Dear Dr. Schwarz and reviewers,

Thank you once again for your time on reviewing our manuscript. We carefully considered all the comments and revised the manuscript accordingly. In this document, the questions of the editor are answered in red, for reviewer 2 in green and for reviewer 3 in blue. Please find attached the revised manuscript, with tracked-changes with respect to the original manuscript.

Editor comment
Dear Dr. Van der Does and co-authors -

Thank you for your detailed response to the reviewers, and your improved manuscript. As you point out in the response, this manuscript represents a marriage across disciplines. Given that ACP is focused on the atmospheric issues addressed here, my sense is that a little bit more concession to the target community is called for.

For example, the widely raised question from the reviewers about potential biological sources of the lithogenic particles; I imagine that they were thinking (as I did) of the marine organisms that contribute their shells to limestone in the form of calcium carbonate. Could you more directly address this issue?

As we have tried to explain in the methods section of the paper, all the biogenic particles of marine origin are chemically removed: this includes all organic matter, (biogenic) carbonates and biogenic silica. What is left is the insoluble or lithogenic fraction, which is only sourced in atmospheric mineral dust. This may potentially include a small fraction of volcanogenic and cosmogenic particles as well (Plane, 2012), however on the other hand, carbonate dust particles and organic particles of non-marine origin are also removed from the samples by the chemical treatment applied. Since this is the case for every sample that we analyzed in this study, they can be compared directly to each other. In addition, we know that most of Saharan dust is made up of siliciclastics (e.g. Scheuvens and Kandler, 2014), which are not affected by this chemical treatment. Again, we would like to stress the fact that all marine particles (including foraminifera, radiolarians, diatoms, coccolithophores, etc.) have been removed from the samples prior to grain-size analysis. To verify if indeed all marine biogenic particles were removed, we performed microscope analysis (Figure 5). This confirmed that the fraction analyzed for particle size is the insoluble or lithogenic fraction, interpreted as mineral dust.

We modified the paragraph from the methods section addressing this issue.

Page 6, lines 7-17:

"The sediment traps collect all particles settling down into the ocean, that besides mineral dust includes the skeletons of marine plankton (foraminifera, radiolarians, diatoms, coccolithophores, etc.), organic matter (marine and aerosols from biomass burning) and potentially volcanogenic and cosmogenic particles (Plane, 2012). All these biogenic constituents were chemically 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 H_2O_2, followed by dissolving the biogenic carbonates using HCl, and removing biogenic silica by adding NaOH. What remains is the lithogenic fraction which is considered to consist mainly of dust, as confirmed by microscope analysis (Fig. 5). Indeed, some of the dust particles have a risk of being removed during this process including lithogenic carbonates and organic particles of non-marine origin. However, lithogenic carbonates are more resistant to the chemical treatment than the biogenic carbonates. Also, since this is the case for every sample analyzed, they can be compared directly to each other."

Other basic questions are still focused on the transport of dust from the surface downwards. Simple calculations of particle sedimentation rates (in the absence of any water currents) indicates that the short transport time between the 1200 m and 3500 m traps cannot be explained simply by particle setting. For example, for a 10 µm particle of density 2700 kg/m^3, a distance of only ~200 m would be traversed in a ~few weeks. Since 10 µm is close to the mass-modal diameter, it's clear that something else must be going on. Hence the interest in currents, I imagine, which the atmospheric community does not have any intuition about. Adding information about the rate and direction of currents will be very helpful. This is also relevant to the apparent discontinuity in results at

the lower traps and at the ocean floor; why would transport be very speedy from 1200 m to 3000m, but then essentially disappear in the lowest 1 - 2 km?

In the ocean, the dust particles deposited onto the ocean's surface do not settle individually, but as part of large marine particles like marine snow. These are aggregates > 500 µm of organic and inorganic particles of different composition and origin (e.g. Nowald et al., 2015). These particles have a much higher settling velocity, usually over 200 m day^{-1}, and the settling velocity may even increase with increased depth (Berelson, 2002;Nowald et al., 2015).

We modified the following lines in the revised manuscript:

Page 5, lines 6-10:

"These high settling velocities can be reached since the mineral dust particles are not deposited individually, but as part of large marine particles like marine snow. These are aggregates > 500 µm of organic and inorganic particles of different composition and origin (e.g. Nowald et al., 2015). These particles have settling velocities of approximately 200 m day^{-1}, and may increase with increased depth (Berelson, 2002;Nowald et al., 2015)."

In addition, current meters on the moorings show that, with few exceptions, current velocities remained well below < 12 cm s^{-1}, which is the threshold below which unbiased collection of settling particles occurs (Knauer and Asper, 1989). Above this threshold undertrapping occurs, with high current velocities resulting in a decreased particle flux in the sediment traps.

We've added the following lines to the revised manuscript:

Page 3, lines 36-39:

"Tilt-meters showed that the sediment traps remained nearly upright for the entire sampling period. With few exceptions, current velocities, as measured by current meters and ADCPs, remained well below 12 cm s^{-1}, the threshold below which unbiased collection of settling particles occur (Knauer and Asper, 1989)."

The apparent discontinuity in particle size between the sediment traps and the seafloor sediments is due to the difference in timing, and is not related to particle settling velocities: the sediment traps collect currently deposited dust over periods of 16 days, while the seafloor sediments contain dust deposited over 100s to 1000s of years, in a single sample. This is also extensively discussed in the manuscript.

Addressing these questions will help the atmospheric community contextualize the potential scale of any assumptions and uncertainties, and will help it grasp the significance of this manuscript as a step in achieving closure between atmospheric transport of mineral dust, and its removal from the atmosphere by dry and wet deposition.

As I had some concern that some portions of the reviewers' comments may not have been fully considered (these are the questions I reformed here; perhaps I am mistaken!), I have asked them, also, to comment on the response.

Thank you very much for working through this review process with ACPD; I have high expectations for the value of your technique and findings to contribute meaningfully to understanding and quantifying total and size-dependent dust transport and removal from the atmosphere.

PS - Minor additional comments:

It would be helpful to use clearer wording to describe the size distribution information throughout the paper. I found myself repeatedly questioning whether number or mass average values were being presented. For example, in the caption of Figure 3, the "average modal grain size" is presented. I think this is the average modal-mass grain size ? ; in Figure 4 the caption is for "average grain-size distributions" , but these represent average grain-volume distributions? Page 19 ,lines 3-7 : "...with modal particle diameters ranging..." could perhaps be more clearly stated "...with mass-modal particle diameters ranging..."? And so on.

The modal particle sizes described in the paper are neither number nor mass average values, but the modal values of the relative volume grain-size distributions. Average modal grain size means the average over several samples, in the case of Figure 3 grouped seasonally.

We modified the following lines in the revised manuscript:

Page 8, lines 2-5:

"Figure 3. Downwind fining and seasonality in average modal grain size per season for all seven traps (an average of all modal values from the relative volume grain-size distributions, grouped per season), for October 2012 – November 2013, and modal grain size of the seafloor sediments (from the grain-size distributions as show in Figure 4B), versus western longitude."

Page 9, lines 3-4:

"Figure 4. A: Average volume grain-size distributions of 24 samples from all seven sediment traps, where U = upper trap (1200 m) and L = lower trap (3500 m)."

On Figure 4, I'd also suggest a clarification of the vertical axis legend. As it looks like the width of the bins of the histogram are not one size in linear space, but one size in log-space, it is standard practice in the atmospheric community to specify the volume fraction as scaled by the log-space bin width (dV/dLog(D), the differential volume per horizontal step in log space). I imagine that this is what is shown already.

We have changed the title of the axis in Figures 4 and 6 (pages 9 and 11), and it now reads: "Frequency (vol. %) ; dV/dLog(D)".

Finally, my own elementary question reflecting lack of familiarity with your water-centric techniques: are the sampling volumes closed when not sampling/during recovery? Is there any chance that some vials sampled substantially longer times than others? If these are non-issues, that will be helpful to know.

The (24) sampling bottles of the sediment traps are mounted on a carrousel underneath the funnel of the trap. This carrousel has a zero-position, which is positioned underneath the funnel during deployment. The motor driving the carrousel is programmed to start sampling at a pre-defined date, such that all sediment traps along the whole transect start sampling simultaneously. All other bottles are sealed underneath the carrousel with O-rings. After the pre-programmed sampling interval, the carrousel switches one position and the next bottle is placed underneath the open funnel. After all bottles have sampled, the carrousel returns to the zero position until the sediment trap is recollected. The sediment-trap motor logs its actions, and from this data we can see that they performed flawlessly according to the pre-programmed intervals, for all seven sediment traps. This sediment-trap sampling method has been used in the marine community for several decades and proved efficient in many studies. Since no extraordinary events occurred, the precise mechanism of the sediment traps are not mentioned in the manuscript.

We modified the following lines in the revised manuscript:

Page 3, lines 35-36:

"All sediment traps operated synchronously over pre-programmed intervals of 16 days, and performed flawlessly."

Co-Editor Decision: Reconsider after major revisions (18 Aug 2016) by Joshua Schwarz
Comments to the Author:
Dear Dr. van der Does,

After having some time to consider the reviewers response to your revised manuscript, I have concluded that the required changes rise to the level of a major revision. In particular, both reviewers felt that the revised text should incorporate more of your responses. Indeed, I have always felt that manuscripts published in ACP should stand on their own without any need for readers to reference discussions in ACPD.

You can be very pleased that the reviewers also still agree on their assessment of the excellent promise of the scientific significance of your work. I very much look forward to another revision of your exciting manuscript! I

Regards,
Shuka

Dr. Joshua (Shuka) Schwarz
Guest Editor, ACP Special Edition: SALTRACE
NOAA ESRL CSD
Boulder, CO

Referee # 2

The authors provided a revised version of the manuscript and point-to-point responses to the referees' comments. The new manuscript is indeed improved, and the authors answered satisfactorily to most of my previous comments. I think that some of those replies should also be incorporated more directly into the manuscript, to enhance the comprehension for all future readers. Here are my final (minor) comments.

3 / 9. "We used time-series FROM submarine sediment traps ..."
3 / 10. "of 16 days. Here we ..."

We modified the following lines in the revised manuscript:

Page 3, lines 9-10:

"We used time-series from submarine sediment traps moored at five locations along this transect, sampling synchronously over successive intervals of 16 days."

3 / 25-27. Not completely accurate (e.g. McGee et al., 2013)

The reviewer is correct that McGee et al. (2013) observed dust fluctuations at a higher frequency than "just" glacial-interglacial scales during the last 20kyr BP, however we wanted to sketch the bigger picture of the Late Quaternary. We now added the words "generally, throughout the Late Quaternary" to emphasize that we are really merely sketching the bigger picture throughout the whole Quaternary.

We modified the following lines in the revised manuscript:

Page 3, lines 24-27:

"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 generally, more dust deposition in dry glacial periods than in humid interglacials, throughout the Late Quaternary."

6 / 33-35. How are these two observations (one about a vertical profile, the other about a longitudinal gradient) connected?

We agree that this is an inconsistent sentence, with two sentences that are not linked.

We modified the following lines in the revised manuscript:

Page 7, lines 17-23:

"All traps show a "shoulder" towards the coarse end of the grain-size distribution, which is most prominent at station M5 (Fig. 4A). This shows that coarse particles are not only deposited at proximal locations, but also transported over great distances. These shoulders are also found in the seafloor sediments (Fig. 4B). They include "giant" particles of more than 100 µm, which are observed as far west as station M5 (57° W; approximately 4400 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)."

9 / 10. I would suggest to explain already here the size distribution metrics that will be shown and discussed. For instance, explain that the modal diameter of particles-volume distributions was chosen to focus on the "main" dust mode, and that this particular metric does not account for the presence of shoulders. On the other hand you will show a complementary metric, i.e. sand%, to discuss the presence of shoulders.

We added a paragraph that discusses the used metrics at the beginning of the Results section.

Page 7, lines 7-11:

"The particle size of Saharan dust can be expressed in many ways. Showing grain-size distributions of all individual samples makes it more difficult to distinguish seasonal and spatial changes. Therefore, the modal particle size is chosen to represent the main dust mode, to better illustrate these changes. In addition, for station M1, the percentage of particles > 63 µm is shown, to highlight the coarse second peak, or "shoulder" of the grain-size distributions."

13 / 12-15. This paragraph (together with the comment that deposition fluxes will be assessed separately) addresses my comment about giving at least an idea of the potential amounts of those large/giant particles. In their reply to my previous comment (8 / 11-15) the authors also seem to confirm that overall their counting statistics are high, since they have very concentrated samples (as opposed to ice cores). While in general I must agree with that, as expected, I still wonder if this is also the case for the tails in particular, which were the object of my concern. The microscope images indeed show the presence of those particles. I wish that the authors could provide some additional insight into the procedure to convince us that those are also a genuine signal from the water column (e.g. not contamination from the laboratory or similar). These data are are very promising, and the first of this kind, and many colleagues should be interested in those, therefore I think it's worth assessing potential problems in their interpretation, or more positively assuring their validity.

Indeed the particles represented in the tails or shoulders of the grain-size distributions are most likely only a few particles, which, due to their size, have a higher impact on the volume-size distribution than smaller particles. Still, when looking at the microscope images, a handful of these "giant" particles are observed in each sample, and when considering the samples are only 1/25 of the original sample, which sampled for 16 days and collected material over 1 m^2, we come to the conclusion that the total transport and deposition of these giant particles over the Atlantic Ocean must be substantial.

To illustrate these giant particles even better, and to show that they are present at every station along the transect, we've added more microscope images to Figure 5 in the revised manuscript (page 10). As with any analysis, contamination could happen at any stage, but this is of course limited as much as possible. All parts of the sediment trap are kept clean throughout the whole sampling procedure. The sampling bottles are cleaned thoroughly at the home institute (cleaned with 1N HCl and rinsed thoroughly with demineralized water) and then closed. Once onboard the ship, where the biocide is added, all handling of the bottles occurs in fume hoods so that we can be confident that contamination is minimal. Since these giant particles are present in almost every sample, at every station along the transect, the chance of these particles being contamination in every case is considered negligible.

We modified the following lines in the revised manuscript:

Page 13, lines 22-30:

"However, we observed giant particles (≥100 µm) as far west as station M5 (57° W; approximately 4400 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). Only a handful of these coarse particles are found in the samples, however when considering these are 1/25 of the original samples, collecting sediments over 1 m^2 of ocean, over a time period of 16 days, this means that the amount of giant particles being transported over the Atlantic Ocean every year can be considered to be substantial. Such coarse particles are generally not incorporated into climate models (Kok, 2011).The underestimation of the coarse size fraction in climate models may have its origin in the sampling of dust of specific size classes, e.g. PM_{10} and $PM_{2.5}$, which form the basis of the guidelines from the World Health Organization (WHO, 2006) on fine-grained particles."

14 / 10. I would make it more clear in the text that this is one possibility, as the authors point out in their replies. E.g. something like "Assuming a consistent relation in the vertical profiles (as discussed above), we speculate that this difference may be related to a change in conditions in the source areas."

We tried to highlight the fact that increased emission as a result of human activity due to commercial agriculture is one possible mechanism that could explain the difference in particle size between the sediment traps and seafloor sediments, by modifying the following lines in the revised manuscript:

Page 14, lines 3-9:

"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 is one possible mechanism that could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we observe now in the sediment traps. However, we do not exclude other mechanisms that could be responsible for the change in particle size of mineral dust as deposited along the sampled transect."

19 / 7-9. This sentence is unclear

We modified the following sentence in the revised manuscript:

Page 18, lines 26-29:

"Coarser dust is found in the sediment traps as opposed to the seafloor sediments, which is in line with increased emission and coarser dust due to the onset of commercial agriculture in the 19th century, and is a possible explanation for the difference in particle size between the two records."

19 / 19. Didn't you suggest this is unlikely, based on HYSPLIT?

In this case, HYSPLIT does not show a distinct seasonal difference in air-layer trajectories, however this does not exclude changing source areas on a seasonal or (multi-)annual scale. HYSPLIT can serve as a good indicator of air-layer trajectories, however when calculated over longer time intervals these trajectories become more uncertain. In addition, from HYSPLIT it is not clear what surface conditions contribute to the uplift of particles into the long-distance transporting air layers. Still, differences in source areas could contribute to differences in composition and particle size of the dust.

We modified the following lines in the revised manuscript:

Page 18, lines 9-13:

"An increased number of coarse particles during spring could mean that the dust originates from a different source area. Backward trajectories calculated over the entire sampling period do not show this. However, these backward trajectories serve only as an indicator for air-layer trajectories, but from these it does not become clear what surface conditions contributed to the uplift of particles in the long-distance transporting air layers."

19 / 20. I would suggest "Multiple-year samples from this transect AND COUPLED DEPOSITION FLUX MEASUREMENTS should clarify"

We agree and modified the indicated sentence in the revised manuscript:

Page 19, lines 1-3:

"Multiple-year samples from this transect and coupled dust deposition fluxes should clarify which of the above mentioned processes are more dominant, in order to be incorporated into e.g. climate models and climate reconstructions."

References:

McGee, D., P. B. deMenocal, G. Winckler, J. B. W. Stuut, and L. I. Bradtmiller (2013), The magnitude, timing and abruptness of changes in North African dust deposition over the last 20,000 yr, Earth Planet. Sci. Lett., 371–372, 163–176, doi:10.1016/j.epsl.2013.03.054.

Referee #3

van der Does et al. present interesting results regarding deposition of dust across the Atlantic Ocean using moored traps. The potential for this study is significant, considering the cross disciplinary effort between sedimentology and atmospheric physics and transport. However, a dearth of information regarding the atmospheric component, even in this version, warrants further revision.

General comments:

I see that many of the authors' responses to the reviewer comments, although sufficient in providing the necessary explanations, did not result in any additions to the manuscript. As in this case, should the authors need to explain themselves to the reviewers for clarity, the readers will certainly need and appreciate the explanations as well. I suggest the authors take their responses from the first revision and incorporate them into the manuscript more than they currently have.

We have tried to incorporate more of the responses to the reviewer's comments in the manuscript. Please see the new revised manuscript with tracked-changes to see the most recent updates to the paper.

I continue to question the speculation that all the particles sized were dust based on the information provided. As an aerosol chemist, I observe quite a bit of spatial and vertical heterogeneity in atmospheric aerosols, even when close to a source region. Without chemical analysis, I find it difficult to believe all the particles from this study were dust, even considering the chemical treatment methods used to reduce the contribution from other aerosol types. Considering the work adds another variable to what could influence the aerosol composition, there could be more room for introducing complexity from additional particulate components from the ocean. With that said, there are a few things the authors could do to alleviate this issue as discussed below.

As we have tried to explain in the methods section of the paper, all the biogenic particles of marine origin are chemically removed: this includes all organic matter, (biogenic) carbonates and biogenic silica. What is left is the insoluble or lithogenic fraction, which is only sourced in atmospheric mineral dust. This may potentially include a small fraction of volcanogenic and cosmogenic particles as well (Plane, 2012), however on the other hand, carbonate dust particles and organic particles of non-marine origin are also removed from the samples by the chemical treatment applied. Since this is the case for every sample that we analyzed in this study, they can be compared directly to each other. In addition, we know that most of Saharan dust is made up of siliciclastics (e.g. Scheuvens and Kandler, 2014), which are not affected by this chemical treatment. Again, we would like to stress the fact that all marine particles (including foraminifera, radiolarians, diatoms, coccolithophores, etc.) have been removed from the samples prior to grain-size analysis. To verify if indeed all marine biogenic particles were removed, we performed microscope analysis (Figure 5). This confirmed that the fraction analyzed for particle size is the insoluble or lithogenic fraction, interpreted as mineral dust.

We modified the paragraph from the methods section addressing this issue.

Page 6, lines 7-17:

"The sediment traps collect all particles settling down into the ocean, that besides mineral dust includes the skeletons of marine plankton (foraminifera, radiolarians, diatoms, coccolithophores, etc.), organic matter (marine and aerosols from biomass burning) and potentially volcanogenic and cosmogenic particles (Plane, 2012). All these biogenic constituents were chemically 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 H_2O_2, followed by dissolving the biogenic carbonates using HCl, and removing biogenic silica by adding NaOH. What remains is the lithogenic fraction which is considered to consist mainly of dust, as confirmed by microscope analysis (Fig. 5). Indeed, some of the dust particles have a risk of being removed during this process including lithogenic carbonates and organic particles of non-marine origin. However, lithogenic carbonates are more resistant to the chemical treatment than the biogenic carbonates. Also, since this is the case for every sample analyzed, they can be compared directly to each other."

There are ways to support conclusions regarding aerosol type, such as the authors do with MODIS. However, MODIS also has its limitations. For instance, in Figure 10, AOD is clearly enhanced in a pathway from Africa, but

these aerosols could be dust or emissions from biomass burning, which is especially predominant in the summer and is evident by the high AOD propagating off central Africa. Did the authors account for biomass burning aerosol? Have the authors considered adding CALIPSO to their analysis to evaluate aerosol type, at least between the coast of Africa and M1? This would greatly reduce the assumptions made regarding aerosol type.

Indeed, some mixing of aerosols (mineral dust and aerosols from biomass burning) could occur (Adams et al., 2012), and it could be possible that these aerosols resulting from biomass burning are collected within the sediment traps. However, since they are of organic origin, they are chemically removed prior to grain-size analysis, using Hydrogen Peroxide (see also the Methods section, page 6 of the revised manuscript), so these are not accounted for in the grain-size distributions as presented in this paper. Based on CALIPSO data, Adams et al. (2012) speculate that smoke is not transported over great distances, opposed to mineral dust. Figure 10 is mainly used to illustrate the seasonal latitudinal movement of the dust cloud. It shows that AOD values are highest during summer, not only over the studied transect but also more to the south, between the equator and 10 °S, the latter possibly being related to biomass burning. We do not make a link between AOD and dust deposition fluxes, since these data are not available for this paper.

We added the following lines to the revised manuscript:

Page 6, lines 7-12:

"The sediment traps collect all particles settling down into the ocean, that besides mineral dust includes the skeletons of marine plankton (foraminifera, radiolarians, diatoms, coccolithophores, etc.), organic matter (marine and aerosols from biomass burning) and potentially volcanogenic and cosmogenic particles (Plane, 2012). All these biogenic constituents were chemically 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)."

Page 14, lines 15-17:

"However, the aerosols over our study area are mostly mineral dust originating from the African continent (Yu et al., 2015), also since Adams et al. (2012) speculate that smoke is not transported over great distances, as opposed to mineral dust."

HYSPLIT is a great tool, but when used cautiously. Typically, the error in a single trajectory results in 30% of its distance, meaning the error in where the air mass traveled can be quite substantial far from its initiation location. This can be alleviated by using an ensemble of HYSPLIT trajectories. However, only 4 trajectories are shown, which are also used to conclude in the abstract that transport occurs at higher altitudes and fast winds. Evidence supporting this conclusion is not provided, thus making it quite speculative. The authors do state in the responses that they did this for the year at M1 and it resulted in a mess of sources, however, these trajectories can be clustered. Showing clusters of trajectories, perhaps at various seasons and locations would increase the statistical significance and reduce the uncertainty in the sources and transport pathways, and better support their arguments.

The HYSPLIT trajectories shown in the paper are only a means to illustrate a typical winter and summer dust transport scenario. These differences in dust transportation altitude between the different seasons have been demonstrated in literature before (e.g. Tsamalis et al., 2013;Adams et al., 2012), and this was used to interpret the data of the present paper. Figure 9A and -B also illustrate the different transport altitudes between summer and winter. In the caption of Figure 9 we also note that these are "typical examples" of summer and winter dust transport, as described in the cited literature.

It is true that sea salt is soluble, but mineral surfaces have been shown to become coated by these soluble species in solution (REFS) and at times can be an irreversible reaction, as been shown in cloud droplets (CZICZO). Considering these particles are much larger than aerosols studied in clouds, there is substantial surface area for these (sometimes) irreversible reactions to take place. How are the authors so sure that the treated dust particles are devoid of soluble components, or other types of particles?

Without a comprehensive chemical analysis we have not verified this, however the microscope images show giant quartz particles with no apparent coatings. For further demonstration of the giant particles in the samples, we have added more microscope images to Figure 5 (page 10 of the revised manuscript). This also illustrates that these giant particles are deposited at every station along the transect, however possibly decreasing in amount towards the west.

Overall, providing more sound, empirical evidence in addition to more statistics on air mass transport would support the authors' conclusion that what they are sizing is most likely dust. At the very least the authors should clearly highlight the assumption that these particles are dust throughout the manuscript, and provide some more background on what other particulate components could be contributing to the samples. Providing examples of previous studies evidencing dust as the major component in this region would also help.

Since these samples were obtained by submarine sediment traps, the bulk samples exist of mainly marine particles, including organic matter, planktonic biomineral shells (e.g. foraminifera, radiolarians, diatoms, coccolithophores, etc.) next to mineral dust, and potentially volcanogenic and cosmogenic particles (Plane, 2012). After the chemical treatment, in the form of sequential leaching, all biogenic particles are removed from the sample and what is left is solely the insoluble, lithogenic fraction, which is almost entirely made up of mineral dust (see also the reply to the first question). As Yu et al. (2015) have pointed out in their study, 182 million tons of Saharan dust are transported from Africa over the Atlantic Ocean, which is calculated to amount to 5 $g/m^2/yr$. This considerable amount evidences dust as the major lithogenic component in the Atlantic Ocean. As mentioned also in the first rebuttal, lithogenic input other than mineral dust is believed to be negligible.

Were any of the extracted dust samples weighed prior to and following chemical treatment to remove non-dust components? This would strengthen the argument that what was left was likely dust, i.e., if total mass concentrations decreased after treatment. Would also provide a more quantitative approach as compared to Figure 2.

As mentioned before, dust mass fluxes would be a valuable addition to the data presented in this paper, but this will be the scope of another paper. However, some masses have been determined. As an example, for station M1, the average weight of the total bulk sample (so including all marine particles) is close to 1800 mg. The average weight of the isolated lithogenic (dust) fraction of the samples is nearly 30 mg. This means that the lithogenic fraction is about 1.7 % of the total sample, and proving the leaching procedure very effective. Figure 2 is intended to illustrate the similarity between the upper (1200 m) and lower (3500 m) traps, and demonstrate the settling velocity of the sediments.

The authors do not provide any discussion of or show uncertainty in the measurements in the manuscript. Please show error bars in the figures and clearly discuss the possible sources of uncertainty or variability in the measurements. For example, Figure 3 shows several markers close together, but it is unknown if these disparities are significant or simply within error. It would be more difficult to draw the conclusions discussed by the authors should these all be within error of each other.

For this study, we did not analyze any duplicate samples, since there simply are not more samples to be analyzed. The reproducibility is checked regularly at MARUM, Bremen (Germany), where author JBS holds a part-time position, using exactly the same equipment set-up as in this study, including the same laser particle sizer and degassed-water system, by Dr. J. Titschack. This is done by replicate analyses of three internal glass-bead standards, and the reproducibility is found to be better than ± 0.7 µm for the mean and ± 0.6 µm for the median particle size (1σ). The average standard deviation integrated over all the size classes is better than ± 4 vol %.

The markers in Figure 3 are an average of seasons (3 months), covering in total 5 to 7 samples per season. The more accurate difference between the seasons can be observed when looking at Figure 7 (Figures 7 – 9 of previous version), which shows more accurately that there is indeed a significant difference between the different seasons. Figure 3 is also meant to illustrate the difference between the sediment trap samples and the seafloor sediment samples, the latter being finer grained.

We have added some lines to the Methods section about the reproducibility of the laser particle sizer.

Page 6, lines 24-28:

"The reproducibility is checked regularly at MARUM, Bremen (Germany), using exactly the same equipment set-up as in this study, by Dr. J. Titschack. This is performed by replicate analyses of three internal glass-bead standards, and the reproducibility is found to be better than ± 0.7 µm for the mean and ± 0.6 µm for the median particle size (1σ). The average standard deviation integrated over all the size classes is better than ± 4 vol %."

There is quite a bit of redundancy in some of the figures (namely 7, 8, and 9). The authors should consider combining these, which will serve to eliminate the redundancy and a more direct comparison.

We initially chose to separate these figures to avoid a figure becoming too crowded. Plotting modal grain sizes of all traps (N=7) together in one plot will result in an unclear image. Separating the upper and lower traps made it harder to compare these for stations M2 and M4, where both are present, thus resulting in Figure 9. However, we have chosen now to combine Figures 7, 8, 9 and 12, as Figure 7A and B in the revised manuscript (page 12).

Specific comments:

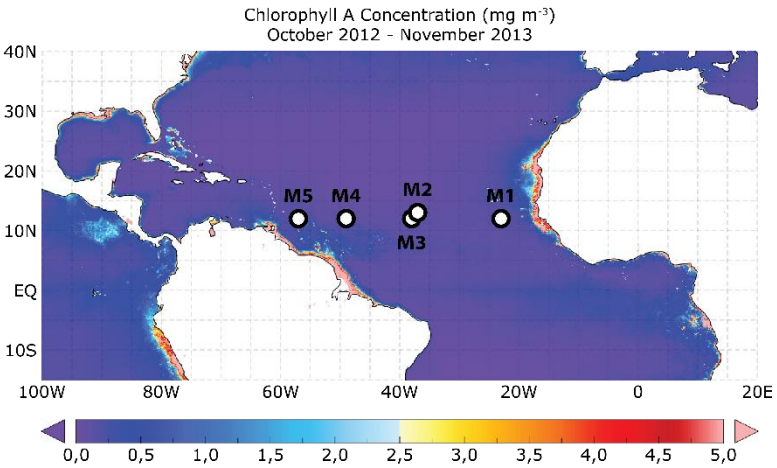
Page 5, lines 11-12: Elaborate on this. Where was chlorophyll and salinity higher? This should perhaps be shown in a supporting information file, especially considering these can change greatly on a seasonal basis throughout the year.

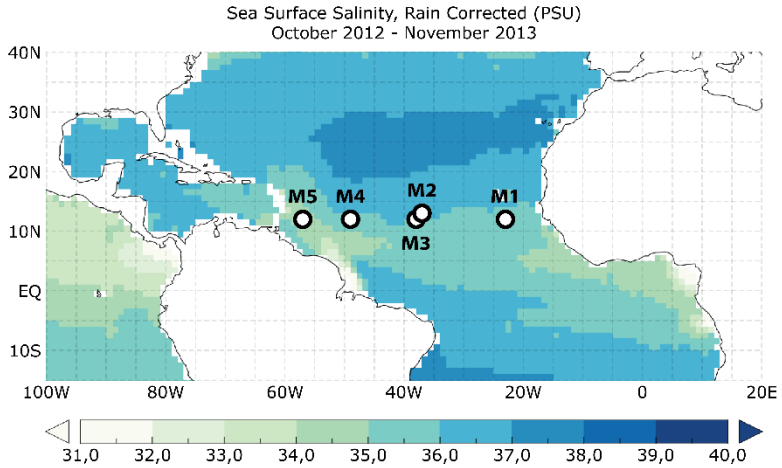
Data of Chlorophyll and salinity can track fresh-water input and productivity from major rivers in the ocean. As seen from this data, the influences at the sampling stations seem to be minor. These graphs (see below) are added as Supplement to the manuscript.

We modified the following lines to the revised manuscript:

Page 5, lines 19-20:

"Limited influence of major rivers is also visible when looking at (satellite) data of chlorophyll or salinity (see Supplement)."





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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.,

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 to 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 pCO_2 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. 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 (Adams et al., 2012;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

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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).

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 from submarine sediment traps moored at five locations along this transect, sampling synchronously over successive intervals 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. (2016) 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 generally, 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 m^2 and an 8mm hexagonal baffle on top to maximize particle collection (Knauer and Asper, 1989) and prevent large swimmers from entering the sediment trap. Underneath the funnel, a rotating carrousel with 24 sampling cups collects discrete samples of the settling particle flux. All sediment traps operated synchronously over pre-programmed intervals of 16 days, and performed flawlessly. Tilt-meters showed that the sediment traps remained nearly upright for the entire sampling period. With few exceptions, current velocities, as measured by current meters and ADCPs, remained well below 12 cm s^{-1}, the threshold below which unbiased collection of settling particles occur (Knauer and Asper, 1989). This paper presents the

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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. 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.

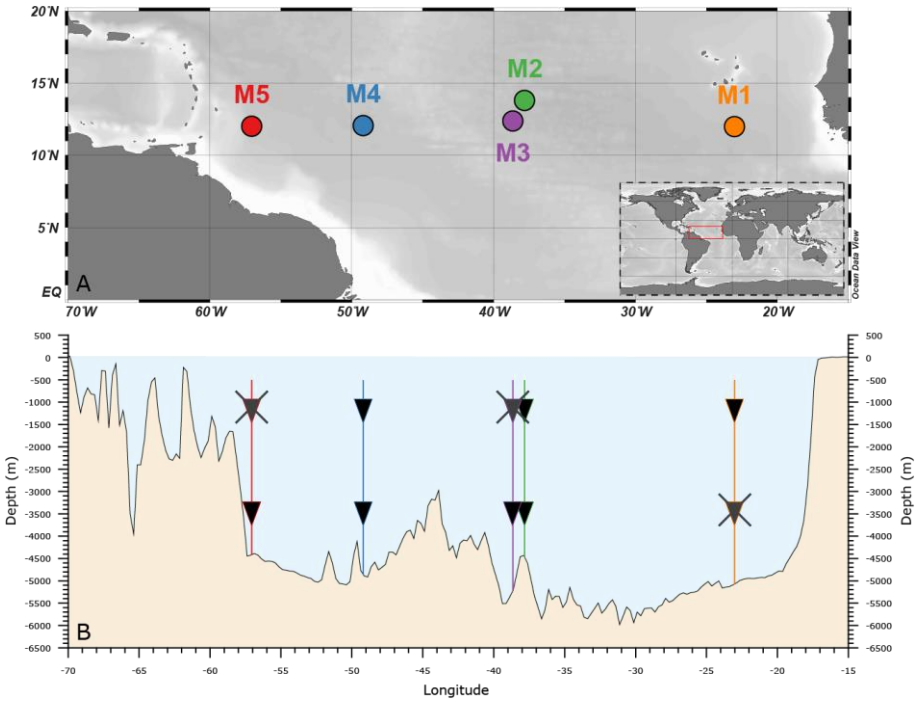

Figure 1. A: Map with sampling stations M1–M5 in the Atlantic Ocean at 12° N. B: Bathymetry along 12° N (from www.gebco.net) with sediment traps at 1200m and 3500m BSL. Crossed-out sediment traps could not be recovered.

Field Code Changed

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 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. These high settling velocities can be reached since the mineral dust particles are not deposited individually, but as part of large marine particles like marine snow. These are aggregates > 500 µm of organic and inorganic particles of different composition and origin (e.g. Nowald et al., 2015). These particles have settling velocities of approximately 200 m day^{-1}, and may increase with increased depth (Berelson, 2002;Nowald et al., 2015). The samples also show 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 (see Supplement). 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.

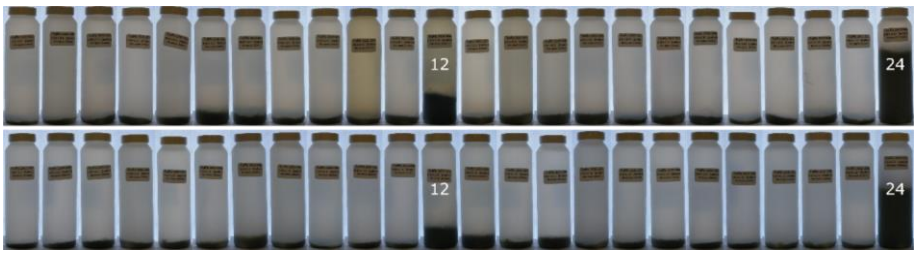

Figure 2: Sediment-trap bottles of the upper (1200 m) and lower (3500 m) traps at station M4 (12°N, 49° W).

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; $Na_2B_4O_7 \cdot 10H_2O$; end concentration 1.3 g L^{-1}, pH ≈ 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

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average weight difference between replicate aliquots of each sample is 2.4% (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 x g with Milli-Q water to remove the $HgCl_2$, borax, and sea-salts.

The sediment traps collect all particles settling down into the ocean, that besides mineral dust includes the skeletons of marine plankton (foraminifera, radiolarians, diatoms, coccolithophores, etc.), organic matter (marine and aerosols from biomass burning) and potentially volcanogenic and cosmogenic particles (Plane, 2012). All these biogenic constituents were chemically 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 H_2O_2, followed by dissolving the biogenic carbonates using HCl, and removing biogenic silica by adding NaOH. What remains is the lithogenic fraction which is considered to consist mainly of dust, as confirmed by microscope analysis (Fig. 5). Indeed, some of the dust particles have a risk of being removed during this process including lithogenic carbonates and organic particles of non-marine origin. However, lithogenic carbonates are more resistant to the chemical treatment than the biogenic carbonates. Also, since this is the case for every sample analyzed, they can be compared directly to each other. Immediately prior to the grain-size measurements sodium pyrophosphate ($Na_4P_2O_7 \cdot 10H_2O$) 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. The system uses degassed water, to minimize the influence of air bubbles. 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. The reproducibility is checked regularly at MARUM, Bremen (Germany), using exactly the same equipment set-up as in this study, by Dr. J. Titschack. This is performed by replicate analyses of three internal glass-bead standards, and the reproducibility is found to be better than ± 0.7 µm for the mean and ± 0.6 µm for the median particle size (1σ). The average standard deviation integrated over all the size classes is better than ± 4 vol %.

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).

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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

The particle size of Saharan dust can be expressed in many ways. Showing grain-size distributions of all individual samples makes it more difficult to distinguish seasonal and spatial changes. Therefore, the modal particle size is chosen to represent the main dust mode, to better illustrate these changes. In addition, for station M1, the percentage of particles > 63 µm is shown, to highlight the coarse second peak, or "shoulder" of the grain-size distributions.

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). This shows that coarse particles are not only deposited at proximal locations, but also transported over great distances. These shoulders are also found in the seafloor sediments (Fig. 4B). They include "giant" particles of more than 100 µm, which are observed as far west as station M5 (57° W; approximately 4400 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 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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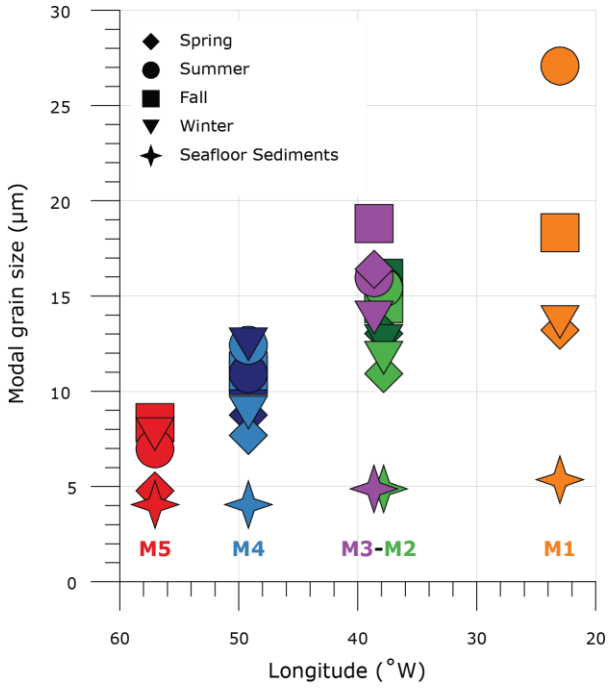

Figure 3. Downwind fining and seasonality in average modal grain size per season for all seven traps (an average of all modal values from the relative volume grain-size distributions, grouped per season), for October 2012 – November 2013, and modal grain size of the seafloor sediments (from the grain-size distributions as show in Figure 4B), versus western longitude. 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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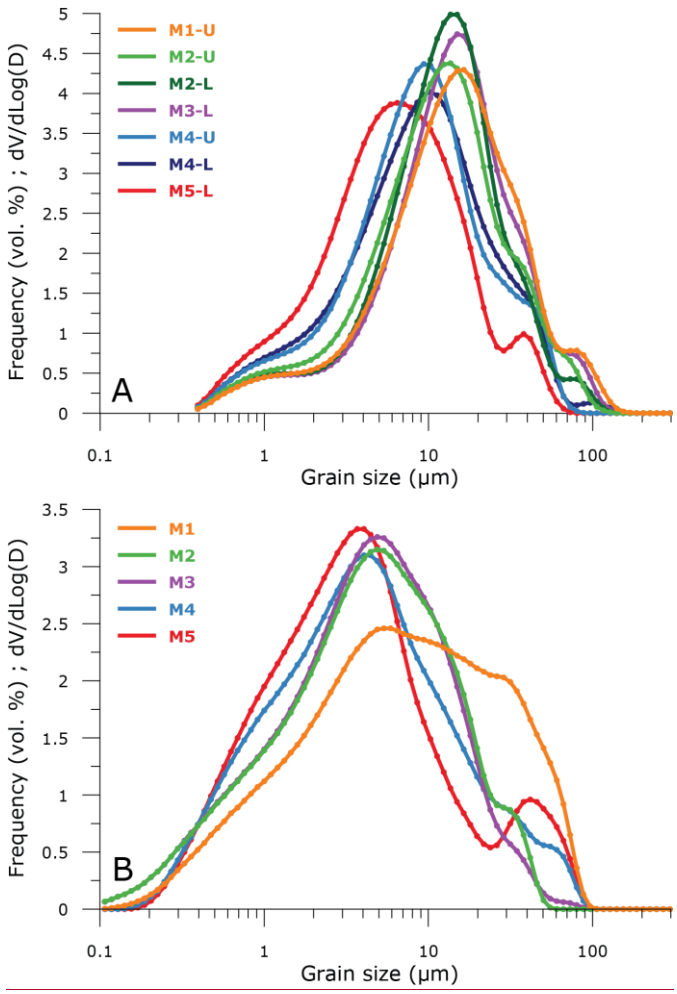

Figure 4. A: Average volume grain-size distributions of 24 samples from all seven sediment traps, where U = upper trap (1200 m) and L = lower trap (3500 m). Collected between October 2012 and November 2013. B: Grain-size distributions of seafloor sediments at the five mooring stations (M1–M5) along the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

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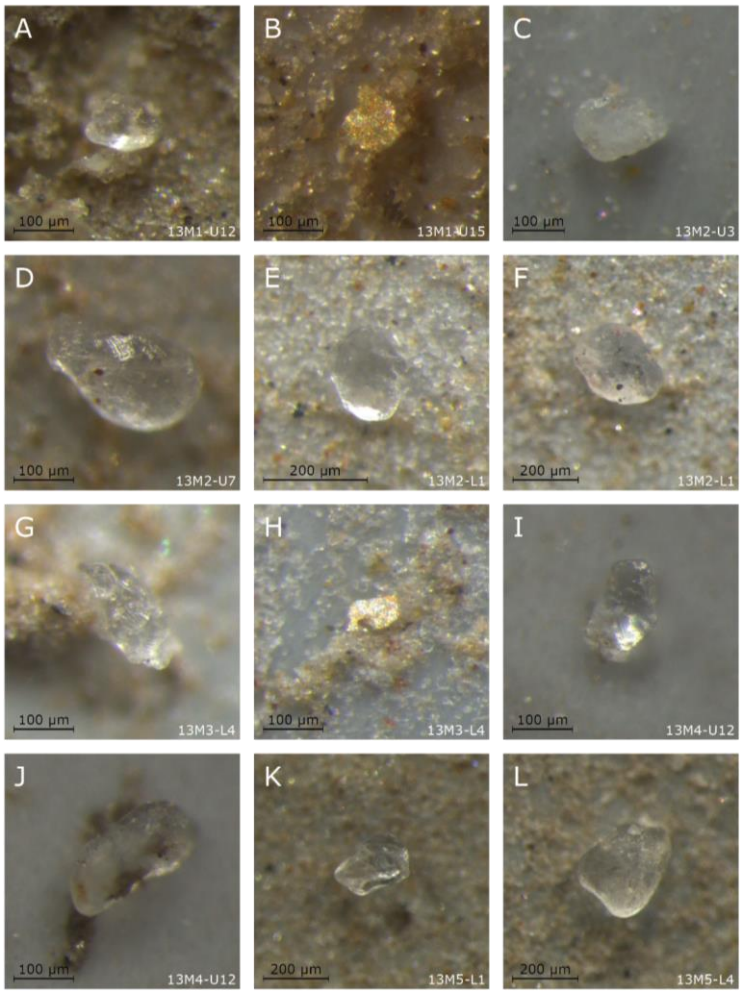

Figure 5. Light-microscope images of large dust particles from the sediment trap samples. Name in lower-right corners: 13Mx denotes the station (see Table 1 for the exact locations). U and L denotes Upper (1200 m) and Lower (3500 m) traps, respectively, followed by the sample number (out of a total of 24). A: Quartz particle (diameter approximately 140 µm); B: Mica particle (120 µm); C: Quartz particle (180 µm); D: Quartz particle (270 µm); E: Quartz particle (180 µm); F: Quartz particle (290 µm); G: Quartz particle (200 µm); H: Mica particle (85 µm); I: Quartz particle (180 µm); J: Quartz particle (250 µm); K: Quartz particle (240 µm); L: Quartz particle (300 µm).

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), more prominent than during the other seasons. Also modal grain sizes illustrate this

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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. 7A) 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.

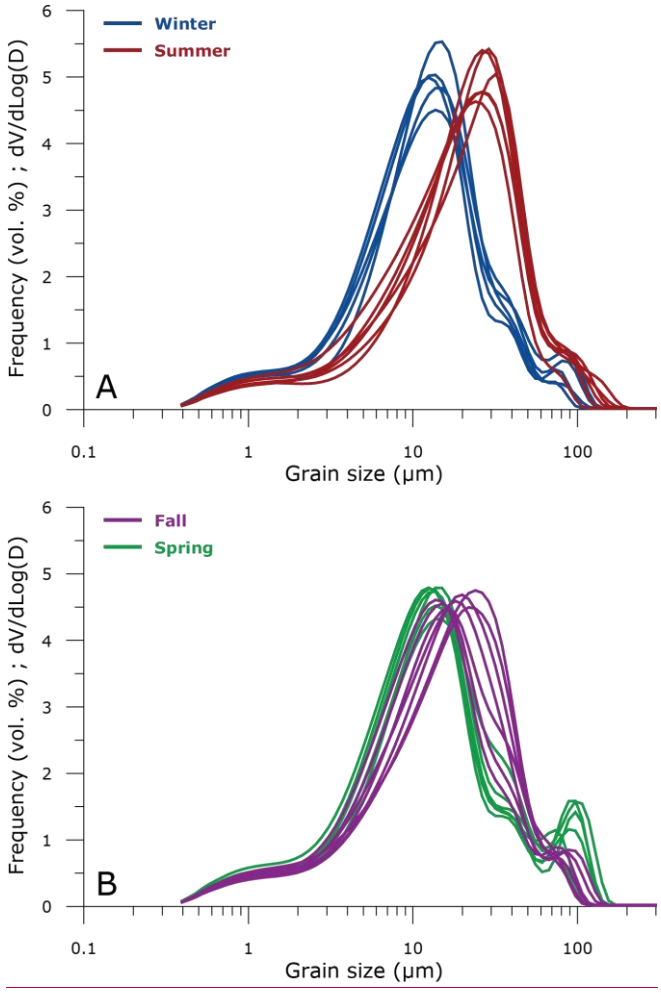

Figure 6. Seasonal grain-size distributions at station M1 (12° N, 23° W; approximately 700 km from the African coast) for October 2012–November 2013, of A: winter (blue), summer (red), B: fall (purple) and spring (green).

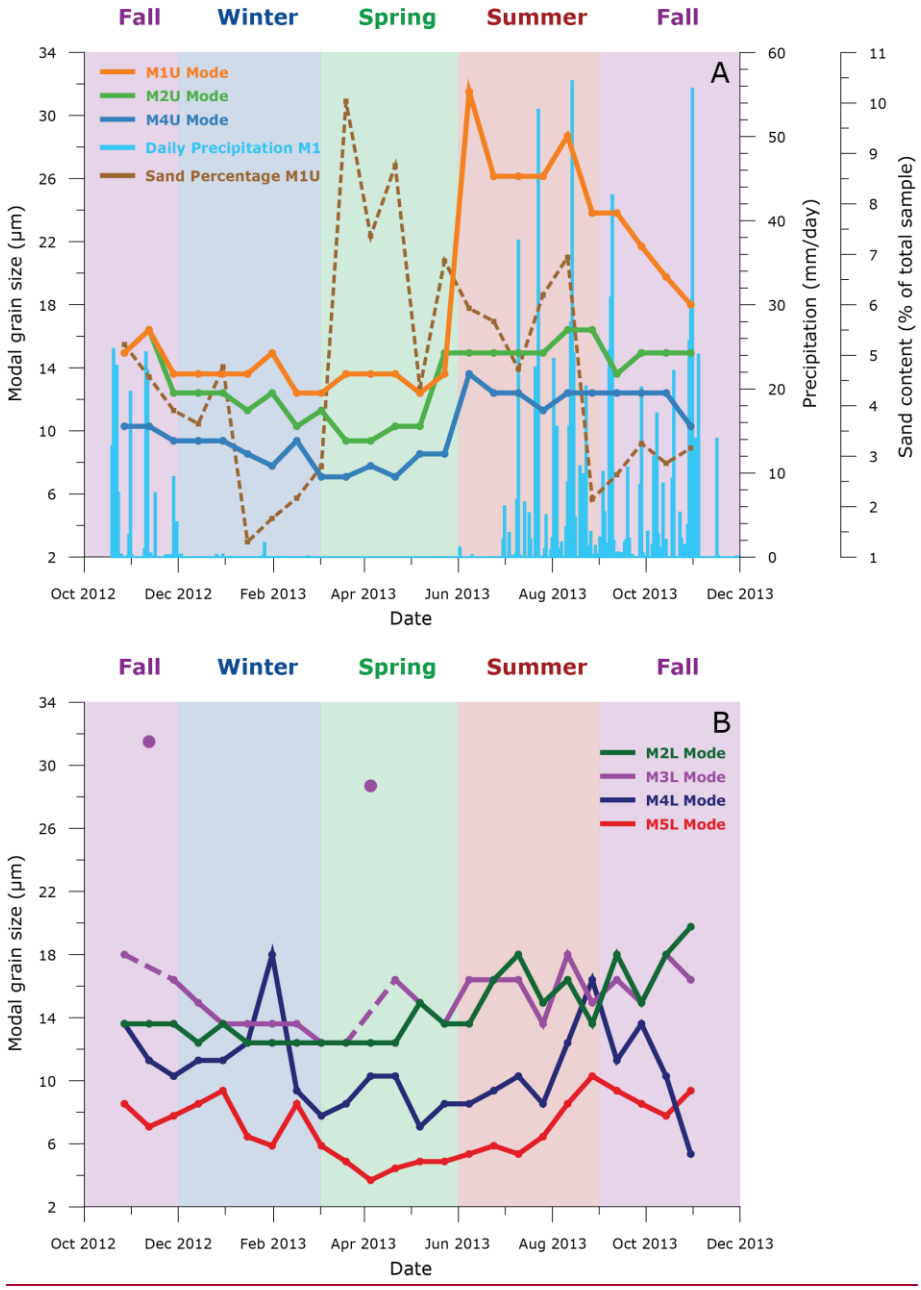

Figure 7. A: **Modal particle diameter of dust collected by the three upper (1200 m) sediment traps at stations M1, M2 and M4** (left axis), daily precipitation (right axis; TRMM) and percentage of sand particles (> 63 µm) (far right axis)

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at station M1. B: Modal particle diameter of dust collected by the three lower (3500 m) sediment traps at station M2, M3, M4 and M5. The two points that are not connected in series M3-Lower are considered outliers.

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. 7A). 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. 7B). 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. 7B). 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. 7).

4 Discussion

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 (≥100 µm) as far west as station M5 (57° W; approximately 4400 km from the African coast) (Fig. 5), and mica particles, whose platy shape allows for aerial transport over greater distances (Stuut et al., 2005). Only a handful of these coarse particles are found in the samples, however when considering these are 1/25 of the original samples, collecting sediments over 1 m^2 of ocean, over a time period of 16 days, it means that the amount of giant particles being transported over the Atlantic Ocean every year can be considered to be substantial. Such coarse particles are generally not incorporated into climate models (Kok, 2011). The underestimation of the coarse size fraction in climate models may have its origin in the sampling of dust of specific size classes, e.g. PM_{10} and $PM_{2.5}$, 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 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 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

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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.¶

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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 is one possible mechanism that could cause the particle size of the deposited Saharan dust to become gradually coarser over time, as we observe now in the sediment traps. However, we do not exclude other mechanisms that could be responsible for the change in particle size of mineral dust as deposited along the sampled transect.

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 depth (AOD) data (Fig. 8). 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), also since Adams et al. (2012) speculate that smoke is not transported over great distances, as opposed to mineral dust. In summer, when AOD values are highest, the cloud is located at its northernmost position (Fig. 8D). Aerosol concentrations are lowest during fall (Fig. 8A and -E), and during winter the cloud is located in its southernmost position (Fig. 8B). However, during winter the aerosols may receive a higher 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.

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. 7B). Thus, during winter the northern station M2 does not receive the same dust as station M3, since 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 of dust. 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 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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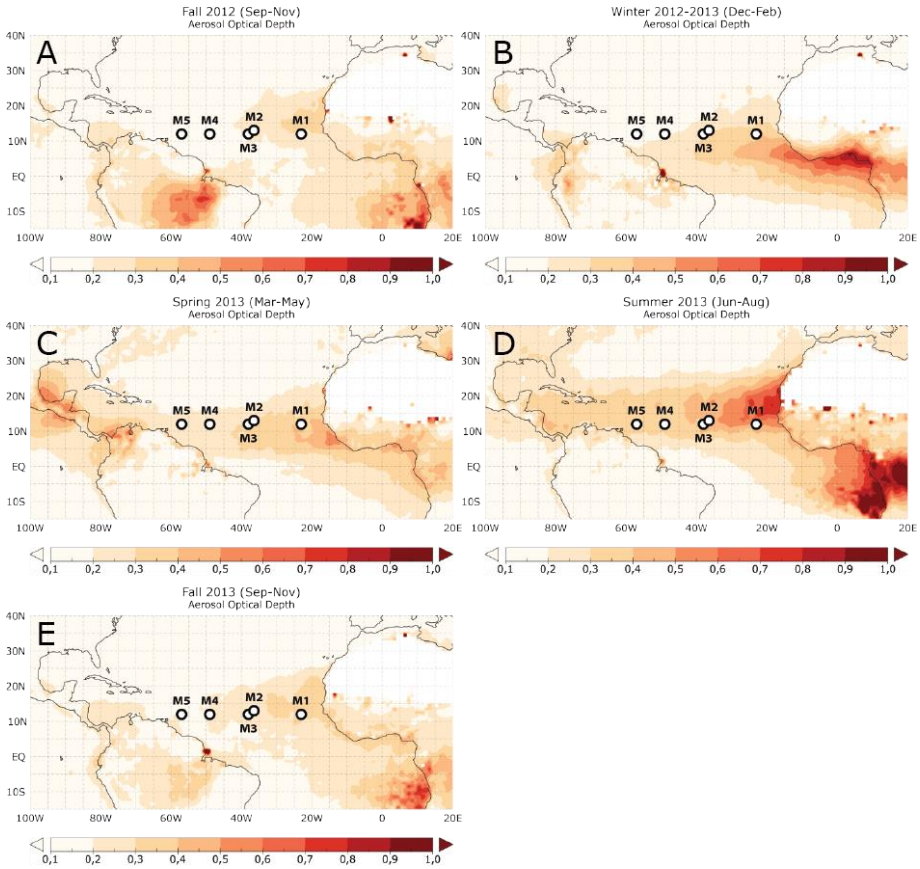

Figure 8. Three-month average aerosol optical depth (AOD) for the sampled seasons, from MODIS Terra. A: Fall (SON) 2012, B: Winter 2012–2013 (DJF), C: Spring 2013 (MAM), D: Summer 2013 (JJA) and E: Fall 2013 (SON). Stations M1–M5 are marked with black/white circles.

In winter, dust is transported at lower altitudes than during summer (Adams et al., 2012;Tsamalis et al., 2013). 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. 9A). 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. 9B), 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. (2014) argue that the dust particle size does not depend on wind speeds at emission. However, high wind velocities in the SAL of >7 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

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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. 9C 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 9A and -B, with the lowest (500 m) elevation representing winter dust transport and the highest (3500 m) elevation representing summer dust. In winter (Fig. 9C), 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. 9A), picked up from the surface and brought to higher altitudes. By contrast, in summer (Fig. 9D) 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. 9D, 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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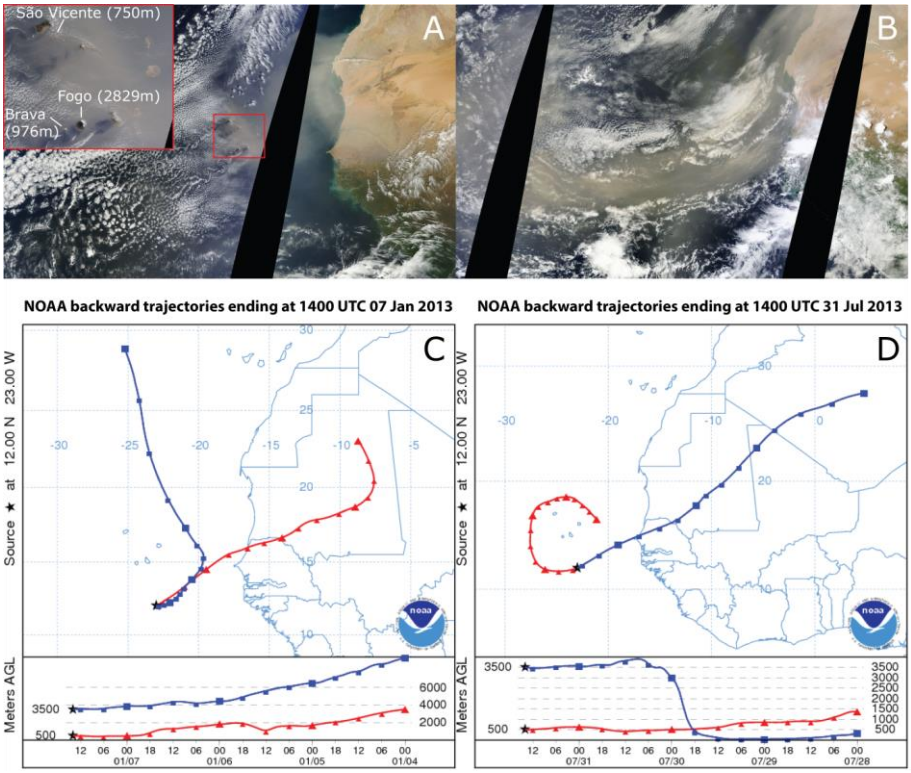

Figure 9. A: Satellite images of typical winter dust transport, with close-up of the Cape Verde islands (7 January 2013) and B: typical summer dust transport (31 July 2013) over the Cape Verde islands, at relatively low and high altitudes, respectively. Images from NASA Worldview, MODIS Terra satellite. 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.

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. 7A). This was also noted off northwest Africa related to wet deposition by Friese et al. (2016). 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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At M1, the percentage of sand-sized particles (> 63 µm) increases sharply in spring while modal grain sizes increase in summer (Fig. 7A). 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 (≥ 100 µm) 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 originates from a different source area. Backward trajectories calculated over the entire sampling period do not show this. However, these backward trajectories serve only as an indicator for air-layer trajectories, but from these it does not become clear what surface conditions contributed to the uplift of particles in the long-distance transporting air layers.

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.

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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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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 as opposed to the seafloor sediments, which is in line with increased emission and coarser dust due to the onset of commercial agriculture in the 19th century, and is a possible explanation for the difference in particle size between the two records. 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 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 µm) 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 and coupled dust deposition fluxes should clarify which of the above mentioned processes are more dominant, in order to be incorporated into 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.

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Acknowledgements

We thank the captain, crew and scientists of FS *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 J. Schwarz and three anonymous reviewers for their helpful comments.

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All data used in this manuscript has been stored at https://doi.pangaea.de/10.1594/PANGAEA.863030.

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Supplement to “Particle size traces modern Saharan dust transport and deposition across the equatorial North Atlantic”

Michèlle van der Does, Laura F. Korte, Chris I. Munday, Geert-Jan A. Brummer, Jan-Berend W. Stuut

Here we provide maps of Chlorophyll A concentrations and Sea Surface Salinity, over the sampled period October 2012 – November 2013, over the equatorial Atlantic Ocean.