial variations in particle size of Saharan dust deposition across the Atlantic Ocean, using an array of submarine sediment traps moored along a transect at 12° N. We show that the particle size decreases downwind with increased distance from the Saharan source, due to higher gravitational settling velocities of coarse particles in the atmosphere. Modal grain sizes vary between 4 and 32 μm throughout the different seasons and at five locations along the transect. This is much coarser than previously suggested and incorporated into climate models. In addition, seasonal changes are prominent, with coarser dust in summer, and finer dust in winter and spring. Such seasonal changes are caused by transport at higher altitudes and at greater wind velocities during summer than in winter. Also the latitudinal migration of the dust cloud, associated with the Intertropical Convergence Zone, causes seasonal differences in deposition as the summer dust cloud is located more to the north, and more directly above the sampled transect. Furthermore, increased precipitation and more frequent dust storms in summer coincide with coarser dust deposition. Our findings contribute to understanding Saharan dust transport and deposition relevant for the interpretation of sedimentary records for climate reconstructions, as well as for global and regional models for improved prediction of future climate.

Keywords Mineral dust; Atlantic Ocean; grain size; Saharan dust transport; seasonality

1 Introduction

Millions of tons of mineral dust are transported from the African continent towards the Atlantic Ocean every year, with several direct and indirect effects on global climate. CALIPSO ([Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation](#)) lidar measurements between 2007 and 2013 show that annually 182 Tg of African dust leaves the African continent towards the Atlantic Ocean, 132 Tg reaches 35° W, and 43 Tg reaches as far west as 75° W (Yu et al., 2015). Approximately 140 Tg is deposited in the Atlantic Ocean between 15 and 75° W and 10° S and 30° N. Atmospheric mineral dust affects the atmosphere's radiation budget by scattering and absorbing incoming and reflected solar radiation, and changes cloud properties by acting as cloud condensation nuclei (Goudie and Middleton, 2001; Highwood and Ryder, 2014; Shao et al., 2011; Wilcox et al.,

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2010). Climatic effects are largely determined by particle characteristics including particle size, particle shape, chemical- and mineralogical composition, and by cloud cover and the albedo of the underlying surface (Claquin et al., 2003;Goudie and Middleton, 2001, 2006;Highwood and Ryder, 2014;Otto et al., 2007;Shao et al., 2011;Sokolik and Toon, 1999). Large particles in the lower atmosphere may have a warming effect on earth's climate by absorbing reflected (long-wave) radiation (Mahowald et al., 2014;Otto et al., 2007). By contrast, small particles in the higher atmosphere may have a cooling effect, by reflecting incoming solar (short-wave) radiation (Claquin et al., 2003;Mahowald et al., 2014). Moreover, dust deposition enhances ocean carbon cycling by delivering nutrients that stimulate phytoplankton growth (Martin and Fitzwater, 1988;Shao et al., 2011). In turn, this not only leads to increased export fluxes but also by faster transport of organic carbon to the deep ocean, as dust particles act as mineral ballast, depending on particle size, shape and mineral density (Armstrong et al., 2002;Bressac et al., 2014;Fischer et al., 2007;Fischer and Karakas, 2009;Klaas and Archer, 2002). Both have the potential to reduce atmospheric pCO₂ levels (Klaas and Archer, 2002).

The distance over which mineral dust is transported depends on the transporting winds and particle characteristics including size, shape and density, which determine settling velocities. Thus, rounded quartz and feldspar particles have a greater settling velocity than platy clay minerals, and are therefore deposited closer to the source (Glaccum and Prospero, 1980;Goudie and Middleton, 2006;Mahowald et al., 2014;Stuut et al., 2005). Saharan dust is transported with the trade winds year-round, from the northwestern Sahara to the eastern Atlantic Ocean. During winter, the Harmattan trade winds prevail, transporting dust from the central Sahara (Glaccum and Prospero, 1980;Stuut et al., 2005) at altitudes between 0 and 3 km (Tsamalis et al., 2013). In summer, when the larger land-sea temperature contrast results in large convective cells over the African continent, dust is emitted from the Sahara and Sahel. During transport towards the Atlantic Ocean, cool marine air blows in the opposite direction and lifts the warm, dusty air high up in the atmosphere. This Saharan air layer (SAL) is confined between two inversion layers, at 1 and 5 km height (Carlson and Prospero, 1972;Kanitz et al., 2014;Prospero and Carlson, 1972;Tsamalis et al., 2013). Due to the latitudinal movement of the ITCZ (Intertropical Convergence Zone), the dust cloud over the Atlantic Ocean also migrates seasonally (Nicholson, 2000), shifting northward (10–20°N) in summer and southward (0–10°N) in winter (Adams et al., 2012;Holz et al., 2004;Moulin et al., 1997;Yu et al., 2015).

The particle size of entrained and transported mineral dust depends on source conditions including surface roughness, wind velocity and erosion threshold, and soil characteristics including particle size, -shape, -density and soil moisture (d'Almeida and Schütz, 1983;Marticorena, 2014). After entrainment, the particle-size distributions are further modified by size-selective processes during transport and deposition (Grini and Zender, 2004). Owing to gravitational settling, dust particle size decreases with increasing distance from the source (Holz et al., 2004;Mahowald et al., 2014;Sarnthein et al., 1981;Schütz, 1980) and generally do not exceed 20 µm when transported over long distances (Gillette, 1979;Tsoar and Pye, 1987). On the Cape Verde islands close to the Saharan source, Glaccum and Prospero (1980) found individual quartz and mica particles of up to 90 and 350 µm, respectively. However, various studies reported giant (> 62.5 µm) mineral dust particles also at much greater distances (> 10,000 km) from their source (Betzer et al., 1988;Goudie and Middleton, 2006;Mahowald et al., 2014;Middleton et al., 2001). Climate models usually do not account for such coarse particles, and generally

overestimate the fine fraction (Grini and Zender, 2004;Kok, 2011). This not only results in an underestimation of the dust flux to the oceans and in turn the fertilizing effect of the transported nutrients, it also produces errors in the sign and magnitude of radiative forcing by dust and the formation of cloud condensation nuclei. This affects weather forecasts and climate predictions, especially in dusty regions (Kok, 2011).

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Due to their vastness, dust over the oceans has remained poorly studied, although specific information is required for predicting future climate and past climate reconstructions (IPCC, 2013). For the present study, we focused on a transect across the Atlantic Ocean, located directly underneath the Saharan dust cloud at 12° N (Yu et al., 2015). We used time-series submarine sediment traps moored at five locations along this transect, sampling synchronously at a resolution of 16 days. Here we present the first-year results on seasonal variability over the full particle-size range, analyzing source-to-sink variation of particle size in relation to large-scale atmospheric processes. Atmospheric Saharan dust has been collected at daily resolution at Barbados for more than 50 years (Prospero and Carlson, 1970;Prospero and Nees, 1977;Prospero et al., 1981;Prospero and Nees, 1986;Prospero and Lamb, 2003). Although the longest dust record sampled to date, it is at a single and distal location relative to the Saharan source. Croot et al. (2004) sampled Saharan dust < 1 μm in fall 2002 from the atmosphere along a transect across the Atlantic Ocean, while Stuut et al. (2005) also considered larger particles by shipboard sampling in winter 1998. Also Skonieczny et al. (2013) observed temporal changes in dust outbreaks and particle characteristics like grain size and chemistry, at a single proximal location on the western African coast. They found higher fluxes during winter, as opposed to coarser particles during summer, and attribute this to the seasonally different transporting dust layers. Similar higher fluxes of coarser-grained lithogenic particles in summer were observed by Ratmeyer et al. (1999a;1999b), using a submarine sediment trap moored at a very proximal location just off NW Africa. Friese et al. (In press) relate seasonal changes of dust particle size in sediment traps to regional meteorological variability such as precipitation, trade-wind speed and dust-storm events. In deep-sea sediments deposited offshore northwest Africa, Holz et al. (2004;2007), Mulitza et al. (2008) and Zühlsdorff et al. (2007) found links between dust deposition and variability in transport mechanisms, and more dust deposition in dry glacial periods than in humid interglacials, throughout the late Quaternary.

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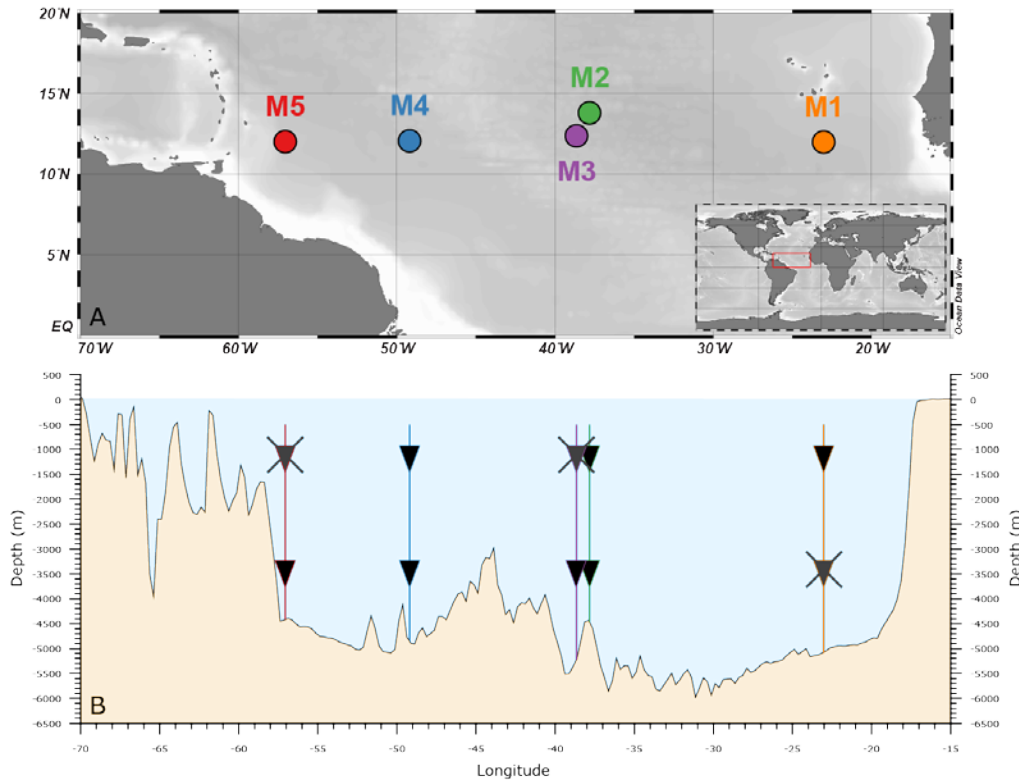
2 Material and methods

Five moorings were deployed in October 2012 (Stuut et al., 2012), of which four were moored along a transect at 12° N across the equatorial North Atlantic Ocean, and a fifth at 13° N (Fig. 1A). Each mooring is equipped with two sediment traps, at depths of 1200 and 3500 meters below sea level (BSL), or “upper” and “lower”, respectively (Fig. 1B, Table 1). The sediment traps are model PPS 5/2 from Technicap that consist of a conical funnel (36°) with a catchment area of 1 m² and an 8mm hexagonal baffle on top to maximize particle collection (U.S. GOFS, 1989) and prevent large swimmers from entering the sediment trap. Underneath the funnel, a rotating carousel with 24 sampling cups collects discrete samples of the settling particle flux. All sediment traps operated synchronously over pre-programmed intervals of 16 days. Tilt-meters showed that the sediment traps remained nearly upright for the entire sampling period. This paper presents the results of successful sampling by seven sediment traps on the five moorings from 19 October 2012 to 7 November 2013 (Stuut et al., 2013). These include three of the upper (1200 m) sediment traps located at mooring stations M1, M2 and M4, and four lower

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(3500 m) sediment traps at stations M2, M3, M4 and M5 (Fig. 1, Table 1). [Three of the ten sediment traps could not be recovered](#). In addition, seafloor sediments were collected by a Multicorer at all five mooring stations, using the top centimeter for comparison with the sediment-trap samples.



5 **Figure 1. A: Map with sampling stations M1–M5 in the Atlantic Ocean at 12° N. B: Bathymetry along 12° N (from www.gebco.net), with sediment traps at 1200m and 3500m BSL. Crossed-out sediment traps could not be recovered.**

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Station	Latitude (° N)	Longitude (° W)	Trap depths (m BSL)	Bottom depth (m BSL)	Distance to African coast (km)
M1	12.00	23.00	1150	5000	700
M2	13.81	37.82	1235, 3490	4790	2300
M3	12.39	38.63	3540	4640	2400
M4	12.06	49.19	1130, 3370	4670	3500
M5	12.02	57.04	3520	4400	4400

Table 1. Locations and depths of the sampling stations M1–M5. BSL = below sea level.

[Since both the upper and lower traps are recovered for two of the five stations \(M2 and M4\), this allows for a direct comparison between the two depths. The upper and lower sediment traps are in very good accordance with each other, demonstrated by images of the sediment-trap bottles after recovery \(Fig. 2\). Two samples, sample 12](#)

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and 24, have a much higher flux than the other samples, and these high-flux samples are present in both the upper and lower trap. Since the sampling interval is only 16 days, it means that the downward transport velocity of the sediments between the traps is at least 140 m day^{-1} and most likely much higher. It also shows that the sediments are deposited in a vertical way down to both sediment traps. It seems however that the higher flux observed in sample 12 of the upper trap is distributed over sample 12 and 13 of the lower trap. This demonstrates that there is a small time-lag between the two traps, of no more than a few days, due to the time it takes for the particles to settle. This could also be true for sample 24, however there is no sample directly after the last sample of the sediment trap.

In this paper we argue that the lithogenic particles found in the sediment traps are of aeolian origin. The sediment traps are located far from the continental shelf, so riverine input of sediments is not affecting the samples. Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll or salinity (not shown; available from Giovanni NASA GES DISC: <http://giovanni.sci.gsfc.nasa.gov/giovanni/>). In addition, the lower sediment traps are positioned 880-1300 m above the seafloor, so resuspension of bottom sediments will not affect the sediment trap samples. When considering the large amounts of Saharan dust being transported across the Atlantic Ocean every year, about 182 Tg (Yu et al., 2015), any other external input is assumed to be negligible. Stuet et al. (2005) also demonstrated the similarity between aerosol samples of Saharan dust collected off west Africa and the lithogenic fraction in sediment traps and seafloor sediments

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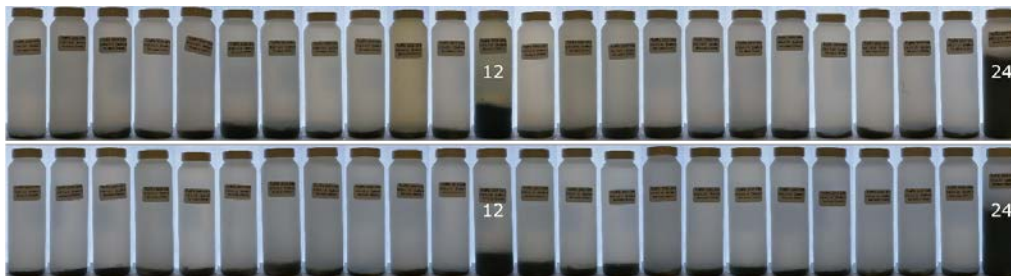


Figure 2: Sediment-trap bottles of the upper (1200 m) and lower (3500 m) traps at station M4 (12°N, 49° W).

Prior to the deployment of each sediment trap the sampling cups were filled with seawater collected at the deployment site depths, to which a biocide (HgCl_2 ; end-concentration 1.3 g L^{-1}) and a pH-buffer (borax; $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$; end concentration 1.3 g L^{-1} , $\text{pH} \approx 8.5$) were added, to a density slightly higher than the ambient seawater. In the laboratory each sample was sieved through a 1mm mesh to remove mostly zooplankton swimmers, then wet-split in five aliquots using a WSD10 Rotor splitter (McLane Laboratories, USA). The average weight difference between replicate aliquots of each sample is 2.4% ($\text{SD} = 2.2$), with 87% of all samples having a weight difference of $< 5\%$ between splits. The highest deviation was found to be 12%. For grain-size analysis, one of these aliquots was split into another 5 subsamples ($1/25$ of the original sample), that were washed and centrifuged repeatedly at approximately $1800 \times g$ with Milli-Q water to remove the HgCl_2 , borax, and sea-salts.

Biogenic constituents were removed in three steps to isolate the insoluble or lithogenic dust fraction from all samples prior to grain-size analysis, following the procedure described by McGregor et al. (2009). Shortly,

organic matter was oxidized using H₂O₂, followed by dissolving the biogenic carbonates using HCl, and removing biogenic silica by adding NaOH. Immediately prior to the grain-size measurements sodium pyrophosphate (Na₄P₂O₇ · 10H₂O) was added to ensure complete disaggregation of the particles. The particle-size distributions were measured with a Coulter Laser Diffraction Particle Sizer (LS13 320) with a Micro Liquid Module (MLM) for small-volume samples, and a magnetic stirrer was used to homogenize the sample during analysis. This resulted in particle-size distributions consisting of 92 size classes ranging from 0.375 to 2000 µm [describing the equivalent-sphere diameter of the particle](#). [Modal particle size is also expressed as particle diameter](#).

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To determine seasonal changes in dust deposition along the trans-Atlantic transect, the sediment-trap samples are grouped per season. The seasons are defined as follows: (boreal) fall includes September, October and November (SON) of 2012 and 2013, winter includes December, January and February (DJF) of 2012/2013, spring includes March, April and May (MAM) of 2013, and summer includes June, July and August (JJA) of 2013. The dates of the samples are referred to as the mid-date of each 16-day sampling period.

In order to determine the provenance of dust carrying air layers, four-day backward trajectories of air parcels were calculated with the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2015), using the GDAS (0.5 degree) meteorological dataset (<http://ready.arl.gov/HYSPLIT.php/>). The heights of these air layers were chosen in accordance with typical winter and summer dust-carrying air layers (see below) [and the starting point of the trajectories is station M1 \(12° N, 23° W\)](#).

[Data for Aerosol Optical Depth \(AOD\) and daily precipitation were obtained from the Giovanni online data system, developed and maintained by the NASA GES DISC. The AOD data were obtained from MODIS Terra, at monthly resolution and averaged over the respective seasons. Daily precipitation data from TRMM was used, averaged over the area between 11 - 13° N and 22 - 24° W \(station M1\).](#)

3 Results

3.1 Spatial trends in grain size

Modal grain sizes of the sediment traps and seafloor sediments show a pronounced downwind fining (Fig. 3). Coarsest Saharan dust was found in the easternmost trap (M1), rapidly fining westward towards M5. Also the seafloor sediments show the same clear and almost linear downwind trend of decreasing particle size (Fig. 3). However, grain sizes in the seafloor sediments are substantially finer than found in the sediment-trap samples, and the downwind decrease in grain size is also less steep for the seafloor sediments. All traps show a “shoulder” towards the coarse end of the grain-size distribution, which is most prominent at station M5 (Fig. 4A). Such shoulders are also found in the seafloor sediments (Fig. 4B), showing that coarse particles are not only deposited at proximal locations, but also transported over great distances. These include “giant” particles of [more than 100 µm](#), which are observed as far west as station M3 (38° W; approximately 2400 km from the African coast, [and thus ever further from the actual dust source](#)), and consist of both platy mica and rounded quartz particles (Fig. 5). Furthermore, the average grain-size distributions show that the differences between stations are larger than

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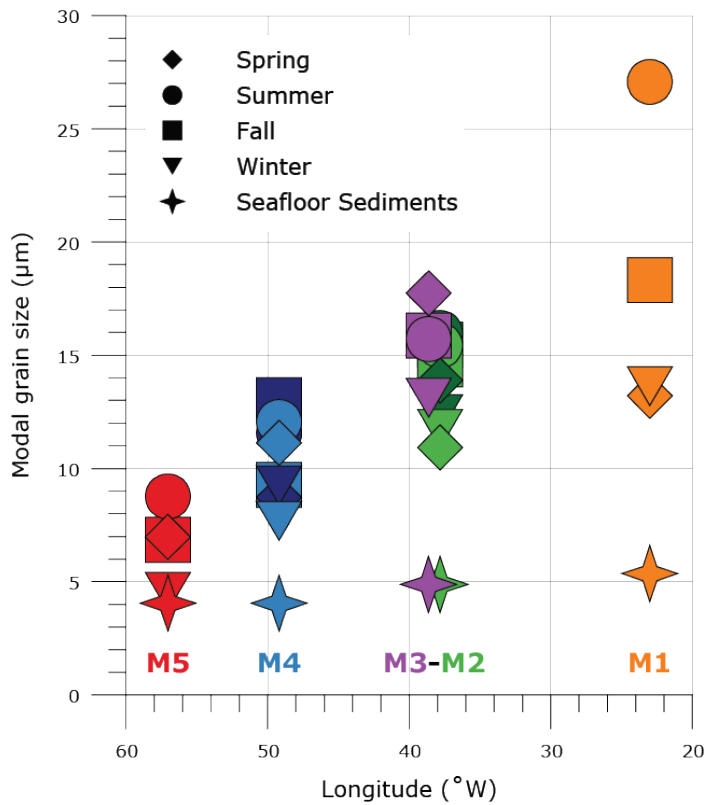
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between the upper (1200 m) and lower (3500 m) traps at stations M2 and M4 (Fig. 4A). The grain-size distributions of the seafloor sediments show that the dust at station M1 is the least sorted, meaning that the widest range of particles of different sizes is deposited closest to the source (Fig. 4B).

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5 Figure 3. Downwind fining and seasonality in average modal grain size per season for all seven traps, and modal grain size of the seafloor sediments, versus western longitude, for October 2012 –November 2013. For M2 (green) and M4 (blue), lighter colors indicate the upper (1200 m) trap, and darker colors indicate the lower (3500 m) trap.

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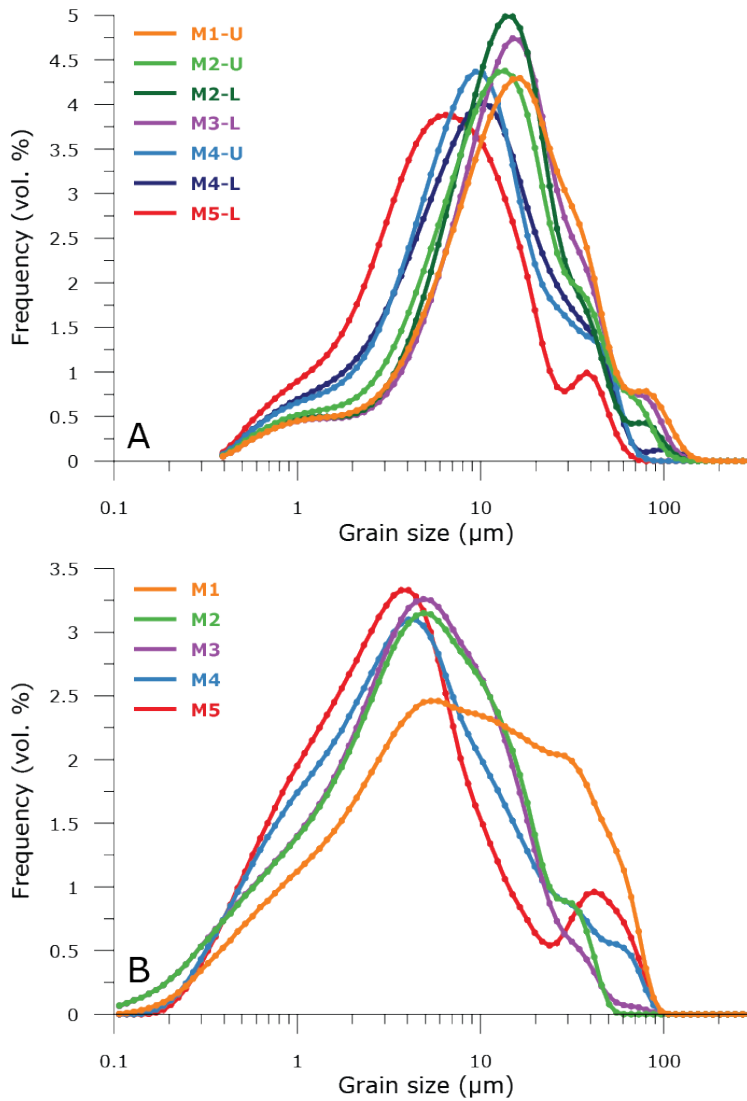


Figure 4. A: Average grain-size distributions of all seven sediment traps, representing the average of 24 samples, where U = upper trap (1200 m) and L = lower trap (3500 m). Collected between October 2012 and November 2013. B: Grain-size distributions of seafloor sediments at the five mooring stations (M1–M5) along the trans-Atlantic transect.

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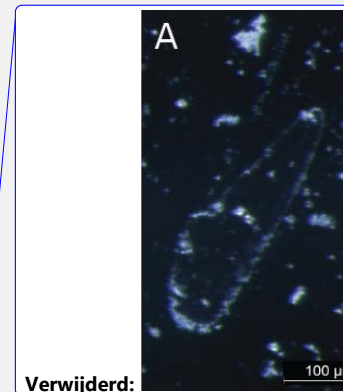
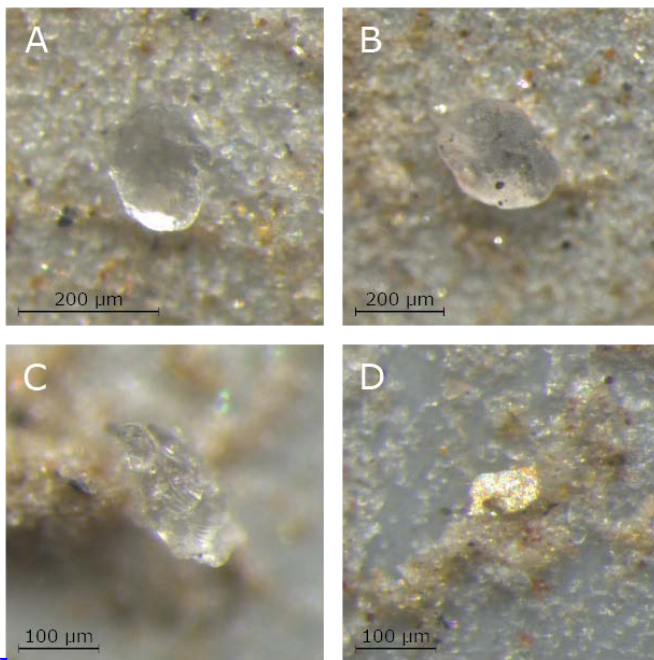


Figure 5. Light-microscope images of large dust particles from the lower (3500 m) traps at station M2 (13° N, 37° W; A and B) and at station M3 (12° N, 38° W; C and D). Both stations are situated at more than 2000 km from the African source. A: Large quartz particle (diameter approximately 180 μm over long axis) from sample 1 (October 19 – November 4, 2012). B: Large quartz particle (diameter approximately 290 μm over long axis) from sample 1. C: Large quartz particle (diameter approximately 200 μm over long axis) from sample 4 (December 6 – 22, 2012). D: Large mica particle (diameter approximately 86 μm over long axis) from sample 4.

3.2 Seasonal grain-size trends

The particle size of Saharan dust deposited in the Atlantic Ocean changes seasonally, and is clearly coarser in summer than in winter at station M1 (Fig. 6A). During spring, a coarse shoulder is present in the grain-size distributions (Fig. 6B), which is more prominent than during the other seasons. Also modal grain sizes illustrate this seasonality, varying between 12.5 and 15 μm from October 2012 to May 2013 (fall to spring), followed by a sharp increase to about 30 μm in June 2013, and stays coarse for the entire summer season (Fig. 7) at station M1. Grain sizes decrease again in late August, and keep decreasing throughout the fall of 2013. At M2 the modal particle size of the upper trap decreases from fall to winter, from 15 μm to about 10 μm, followed by an increase to around 15 μm in May, continuing into summer and fall 2013. At M4 particle sizes of the upper trap decrease from 10 μm in fall 2012 to 7 μm in mid-spring 2013, after which they increase to around 12.5 μm throughout summer and fall 2013.

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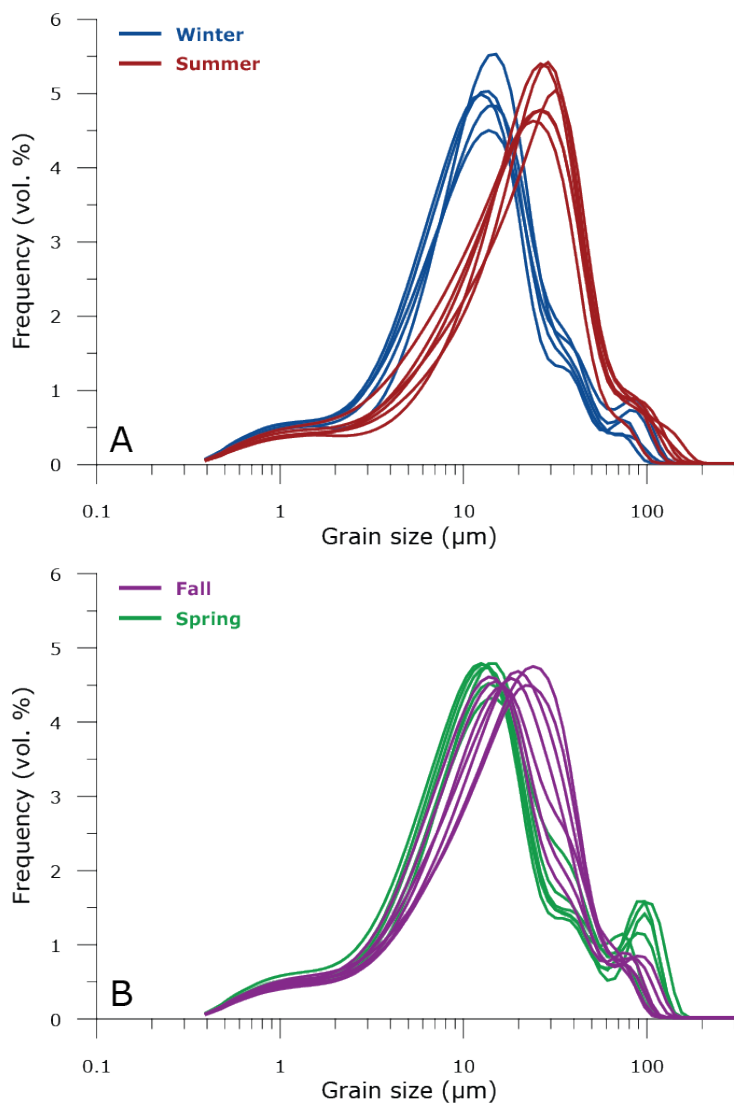


Figure 6. Seasonal grain-size distributions at station M1 (12° N, 23° W; approximately 700 km from the African coast) for October 2012–November 2013, of A: winter (blue), summer (red), B: fall (purple) and spring (green).

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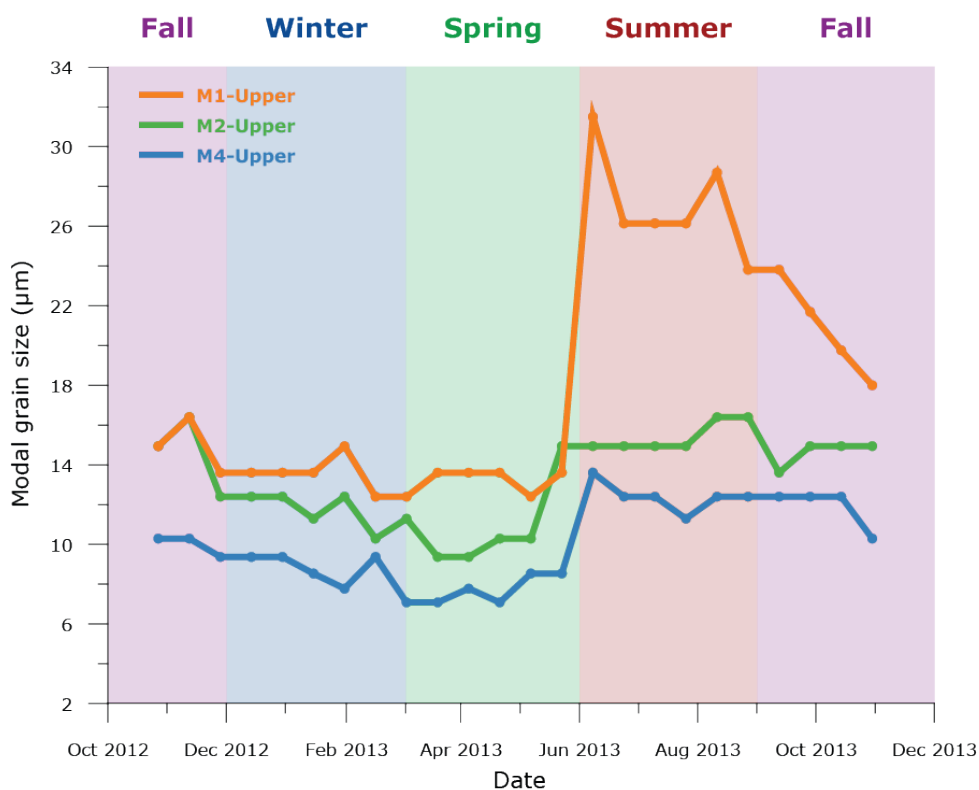


Figure 7. Modal particle diameter of dust collected by the three upper (1200m) sediment traps at stations M1, M2 and M4, from October 2012–November 2013.

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Overall, the particle size at the three sites show the same seasonality, with coarser dust in summer and fall and finer dust in winter and spring (Fig. 7). However, the difference in particle size between these seasons is greatest at M1, close to the source (Fig. 3). Here, particles are also least sorted and have the widest range in particle size, which gradually decreases westward towards M5. However, seasonal trends in modal grain size are more pronounced in the three upper traps at 1200 m than in the four lower sediment traps at 3500 m (Fig. 8). In the lower traps, the modal particle size at the more northern station M2 is slightly finer than at the more southern station M3 from fall 2012 to spring 2013, with the exception of two samples that show unusually high modal grain sizes (in November 2012 and April 2013, shown as “outliers” in Fig. 8). From summer 2013 onwards, the modal grain size of M2 and M3 converge, with synchronous fluctuations between 14 and 18 µm. Seasonality at M4 is even weaker, with grain sizes varying between 5 and 18 µm. At the westernmost station M5 modal particle size ranges between 4 and 10 µm, with a decrease in spring 2013 and an increase in summer. In all seven traps, dust is finest during spring. When comparing modal grain sizes found in the upper (1200 m) and lower (3500 m) traps from stations M2 and M4, it shows that the lower sediment traps have slightly coarser dust than the upper traps (Fig. 9).

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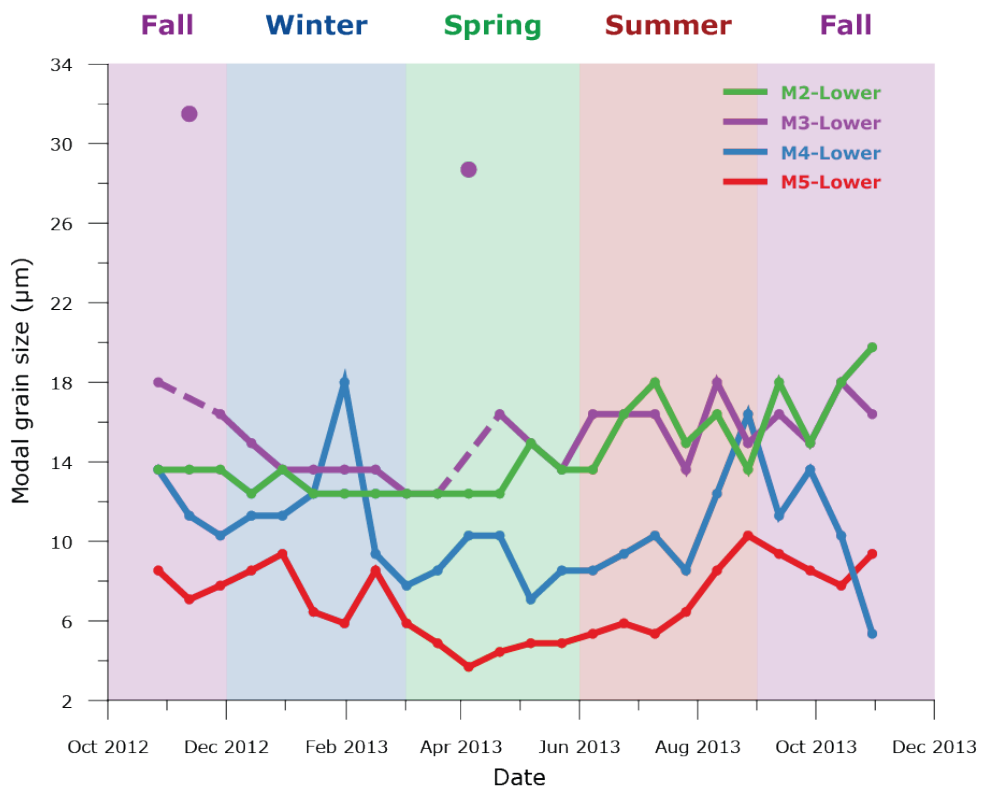


Figure 8. Modal particle diameter of dust samples from the four lower (3500m) sediment traps at stations M2, M3, M4 and M5, for October 2012–November 2013. The two points that are not connected in series M3-Lower are considered outliers.

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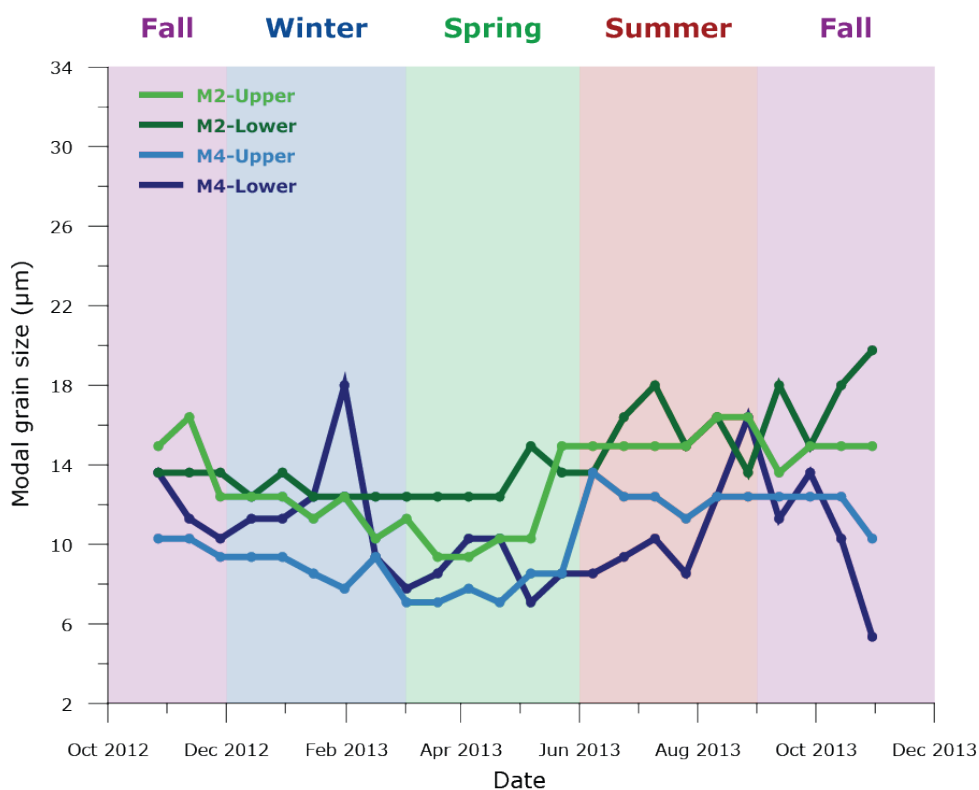


Figure 9. Modal particle diameter of dust samples from the upper (1200 m) and lower (3500m) sediment traps at stations M2 and M4, for October 2012–November 2013.

4 Discussion

5 The grain size of dust decreases with increased distance from the source (Glaccum and Prospero, 1980;Goudie and Middleton, 2006;Mahowald et al., 2014;Stuut et al., 2005): coarse particles have a higher settling velocity and smaller particles can be transported over greater distances (Gillette, 1979;Tsoar and Pye, 1987). This mechanism accounts for the downwind fining observed in both the sediment traps and the seafloor sediments along the trans-Atlantic transect (Fig. 3). However, we observed giant particles ($\geq 100 \mu\text{m}$) at station M3 (38°W ;

10 approximately 2400 km from the African coast) (Fig. 5), and also mica particles, whose platy shape allows for aerial transportation over greater distances (Stuut et al., 2005). Such coarse particles are generally not incorporated into climate models (Kok, 2011). Only a handful of these coarse particles are found in the samples, however when considering these are 1/25 splits of the original samples, collecting sediments over only 1 m^2 of ocean, over a time period of only 16 days, this means that the amount of giant particles being transported over the Atlantic Ocean is substantial. This underestimation of the coarse size fraction may have its origin in the

15 sampling of dust of specific size classes, e.g. PM_{10} and $\text{PM}_{2.5}$, which form the basis of the guidelines from the World Health Organization (WHO, 2006) on fine-grained particles.

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Omlaag verplaatst [1]: Since the seafloor sediments represent a longer time average of Saharan dust deposition than the sediment-trap samples, it implies that the downwind fining is a long-lived trend.

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Verwijderd: Indeed, the coarse particles causing the secondary peak in the grain-size distributions appear to increase downwind from M3 to M5 (Fig. 3), confirming that the coarse but platy mica particles are preferentially transported over greater distances.

5 Since the seafloor sediments represent a longer time average of Saharan dust deposition than the sediment-trap samples, it implies that the downwind fining is a long-lived trend. However, the modal particle size of the
sediment-trap samples is substantially coarser than that of the seafloor sediments at the same stations along the
transect. The particle-size distribution found in the sediment-trap samples closely resemble Saharan dust
10 sampled directly from the atmosphere, by shipboard dust samplers along a transect off the West African coast,
which has modal grain sizes varying between 8 and 42 μm (Stuut et al., 2005). This is in close resemblance with
the observed modal grain size of 4 – 32 μm in the sediment traps. By contrast, modal grain sizes in the
underlying seafloor sediments range between 4 and 5.5 μm . Since the seafloor sediments represent a longer time
15 period, this suggests that Saharan dust was significantly finer in the recent past and increased over the last
centuries. Deposition of coarser dust is in line with increased emission as a result of human activity since the
nineteenth century due to commercial agriculture (Mulitza et al., 2010). Not only does increased human activity
in the source region increase dust emissions, it also enables larger particles to be emitted (McTainsh et al., 1997),
20 which could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we
see now in the sediment traps.

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15 The seasonal variability in particle size can be the result of several factors. First, it could result from the seasonal
movement of the dust cloud, associated with the latitudinal movement of the ITCZ (Nicholson, 2000). As a
result, in summer dust is transported at more northern latitudes than in winter, as indicated by the aerosol optical
20 depth (AOD) data (Fig. 10). These aerosols can include sea salts, organic and black carbon, sulfates and mineral
dust. However, the aerosols over our study area are mostly mineral dust originating from the African continent
(Yu et al., 2015). In summer, when AOD values are highest, the cloud is located at its northernmost position
(Fig. 10D). Aerosol concentrations are lowest during fall (Fig. 10A and -E), and during winter the cloud is
located in its southernmost position (Fig. 10B). However, during winter the aerosols may receive a higher
25 contribution from soot by bushfires released more to the south (as also visible during the other seasons), thereby
moving this high-AOD cloud southward and possibly falsely implying the latitudinal movement of the dust
cloud.

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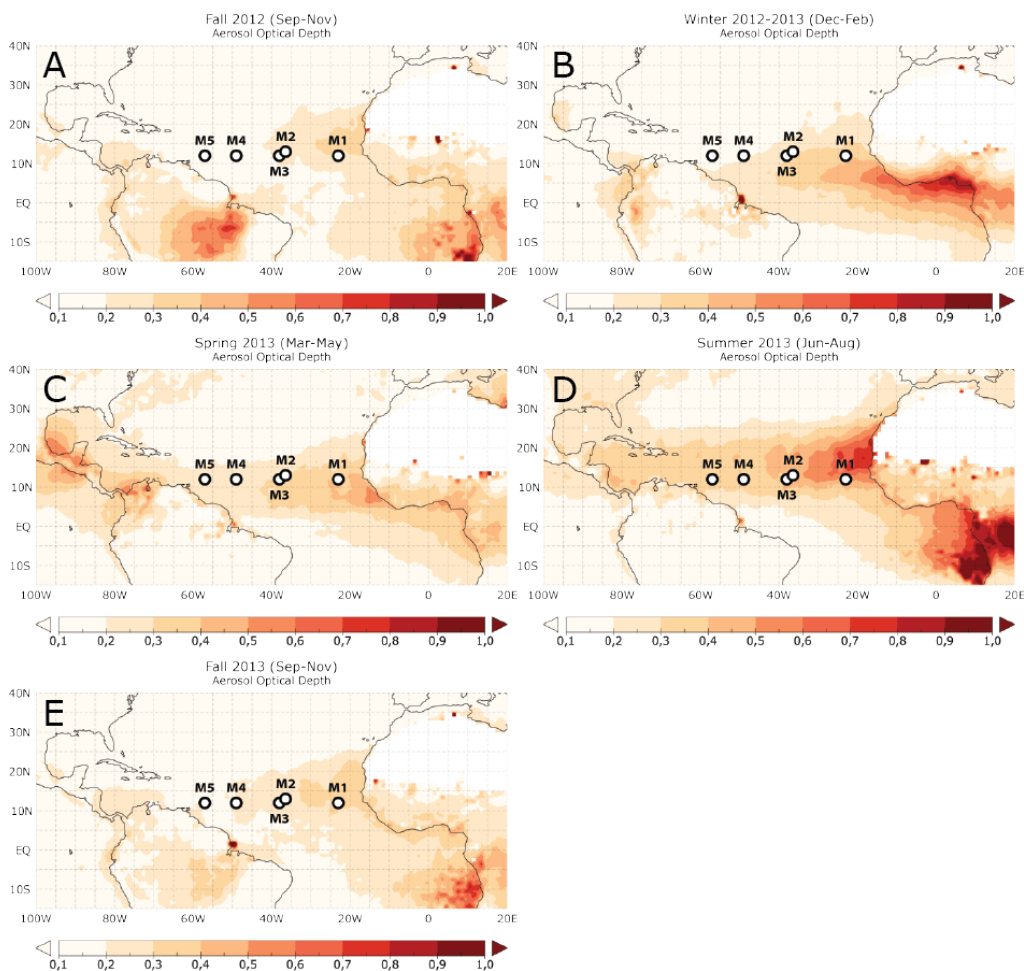
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30 This seasonal, latitudinal shift of the dust cloud is reflected in the samples from stations M2 and M3, which are
positioned at one degree northern latitude from each other. During winter, modal grain sizes at the northern
station (M2, 13° N) are finer than at the southern station (M3, 12° N), while similar at both stations during
summer (Fig. 8). Thus, during winter the northern station M2 does not receive the same dust as station M3, since
35 the dust cloud is located more to the south. In summer the dust cloud is located more to the north, delivering
coarser particles and at the latitude of both stations. However, the difference in grain size between the two traps
is small, due to the close proximity of the two stations (about 200 km). In addition, the seasonal shift of the ITCZ
also causes a latitudinal shift of the seasonal rain belt, affecting different sources during the year (Nicholson,
2000) and changing the amount and location of wet deposition. An alternative explanation is provided by
different wind systems that are active throughout the year, along different trajectories and at different wind
speeds. These can entrain dust from different source areas. The elevation of these wind systems, in combination
with wind speeds and the particle size of the source soils, determine the particle-size distributions of the

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entrained dust (Marticorena, 2014; Tsoar and Pye, 1987), and are further influenced during transport and deposition, creating different grain-size signatures for summer and winter dust.



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Figure 10. Three-month average aerosol optical depth (AOD) for the sampled seasons, from MODIS Terra, A: Fall (SON) 2012, B: Winter 2012–2013 (DJF), C: Spring 2013 (MAM), D: Summer 2013 (JJA) and E: Fall 2013 (SON). Stations M1–M5 are marked with black/white circles.

In winter, dust is transported at lower altitudes than during summer. This is evidenced by satellite images of the Cape Verde islands, which show the high mountain tops (highest point is Fogo at 2829 m) piercing through the dust cloud, deflecting it around the islands (Fig. 11A). The lowest peak that is still visible above the dust cloud is Brava (976 m), but the top of São Vicente (750 m) is not. This means that the top of the dust cloud is at an elevation between 976 and 750 m. In summer, dust is transported at much higher altitudes than winter, covering the Cape Verde islands in a thick blanket of dust (Fig. 11B), meaning that the top of the cloud is at an elevation of at least 2829 m. During summer, dust is transported in the high-altitude Saharan Air Layer (SAL) (Carlson and Prospero, 1972; Kanitz et al., 2014; Prospero and Carlson, 1972; Tsamalis et al., 2013). Mahowald et al.

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Verwijderd: Maps visualized with the Giovanni online data system, NASA GES DISC.

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(2014) argue that the dust particle size does not depend on wind speeds at emission. However, high wind velocities in the SAL of $>7 \text{ ms}^{-1}$ (Tsamalis et al., 2013), enables coarser dust particles to remain in suspension in summer, and due to the high altitude these coarse particles are transported over great distances. In addition, increased convection in the source areas in summer, related to larger differences in temperature, can result in the uplift of coarser dust particles (Heinold et al., 2013).

Four-day backward trajectories of air parcels also illustrate the difference in the elevation of the dust-transporting air layers between winter and summer (Fig. 11C and -D). The altitudes of the starting points of these backward trajectories were chosen in accordance with the hypothesized heights of the dust-carrying air layers, as demonstrated in Figures 11A and -B, with the lowest (500 m) elevation representing winter dust transport and the highest (3500 m) elevation representing summer dust. In winter (Fig. 11C), the higher trajectory is not originating from the African continent, and therefore the winds at these altitude are unlikely to transport dust to the sample location. The lower trajectory has a more eastern origin, and air layers at this altitude could be transporting dust (Fig. 11A), picked up from the surface and brought to higher altitudes. By contrast, in summer (Fig. 11D) this situation is reversed: the higher trajectory has a more continental origin and is the most likely dust-carrying air layer over the lower trajectory. The elevation profile shows that this high-elevation trajectory started at lower altitudes, but upon reaching the coastline it was uplifted to about 3500 m AGL (Fig. 11D, bottom panel). This is in accordance with how the Saharan Air Layer (SAL) is described, when dust-carrying air from the continent is uplifted by a cool marine inversion layer (Carlson and Prospero, 1972; Prospero and Carlson, 1972). This inverted air layer is visible in the 500 m air layer, moving in an opposite direction, from west to east. After this sharp increase in altitude, the trajectory decreases in altitude, which persists across the Atlantic Ocean (Tsamalis et al., 2013).

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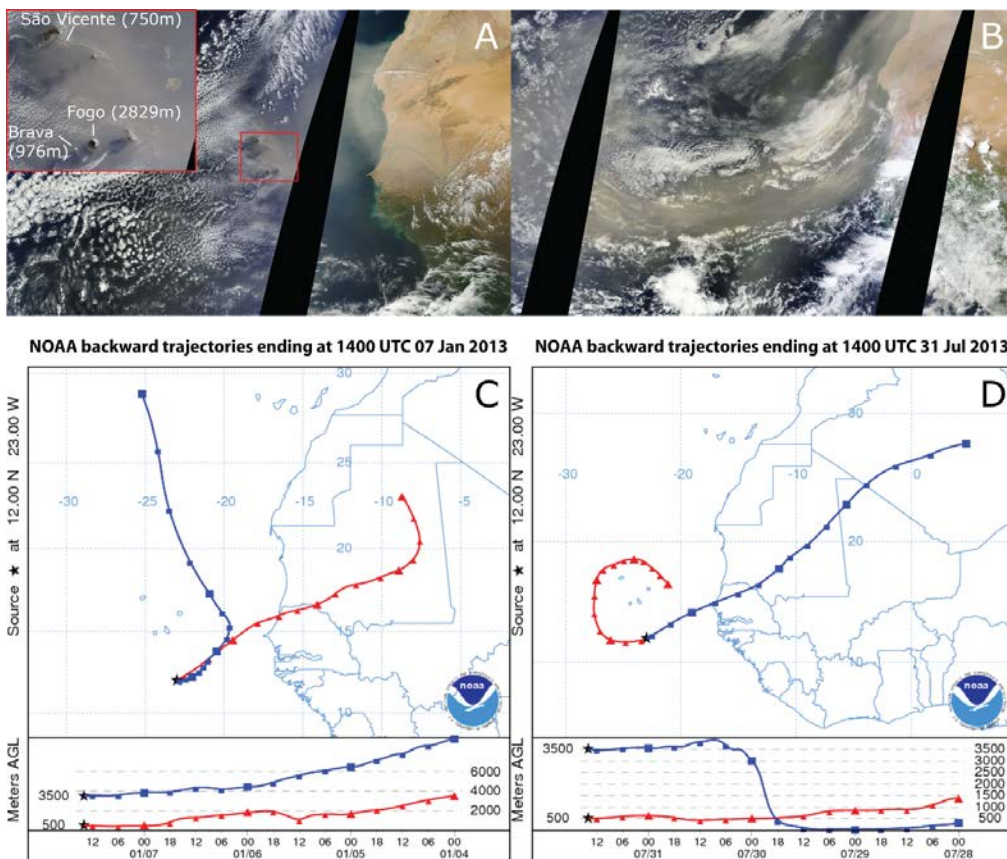


Figure 11. A: Satellite images of typical winter dust transport, with close-up of the Cape Verde islands (7 January 2013) and B: typical summer dust transport (31 July 2013) over the Cape Verde islands, at relatively low and high altitudes, respectively. Images from NASA Worldview, MODIS Terra satellite. Black areas are artefacts from satellite passage. C & D: Concurrent four-day backward trajectories of air parcels from station M1 (star), at 500 m (red) and 3500 m (blue) AGL, showing trajectory maps (top) and elevation profiles (bottom). C: ending at 7 January 2013, D: ending at 31 July 2013.

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The summer season is also characterized by an increased number of more intense dust storms (e.g. Adams et al. (2012)). From May to September, dust is almost continuously emitted from the African continent, as shown by satellite images (MODIS Terra and Aqua satellites; NASA Worldview). Within five days, the dust cloud propagates towards the Caribbean and becomes progressively thinner by dust deposition in the Atlantic Ocean along its track. Increased deposition of coarse particles can also be caused by increased precipitation in summer and fall, as opposed to almost no precipitation in winter and spring (Fig. 12). This was also noted off northwest Africa related to wet deposition by Friese et al. (In press). Increased precipitation at station M1 seems to coincide with increased modal grain sizes, and this relation commences with lowest precipitation early June 2013. This suggests that little precipitation is already sufficient to wash out the suspended dust from the atmosphere by wet deposition.

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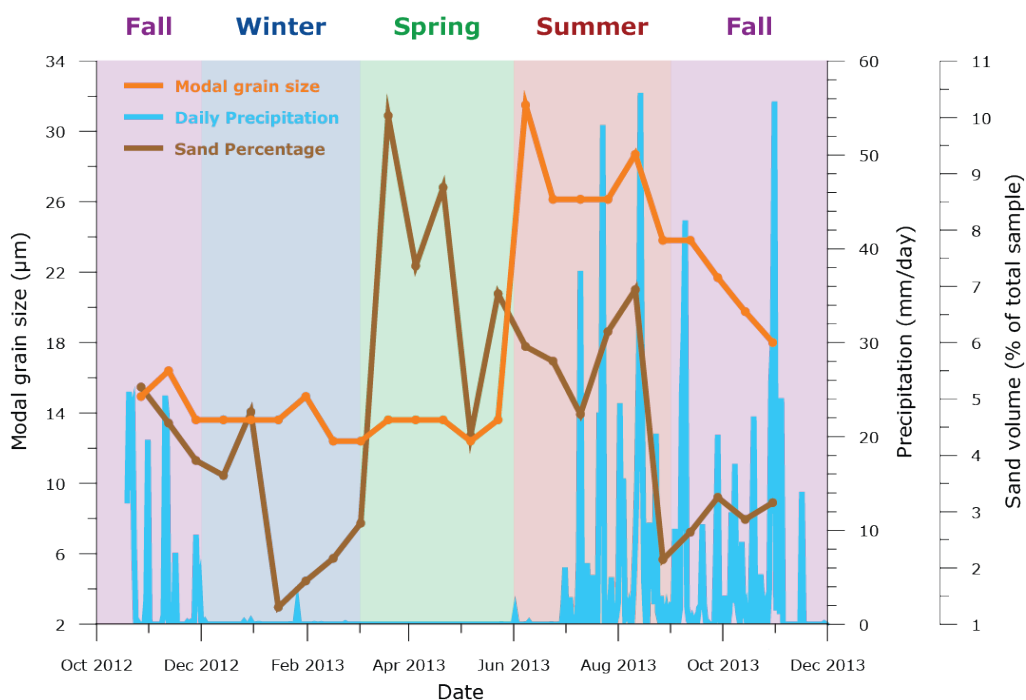


Figure 12. Modal grain diameter (left axis), daily precipitation (right axis; from Giovanni online data system, NASA GES DISC) and percentage of sand particles (> 63 µm) (far right axis) at station M1 (12° N, 23° W).

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At M1, the percentage of sand-sized particles (> 63 µm) increases sharply in spring while modal grain sizes increase in summer (Fig. 12). This increase in coarse particles is related to coarse shoulders in the grain-size distributions of the spring samples (Fig. 6B) that are absent or less prominent in fall, winter and summer (Fig. 6A and -B). These coarse particles mostly include micas: due to their platy shape, these particles have a different aerodynamical behavior than more spherical quartz particles and are therefore more easily transported by wind than spherical particles with a similar diameter (Stuut et al., 2005). However, also large ($\geq 100 \mu\text{m}$) more spherical particles were observed in the samples, at very large distances from the source (Fig. 5). These coarse particles, visible in the grain-size distributions as coarse shoulders, are found in all the traps at all stations, and appear most frequent during spring. An increased number of coarse particles during spring could mean that the dust could originate from a different source area. However, backward trajectories calculated over the entire sampling period do not suggest this.

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The lower (3500 m) traps show less seasonality and are generally slightly coarser than the upper (1200 m) traps. This may be due to the disaggregation of marine snow, releasing the individual dust particles and thus decreasing their settling velocity. Therefore, it would take longer for particles to reach the lower traps at 3500 m, especially very fine particles, and as a result the particle-size distributions lose their seasonal characteristics. This would also explain why the dust in the lower traps (at M2 and M4) is slightly coarser than their upper counterparts, since these coarse particles settle more quickly, and the very fine particles may not reach the lower traps.

5 Conclusions

We have shown seasonal and spatial changes in Saharan mineral dust transport and deposition across the Atlantic Ocean by means of sediment-trap sampling between October 2012 and November 2013, and [spatial changes in the](#) seafloor sediments at the same stations. Our results show strong seasonal variations and significant fining in particle size with increasing distance from the source [in the sediment trap samples](#), with modal [particle diameters](#) ranging from 4 to 32 μm . Coarser dust [is found in the sediment traps](#) opposed to the seafloor sediments, [in line with increased emission and](#) coarser dust due to the onset of commercial agriculture in the 19th century. [A down-wind decreasing particle size](#) reflects the greater gravitational settling velocity of coarse particles, resulting in deposition closer to the source. The largest seasonal difference in particle size occurs closest to the source, however the lower sediment traps (3500 m) show less seasonality than the upper sediment traps (1200 m). This may be due to marine snow disaggregating, decreasing the settling velocity of individual dust particles, resulting in a decreased expression of the seasonal particle-size signatures. Coarser grain sizes during summer and finer during winter and spring suggest: (1) summer transport at higher elevations of up to 5 km within the Saharan Air Layer at high wind speeds ($> 7 \text{ ms}^{-1}$), compared to winter transport; (2) coupling to the latitudinal movement of the dust cloud with the ITCZ; (3) increased emission by more frequent dust storms in summer combined with wet deposition by increased precipitation. Increased contribution of coarse ($> 63 \mu\text{m}$) particles in spring is likely caused by large platy minerals (e.g. micas) of small aerodynamic size that are easily uplifted and transported, possibly from a different source area. These coarse particles are transported thousands of kilometers away from the Saharan source. Multiple-year samples from this transect should clarify which of the above mentioned processes are more dominant, in order to be applied in e.g. climate models and climate reconstructions. Our results contribute to a better understanding of the seasonal and spatial variability of Saharan dust, which still remains a poorly constrained factor in global climate.

Acknowledgements

We thank the captain, crew and scientists of RV *Meteor* cruise M89 and RV *Pelagia* cruise 64PE378, for deployment and retrieval of the sediment-trap moorings. Funding is provided by NWO [with the TRAFFIC](#) project 822.01.008, as well as by the ERC with starting grant 311152: DUSTTRAFFIC. [We thank three anonymous reviewers whose comments greatly improved the paper.](#)

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Gewijzigde veldcode

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