

Saharan Dust from a Marine Perspective

Michèle van der Does

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Transport and Deposition Along
a Transect in the Atlantic Ocean

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VRIJE UNIVERSITEIT

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Transport and Deposition Along a Transect in the Atlantic Ocean

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Summary

Saharan dust is transported in great quantities across the Atlantic Ocean, and a large part is deposited before it reaches the Caribbean. However, little is known about this dust deposition across the entire Atlantic, and the physical characteristics of the particles involved. Therefore, a transect of subsurface sediment traps and dust-collecting buoys was deployed, which provides a unique opportunity to study dust deposition in a spatial and temporal high resolution over multiple years. In this thesis dust deposition fluxes and atmospheric dust concentrations along the transect, particle size of the dust and their downwind trends are described. In addition, evidence that giant mineral dust particles ($>75\text{ }\mu\text{m}$) are blown all the way across the ocean is given, transported over thousands of kilometres, which is explained with a range of possible mechanisms. Also, a possible influence of Amazon River sediments to the western sediment traps by means of geochemical analysis is investigated.

Chapter 2 '*Particle-Size Analysis Methods*' contains a detailed description of the different sample preparations, which were employed for the analyses of the dust samples used for this study. This ranges from atmospheric dust samples over the Atlantic Ocean, down to seafloor sediments along the studied transect. Also, three different particle-size analysers are described here. These include the *Coulter LS13 320*, which uses the angle of light scattering as a measure for particle size; the *Sympatec QicPic*, which determines the size of individual particles from two-dimensional particle contours based on image analysis; and the *Coulter "Counter" Multisizer 3*, which determines the volume of a particle suspended in an electrolyte that disturbs an electric field as it passes through an aperture between two electrodes.

The particle-size results for the first year of dust sampling along the trans-Atlantic sediment-trap array are presented in **Chapter 3** '*Particle Size Traces Modern Saharan Dust Transport and Deposition Across the Equatorial North Atlantic*', along with seafloor surface sediments and the identification of giant ($>75\text{ }\mu\text{m}$) mineral dust particles as far west as 4,400 km from the west African coast. A clear decreasing particle size with increased distance from the source is observed, as the coarsest-grained particles are deposited close to their source, and finer-grained particles can be transported over greater distances. In addition, distinct seasonal variations in particle size were observed, with coarser-grained dust deposited in summer, and finer-grained dust in winter and spring. This is attributed to atmospheric dust transport at different altitudes across the Atlantic Ocean during the different seasons, at high altitudes (up to 5–7 km) and high wind speed in summer within

the Saharan air layer (SAL), and lower altitudes (0–3 km) in winter with the Harmattan trade winds. We hypothesize that coarser-grained dust deposition may also be related to increased wet deposition by precipitation in summer, and more frequent dust storms emerging from the north-western African coast.

The different mechanisms of dust deposition, namely dry- and wet deposition, and their impacts on the bio-availability of carried nutrients, are discussed in **Chapter 4** ‘*Tropical Rains Control Saharan Dust Deposition and Bio-Availability of Nutrients*’. This chapter first combines two years of sediment-trap deposition fluxes and dust collected at Barbados with model simulations and satellite observations. The sediment traps reveal highest deposition fluxes in summer, at the same time of increased precipitation at the sampling sites, indicating wet deposition of dust. In winter, both deposition fluxes and precipitation are low. These observations match well with the model simulations, that also show highest (wet) deposition fluxes in summer, and dry deposition only in winter, in low amounts. At Barbados, furthest downwind along the transect, dust concentrations peak prior to the onset of precipitation, implying that the dust reaching the Caribbean is the result of dust not washed out by rain during the trans-Atlantic crossing. Therefore, we argue that North Atlantic precipitation controls airborne dust reaching Barbados. Furthermore, bottle incubation experiments demonstrate the release of nutrients from dust during atmospheric processing and wet deposition. This shows that wet deposition of dust likely has the largest marine-environmental effect by increasing nutrient availability (phosphorous, silicon, and iron), enhancing primary production and carbon sequestration.

A possible mechanism for the transport of giant (>75 µm) dust particles over thousands of kilometres across the Atlantic Ocean is presented in **Chapter 5** ‘*The Mysterious Long-Range Transport of Giant Mineral Dust Particles*’. The traditional physical laws describing the settling speed of particles overestimates the settling for particles >20 µm, so that they are seemingly impossible to transport over long distances. However, with the evidence of giant mineral dust particles collected from the atmosphere with a passive dust sampler as far as 3,500 km from the west African coast, we investigated possible mechanisms enabling the long-range transport of these giant particles. First, the SAL, that transports dust at high altitudes and high wind speeds, is most likely a key aspect. Turbulence in this layer causes particles to continuously be recirculated, and high concentrations of dust particles allows for tribo-electrification, resulting from particles colliding. This electric charge is found to act as a force compensating a particle’s weight, keeping them aloft. This seems especially efficient for large quartz particles, which also happens to be the main mineralogy of the giant particles we find at the sampling locations. In summer, convection can play an additional role, as giant particles can be brought to high altitudes by large convective cells or tropical cyclones. The probability of such repeated uplift by convection was determined by calculating backward trajectories.

Dust particle size and deposition flux in the sediment traps along the Atlantic transect of multiple years are presented in **Chapter 6** ‘*Seasonality in Saharan Dust Particle Characteristics Across the Atlantic Ocean: Transport Versus Deposition*’, combined with two years of dust concentrations and particle size as sampled by two moored surface buoys and passive MWAC (Modified Wilson and Cooke) samplers. The data show a high similarity between the atmospheric dust and the deposited dust in the ocean, but also differences are observed between transported and deposited dust. The sediment traps collect dust deposited by both dry and wet deposition. Dust deposition fluxes are increased in summer, coinciding with large amounts of precipitation and thus wet dust deposition. The buoys collect transported (dry) dust only, and do not show increased dust concentrations in summer as the dust is transported at higher altitudes, outside the sampling range of the buoys. This implies that the large amounts of dust transported at high altitudes in summer, deposited by wet deposition, determines the seasonality of the deposition flux and particle size in the sediment traps.

Strontium, Neodymium and Hafnium isotopes, Rare Earth Elements (REE), and dust particle size for sediment-trap samples and shipboard-collected aerosols are described in **Chapter 7** ‘*North African Mineral Dust Across the Tropical Atlantic Ocean: Insights from Dust Particle Size, Radiogenic Sr-Nd-Hf Isotopes and Rare Earth Elements (REE)*’. These results show no relation between Sr and Nd isotopic compositions and dust particle size. The good correlation between Hf isotopic composition and particle size is related to the “zircon effect”, as coarse-grained zircon minerals have low $^{176}\text{Hf}/^{177}\text{Hf}$ isotopic ratios. The differences in isotopic signatures between the different samples are attributed to the distance over which the dust has travelled and dust provenance. This in turn influences the mineralogy and particle size of the sampled dust. In addition, alternative sources of lithogenic particles to the western sediment trap are explored, and the influence of Amazon River-derived sediments is considered absent or negligible, based on the geochemical signature of Amazon Basin sediments and sediments from Amazon River tributaries. This confirms that the primary source of lithogenic particles in the sediment traps is of north African origin. The combined Sr and Nd isotopic signature indicates a source of the dust in the western region of northern Africa.

In **Chapter 8** ‘*Concluding Remarks*’ I elaborate on possible future research related to this project, on additional analyses on existing samples and alterations to the sampling transect. I discuss the downwind evolution of particle size and particle flux with different seasons. In addition, I present preliminary results of particle-size analysis of dust samples on Barbados, which can extend the current transect of sediment traps and dust-collecting buoys. Furthermore, I discuss the analysis of dust-particle shape and hypothesize on its possible outcome and importance, and how it would contribute to the characterization of mineral dust from source to sink.

Samenvatting

Woestijnstof uit de Sahara wordt in grote hoeveelheden getransporteerd over de Atlantische Oceaan. Een groot deel hiervan wordt, voor het stof het Caraïbisch gebied bereikt, al afgezet. Er is vrij weinig bekend over deze sedimentatie van stof over de Atlantische Oceaan en de fysieke eigenschappen van deze stofdeeltjes. Daarom is er een transect van sedimentvallen onder water en stof-vangende boeien boven water uitgezet, die een unieke kans bieden om de sedimentatie van stof in detail te bestuderen. Dit proefschrift beschrijft de depositiefluxen en atmosferische stofconcentraties, de korrelgrootte van het woestijnstof en ook de trends langs het transect. Het laat zien dat reuze stofdeeltjes ($>75\text{ }\mu\text{m}$) naar de andere kant van de Atlantische Oceaan worden gebracht, getransporteerd over duizenden kilometers. Dit hebben we geprobeerd te verklaren aan de hand van een aantal mogelijke mechanismen. Ook is er gekeken naar een mogelijke invloed van sedimenten afkomstig van de Amazone Rivier in de westelijk gelegen sedimentvallen door middel van geochemische analyse.

Hoofdstuk 2 ‘*Korrelgrootte Analyse Methoden*’ geeft een gedetailleerde beschrijving van de verschillende voorbehandelingsmethoden die zijn gebruikt voor de verschillende stofmonsters. Dit varieert van atmosferisch stof verzameld op de Atlantische Oceaan tot sedimenten van de zeebodem. Ook worden drie verschillende apparaten beschreven die zijn gebruikt voor de analyse van de korrelgrootteverdelingen van woestijnstof. Dit zijn de *Coulter LS13 320*, welke de breking van licht als maat voor korrelgrootte gebruikt; de *Sympatec QicPic*, die de grootte van een deeltje door middel van tweedimensionale contouren van een korrel bepaalt met camerabeelden; en de *Coulter ‘Counter’ Multisizer 3*, die het volume van een deeltje kan bepalen dat het elektrisch veld tussen twee elektroden verstoort.

Korrelgroottes van de stofdeeltjes van het eerste jaar van bemonstering langs het Atlantische transect van sedimentvallen worden gepresenteerd in **Hoofdstuk 3** ‘*Hedendaags Transport en Depositie van Sahara Stof over de Equatoriale Noord Atlantische Oceaan Herleid door Korrelgrootte*’, samen met zeebodem sedimenten en de identificatie van reuze stofdeeltjes ($>75\text{ }\mu\text{m}$) tot op 4.400 km vanaf de west Afrikaanse kust. We zien een duidelijke afname in korrelgrootte met toenemende afstand tot de bron, doordat de grootste deeltjes dichtbij deze bron worden afgezet en kleinere deeltjes over grotere afstanden kunnen worden getransporteerd. Ook zien we duidelijke seizoensgebonden trends, waarbij er grofkorreliger stof wordt afgezet in de zomer en fijn-korreliger stof in de winter en het voorjaar. Dit wordt

toegeschreven aan stoftransport op verschillende hoogten in de atmosfeer over de Atlantische Oceaan tijdens de verschillende seizoenen, waarbij het woestijnstof 's zomers op grote hoogten (tot 5–7 km) en bij hoge windsnelheden wordt vervoerd in de Sahara Luchtlag en 's winters op een lager niveau (0–3 km) met de Harmattan passaatwinden. Bovendien kan de afzetting van grofkorreliger stof ook gerelateerd zijn aan een toename van natte depositie door neerslag in de zomer en door frequentere stofstormen vanaf de noordwestelijke Afrikaanse kust.

De verschillende mechanismen van stofafzetting, namelijk droge- en natte depositie en hun invloed op de biologische beschikbaarheid van de getransporteerde nutriënten, worden beschreven in **Hoofdstuk 4** *'Tropische Regenbuien Bepalen de Afzetting van Sahara Stof en de Biologische Beschikbaarheid van Nutriënten'*. Allereerst wordt twee jaar van stof depositiefluxen uit de sedimentvallen en woestijnstof dat is verzameld op Barbados gecombineerd met modelsimulaties en satellietobservaties. De sedimentvallen onthullen dat de meeste depositie plaatsvindt in de zomer, tegelijk met een toename van neerslag, een indicatie voor natte depositie. In de winter zijn zowel de depositieflux als de neerslag laag. Deze observaties komen overeen met de modelsimulaties, die ook de hoogste (natte) depositiefluxen laten zien in de zomer en droge depositie alleen in de winter en in kleine hoeveelheden. Op Barbados, ver van de Afrikaanse bron verwijderd, zijn de stofconcentraties het hoogst vlak voordat de zomerregens beginnen. Dit impliceert dat het woestijnstof dat de Caraïben bereikt het overblijfsel is van stof dat niet door regen is weggespoeld tijdens de Atlantische oversteek. Wij betogen daarom dat het stof dat Barbados bereikt bepaald wordt door neerslag over de Noord Atlantische Oceaan. Verder laten incubatie experimenten zien dat tijdens atmosferische processen en natte depositie nutriënten vrijkomen van woestijnstof. Daarom heeft natte depositie waarschijnlijk het grootste effect op het mariene milieu door de beschikbaarheid van nutriënten (fosfor, silicium en ijzer) te vergroten en daardoor ook op de primaire productie en de langdurige opslag van koolstof.

Een mogelijk mechanisme voor het transporteren van reuze stofdeeltjes ($>75\ \mu\text{m}$) over duizenden kilometers over de Atlantische Oceaan wordt gepresenteerd in **Hoofdstuk 5** *'Het Mysterieuze Transport van Reuze Stofdeeltjes over Grote Afstanden'*. De traditionele natuurkundige wetten die de valsnelheid van deeltjes beschrijven overschatten de valsnelheden voor deeltjes $>20\ \mu\text{m}$, waardoor hun transport over lange afstanden onmogelijk lijkt. Naar aanleiding van het bewijs dat deze reuze stofdeeltjes tot wel 3.500 km vanaf de west Afrikaanse kust in de atmosfeer gevonden worden hebben wij de mogelijke mechanismen onderzocht die het transport van deze reuzedeeltjes mogelijk kunnen maken. Allereerst is de Sahara Luchtlag, die stof transporteert op grote hoogten en met hoge windsnelheden, hoogstwaarschijnlijk een belangrijk en essentieel aspect. Turbulentie in deze luchtlag zorgt ervoor dat de stofdeeltjes continu worden gerecirculeerd, en de hoge concentraties zorgen voor het zogenaamde tribo-elektrisch effect, oftewel de door wrijving verkregen elektrische lading. Deze lading werkt als een kracht die het gewicht van een deeltje compenseert waardoor deze langer in de lucht kan blijven. Dit lijkt vooral efficiënt

voor grote kwartskorrels die het voornaamste deel uitmaken van de reuzedeeltjes in de stofmonsters. Convectie kan ook nog een rol spelen in de zomer wanneer deze reuzedeeltjes tot grote hoogten gebracht kunnen worden door grote convectiecellen of tropische cyclonen. We hebben hier bepaald hoe waarschijnlijk het is dat stofdeeltjes meerdere keren omhoog gebracht kunnen worden door convectie.

De korrelgrootte en depositie fluxen van het bemonsteren van woestijnstof over meerdere jaren langs het Atlantische transect worden gepresenteerd in **Hoofdstuk 6** *'Seizoenaliteit van Saharastof Deeltjes en de Fysieke Eigenschappen Hiervan over de Atlantische Oceaan: Transport Versus Depositie'*, gecombineerd met twee jaar stofconcentraties en korrelgrootte bemonsterd door twee verankerde boeien en passieve MWAC (Modified Wilson and Cooke) samplers. De resultaten laten grote overeenkomsten zien tussen het stof in de atmosfeer en het afgezette stof in de oceaan, maar ook belangrijke verschillen tussen de karakteristieken van stof tijdens transport en na depositie. De sedimentvallen vangen stof dat met zowel de zwaartekracht naar beneden komt (droge depositie), als ook door middel van regen (natte depositie). De sedimentvallen laten zien dat er meer stof wordt afgezet in de zomer, tegelijk met veel neerslag. De boeien vangen alleen het stof tijdens (droog) transport op zeeniveau, en laten geen hogere stofconcentraties in de zomer zien. Dit komt doordat het stof op grote hoogten wordt getransporteerd in de zomer wat buiten het bereik van de boeien valt. Hierdoor lijkt het alsof de grote hoeveelheden van natte depositie in de zomer de seizoenaliteit bepalen van zowel de hoeveelheid stofdepositie alsmede de korrelgrootte ervan.

Strontium, Neodymium en Hafnium isotopen, zeldzame aarden en hun verwantschap met de korrelgrootte van stofdeeltjes, voor zowel de sedimentvallen als de aerosolen die zijn bemonsterd tijdens zeegaande expedities, worden beschreven in **Hoofdstuk 7** *'Noord-Afrikaans Woestijnstof over de Tropische Atlantische Oceaan: Inzichten door Korrelgrootte, Radiogene Sr-Nd-Hf Isotopen en Zeldzame Aarden van Woestijnstof'*. Deze resultaten laten geen relatie zien tussen Sr en Nd isotopen en de korrelgrootte van de stofdeeltjes. Daarentegen is de goede correlatie tussen Hf isotopen en korrelgrootte verwant aan het 'zirkoon effect' omdat grofkorrelige zirkoon mineralen lage $^{176}\text{Hf}/^{177}\text{Hf}$ isotoop ratios hebben. De verschillen in isotoopsamenstellingen tussen de verschillende monsters is te wijten aan de afgelegde afstand vanaf de bron van het stof en de oorsprong van het stof. Dit is van invloed op de mineralogie en korrelgrootte van het bemonsterde stof. Verder is er gekeken naar alternatieve bronnen van mineraaldeeltjes in de meest westelijke sedimentvallen. De invloed van sedimenten afkomstig uit de Amazone Rivier wordt verwaarloosbaar geacht, gebaseerd op de geochemische eigenschappen van sedimenten uit het Amazone bekken en van zijrivieren van de Amazone Rivier. Dit bevestigt dat de primaire bron van sedimenten in de sedimentvallen in het noorden van Afrika ligt. De combinatie van Strontium en Neodymium isotopen laat zien dat de bron van het stof zich in de westelijke regio van Noord-Afrika bevindt.

In **Hoofdstuk 8** ‘*Slotopmerkingen*’ wijd ik uit over mogelijk toekomstig onderzoek gerelateerd aan dit project, over andere analyses die toegepast kunnen worden op de bestaande monsters en veranderingen aan de opstelling van het huidige transect. Ik bespreek het verschil in korrelgrootte langs het transect, dat verschilt per seizoen. Ook laat ik de eerste resultaten van korrelgrootte analyse van stofmonsters van Barbados zien waarmee het huidige transect van sedimentvallen en boeien uitgebreid kan worden. Verder bediscussieer ik de analyse van korrelvorm en de mogelijke uitkomsten en belangen en hoe dit bijdraagt aan de karakterisatie van stofdeeltjes vanaf de bron tot de sedimentatie.

Chapter 1:

Introduction

1.1 Mineral Dust and its Role in the Earth System

Mineral dust is an atmospheric aerosol made up of soil particles emitted from arid and semi-arid regions (Knippertz and Stuut, 2014). Mineral dust impacts the atmosphere as well as oceanic and terrestrial ecosystems in various ways. These impacts can be far reaching as dust can be widely transported over great distances and forms the most important aerosol by mass compared to sea salt, soot, smoke and sulphate particles (Knippertz and Stuut, 2014). First, atmospheric dust can alter the atmospheric radiation budget by scattering incoming short-wave solar radiation resulting in atmospheric cooling (Claquin et al., 2003), and by absorbing the Earth's reflected long-wave radiation causing atmospheric warming (Otto et al., 2007), which in turn depends on the size, shape and mineralogy of the dust particles (Ryder et al., 2013b; Highwood and Ryder, 2014). Changes in dust load and the size of the individual particles will alter the radiation balance and thus surface temperature. Although the magnitude and even the sign of this change in radiation remains uncertain, the unknown amount of large and giant particles ($>74\text{ }\mu\text{m}$; Betzer et al., 1988) may have large effects, which are thus far not incorporated into global climate models (Kok et al., 2017). Dust particles can also act as cloud condensation nuclei and ice nuclei (Shao et al., 2011b; Atkinson et al., 2013; Nenes et al., 2014; Twohy, 2015; Boose et al., 2016), and decrease the surface albedo when deposited on snow and ice (Shao et al., 2011b). All these depend on the optical properties of the dust, the presence of clouds, moisture content, the albedo of the underlying surface, and the amount of dust in the atmosphere and its horizontal and vertical distribution (Goudie and Middleton, 2006).

Mineral dust can enhance the ocean's carbon cycle by delivering important nutrients that stimulate phytoplankton growth and thus the CO_2 drawdown from the atmosphere (Martin and Fitzwater, 1988; Jickells et al., 2014). Several experiments of artificial iron addition to the surface ocean have shown to have largest impacts in high nutrient low chlorophyll (HNLC) regions, although light conditions play a very important role as well (de Baar et al., 2005; de Baar et al., 2008). It is thought that the very low atmospheric CO_2 levels ($\sim 200\text{ ppm}$) during glacial stages over the past 4 million years were the result of increased dust deposition and iron supply to the oceans (Martin, 1990; Martínez-García et al., 2011). Export of the increased amount

of organic particles to the deep ocean is needed for effective carbon sequestration from the atmosphere, which can be limited by increased zooplankton grazing, resulting in recycling of nutrients in the upper ocean (Thiele et al., 2012). On the other hand, dust particles acting as mineral ballast particles can aid the transport of organic particles to the deep (Armstrong et al., 2002; Bressac et al., 2014; Van der Jagt et al., 2018) as virtually no dust particle settles individually, but as part of organic aggregates, sometimes referred to as marine snow. These aggregates >500 μm are composed of organic and inorganic particles and have settling velocities around 200 m day^{-1} which may increase with depth (Berelson, 2002; Nowald et al., 2015) and under high dust deposition conditions (Van der Jagt et al., 2018). Again, particle size is important as this is related to the iron content (Shao et al., 2011b) and ballasting potential (Armstrong et al., 2002; Klaas and Archer, 2002; Fischer et al., 2007; Fischer and Karakas, 2009; Bressac et al., 2014; Van der Jagt et al., 2018). Not only do these nutrients impact the ocean, they also play a role in fertilising the Amazon rainforest (Bristow et al., 2010; Yu et al., 2015b).

Many viable spores, pathogens and microbes can also be transported along with the dust over great distances (Griffin, 2007), that may cause e.g. Black Band disease in corals (De Deckker et al., 2008). Moreover, humans are affected as well, including cardiovascular and respiratory diseases, and increased mortality (Morman and Plumlee, 2014). Atmospheric dust also impacts human traffic as it decreases visibility on roads and airports, and high atmospheric dust loads decrease solar-panel efficiency (Bendetti et al., 2014).

There are many links between dust and precipitation. First, dust transport is a good indicator for aridity in the source area, as dust is easily emitted from semi-arid and arid regions, and low amounts of dust are emitted from vegetated, humid areas (Rea, 2007). Also, there seems to be a relation between the intensity of the preceding north-African rainy season and dust emission from the African continent (Swap et al., 1996) and transport to Barbados in the Caribbean (Prospero and Nees, 1986; Prospero et al., 2002; Prospero and Lamb, 2003). Third, a moist atmosphere can trigger the sedimentation of dust particles, as the dust particles act as condensation nuclei for raindrops (Kalu, 1979). The dust particles absorb water when entering a moist section of the atmosphere and the added water increases the weight of the particles causing them to settle. This depositional process by precipitation, called wet deposition, is the main mechanism of dust deposition over oceans and is about 5-10 times more efficient than gravitational settling in removing dust from the free troposphere (Zender et al., 2003). By gaining a better understanding of the relation between precipitation and dust, precipitation patterns might serve as an indicator of dust emission or deposition. In addition, human activity in the source area, such as land use and deforestation, also influence dust generation and emissions (Prospero and Nees, 1977; Mulitza et al., 2010).

1.2 Saharan Mineral Dust

One of the earliest written scientific reports on Saharan dust observations goes back several centuries. In the 18th century, Matthew Dobson observed hazy skies offshore northwest Africa that accompanied strong and dry easterly winds, called Harmattan (Dobson and Fothergill, 1781). In the 19th century, Charles Darwin observed a hazy horizon, followed by dust being deposited onboard the Beagle (Darwin, 1846). Today we know that the Sahara Desert is the world's largest dust source (Muhs, 2013), accounting for 40 to 60% of all global dust emissions (Maher et al., 2010). Other major dust sources include the Middle East, deserts in China and Mongolia, Australia, and regions in North and South America (Shao et al., 2011b; Muhs, 2013). Annually, about 2000 Tg of dust are emitted into the atmosphere, of which 1500 Tg is deposited on land and 500 Tg over the Ocean (Shao et al., 2011b). Recent Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) observations show that on average 182 Tg of dust is emitted and transported westward over the Atlantic Ocean every year, of which approximately 140 Tg is deposited in the Atlantic Ocean between 15 and 75°W and 10°S and 30°N (Yu et al., 2015a). The region of the Lake Chad Basin in general and the Bodélé Depression in particular is presently the most intense single dust source in the world (Washington et al., 2006). Historical records mention the activity of this region for at least several hundred years (Prospero et al., 2002), and paleo-records suggest even several thousand years (Armitage et al., 2015). Other sources include regions in Mauritania, Mali, southern and northeast Algeria, the Horn of Africa and Djibouti, the Nubian Desert in southern Egypt, Northern Sudan, the Libyan Desert and Western Desert (Sahara el Gharbiya) and Tunisia (Schütz, 1980; Goudie and Middleton, 2001; Prospero et al., 2002; Maher et al., 2010; Scheuven et al., 2013). Source areas are typically associated with aridity, topographical lows and alluvial deposits (Glaccum and Prospero, 1980; Prospero et al., 2002; Muhs et al., 2014). The transport of Saharan dust westward over the Atlantic is by far the most important, but Saharan dust is also transported northward towards Europe (Prospero, 1996; Stuut et al., 2009), eastward to the Middle East and even China and Japan (Tanaka et al., 2005).

1.3 Dust Generation

Wind in arid and semi-arid regions is the main driver of dust emissions, but surface characteristics are important for emission frequency, intensity and spatial distribution (Marticorena, 2014). Soil particles are mobilized in three ways, namely aerodynamic entrainment, saltation and aggregate disintegration (Fig. 1.1) (Shao, 2001; Shao et al., 2011a; Marticorena, 2014). During aerodynamic entrainment dust is lifted directly from the surface, which is generally fine-grained dust (Fig. 1.1A). Larger particles (~100 μm) are transported in a saltating motion, and their impacts on the surface eject more dust particles, a process known as sandblasting (Fig. 1.1B). Aggregate disintegration occurs when large fragments of sand particles coated with dust and clay particles behave as individual particles, and disintegrate when they hit the surface (Fig. 1.1C).

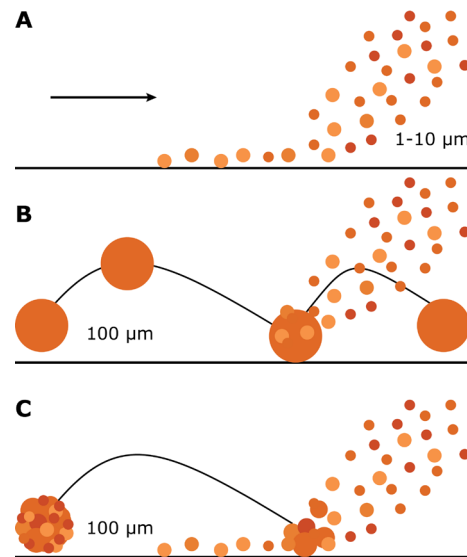


Figure 1.1 Three mechanisms for dust emission. A: Aerodynamic entrainment, B: Saltation and bombardment, and C: Aggregate disintegration. Arrow represents wind direction. After Marticorena (2014).

The minimum wind velocity for the initiation of soil particle movement, or erosion threshold, depends on soil moisture and surface roughness, and on particle characteristics including size, shape and density (Marticorena, 2014). For particles smaller than 100 μm, erosion increases as particle size decreases due to inter-particle cohesive forces (Shao, 2001; Marticorena, 2014). The optimum particle size for wind erosion is ~60 μm, a trade-off between inter-particle cohesion and gravitational force. This makes the wind the most important determining factor for the particle size of emitted dust: increased wind strength increases mobilization of larger and smaller particles, emitting a larger size range of dust particles. This depends in the first place on the size distribution of the source soil and aggregation of individual particles (Marticorena, 2014).

1.4 Dust-Transporting Wind Systems

Dust generation is often associated with hot rising air, but can also be the result of orographic lift (Dubief, 1979). The meteorological processes involved in dust generation and emission range from microscale dust devils to continental-scale dust outbreaks (Knippertz, 2014). The long-range transport of the emitted dust is facilitated by three main wind systems (Fig. 1.2) which are active during different seasons, and transport the dust at different altitudes (Stuut et al., 2005; Muhs, 2013). First, dust is transported year-round from the northwest Sahara to the proximal North Atlantic coastal zone, along the coast between the Canary Islands and the Cape Verde Islands, in the shallow north-easterly trade-wind layer. These winds carry the

dust at low altitudes below 1.5 km, from sources in the region of the Atlas Mountains, the Moroccan coastal region and the northern Sahara (Muhs, 2013). The second main dust-transporting wind system is the Harmattan wind system, transporting dust from the central Sahara as part of the northeast trade winds. This dust transport occurs in highest intensities during boreal winter, from October to May, concurrent with the southward migration of the intertropical convergence zone (ITCZ; Muhs, 2013). These winds occur at low altitudes, between 0 and 3 km over the eastern Atlantic and between 0 and 2 km over the western Atlantic (Muhs, 2013; Tsamalis et al., 2013), at wind speeds up to 25 m s⁻¹ (Sarnthein et al., 1981), and travel over a wide area of western Africa, from Niger to Burkina Faso and from Nigeria to Ghana (Fig. 1.2) (Muhs, 2013). In summer, dust from the Sahara and Sahel is transported in the Saharan Air Layer (SAL), which carries the dust at mid-tropospheric levels above the trade wind zone, with a maximum altitude of 5 to 7 km across the Atlantic Ocean (Muhs, 2013). These winds are created when rising hot air forms large convective cells over the African continent. Upon reaching the west African coast, the warm, dusty air layer is uplifted high in the atmosphere when it is undercut by cool marine air blowing in an opposite direction (Carlson and Prospero, 1972; Chouza et al., 2016). Moving westward over the Atlantic Ocean, the SAL decreases in thickness and altitude until it reaches altitudes of 0-2 km near the Caribbean (Tsamalis et al., 2013). Summer is also the season of maximum dust emission, while in winter dust

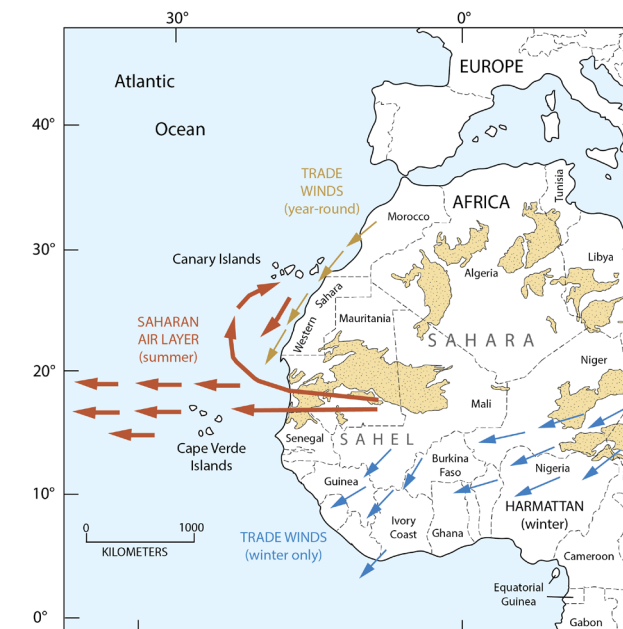


Figure 1.2 Main dust-transporting winds and their season of transport (arrows) and major active dust sources (yellow areas) in north-western Africa. From Muhs (2013), reprinted with permission from Elsevier.

emissions are at a minimum (Adams et al., 2012). The southern border of the trans-Atlantic dust transport is closely linked to the position of the ITCZ (Ben-Ami et al., 2012), and the centre of the main dust cloud migrates seasonally from 10–20°N in summer to 0–10°N in winter (Adams et al., 2012). It takes about a week for dust leaving the African coast to reach the Caribbean (Prospero et al., 1981; Muhs, 2013).

1.5 Characteristics of Saharan Dust Deposition

Dust emissions from the African continent may show large inter-annual variability, and the amount of dust deposited over the ocean is influenced mainly by the distance the dust has travelled, as most dust is deposited closest to the source (Sarnthein et al., 1981; Goudie and Middleton, 2001). Dust particle size shows similar trends, with dust deposited at proximal locations being the coarsest-grained, and the dust deposited far away from the source the finest-grained (Van der Does et al., 2016). This sorting is affected by the mineralogy of the dust, as large, round and heavy quartz and feldspar particles have a greater settling velocity than light platy clay minerals, and are therefore deposited close to the source (Glaccum and Prospero, 1980; Stuut et al., 2005; Goudie and Middleton, 2006; Scheuven et al., 2013; Mahowald et al., 2014; Korte et al., 2017).

Long-range transport of dust is generally assumed to be limited to particle sizes of 20–30 µm (Gillette, 1979; Tsoar and Pye, 1987; Ryder et al., 2013a), and dust transport simulated by climate models is often limited to only 10 µm (Zender et al., 2003; Albani et al., 2014; Smith et al., 2017). However, on many occasions so-called giant (>74 µm) dust particles were found at large distances from the source. Over the Pacific Ocean, individual quartz particles >200 µm were found at >10,000 km from their Asian source (Betzer et al., 1988), and giant Saharan dust particles were found >4,000 km from their source in Europe (Middleton et al., 2001) and over the Atlantic Ocean (Van der Does et al., 2016). This long-range transport is more easily explained for platy clay minerals than for spherical quartz particles. Due to their platy shape and aerodynamical behaviour, clay minerals and mica particles are more easily transported over great distances (Stuut et al., 2005; Scheuven et al., 2013). For more solid particles such as quartz, the long-range transport cannot be explained by the conventional law of settling particles formulated by Stokes (Bagnold, 1941). Dust particles are removed from the atmosphere by dry and wet depositional processes (Bergametti and Forêt, 2014), in the form of gravitational settling, turbulent mix-out, nucleation scavenging and sub-cloud scavenging (Zender et al., 2003).

1.6 Composition of Saharan Dust

The compositional characteristics of dust yields information on the source area of the dust and erosional processes during transport and deposition (Scheuven et al., 2013), for present-day deposited dust as well as dust found in geological archives. Compositional characteristics include dust mineralogy (Caquineau et al., 1998; Kandler et al., 2009; Kandler et al., 2011; Friese et al., 2017), elemental composition

(Moreno et al., 2006; Muhs et al., 2007; Castillo et al., 2008; Bozlaker et al., 2018), biomarkers (Schreuder et al., 2018b), and radiogenic isotopes (Grousset and Biscaye, 2005; Meyer et al., 2011; Abouchami et al., 2013; Pourmand et al., 2014).

The most dominant mineral in Saharan dust samples is quartz, and the amount of quartz in such a sample ranges from 15 to 80 wt% (Scheuven and Kandler, 2014). The fine fraction (<2 µm) is dominated by clay minerals, of which illite and kaolinite generally contribute more than 50% (Scheuven et al., 2013). The total amount of clay minerals in a Saharan dust sample varies significantly, from as large as 60 wt%, down to nearly 0 wt% (Scheuven and Kandler, 2014). Other important dust minerals include chlorite, microcline, palygorskite, and calcite (Glaccum and Prospero, 1980; Prospero et al., 1981; Scheuven et al., 2013). Feldspar minerals are mostly present but in smaller amounts, up to 10 wt% (Scheuven and Kandler, 2014).

The compositional patterns of Rare Earth Elements (REE) reveals information about dust sources, and depletion or enrichment of specific elements resulting from natural or anthropogenic processes can be traced to particular source regions (Meyer et al., 2011; Meyer et al., 2013; Scheuven et al., 2013). Isotopic ratios of Neodymium and Strontium in mineral dust yields information on weathering and the dust's provenance, while Hafnium isotopic composition informs about dust transport and deposition (Scheuven et al., 2013).

1.7 Saharan Dust in Sedimentary Archives

In sedimentary archives, including deep-sea sediment cores, ice and snow cores, in lakes, soils, peat bogs and in loess deposits, dust can be used as an indicator of past conditions like aridity, wind strength and provenance, involved in the atmospheric transport of dust (Rea, 2007; Muhs, 2013). Especially in the tropical Atlantic Ocean, where most of the terrigenous fraction of the marine sediments is of aeolian origin (Skonieczny et al., 2011). Dust deposition fluxes may serve as a paleoclimatic indicator for aridity or humidity in the source area (Tjallingii et al., 2008; McGee et al., 2013), and dust particle size can be related to past wind strength (Rea, 1994; Stuut et al., 2002). Studies have shown that dust emission and deposition varied greatly through time, with dust fluxes two to five times higher in glacial stages than in interglacial stages (Rea, 1994; Winckler et al., 2008; Maher et al., 2010). Dust particle size was coarser during the Last Glacial Maximum (18,000 yrs BP), related to increased wind strength of up to 40% compared to today (Sarnthein et al., 1981). Even correlations between Milankovitch cycles of orbital forcing and dust flux and particle size were found on timescales of thousands of years (Rea, 1994; Mulitza et al., 2008).

Next to natural relationships between climate and dust emissions they are also influenced by anthropogenic processes. In the 19th century, North African dust emissions increased parallel to the onset of commercial agriculture in the Sahel region, which led to increased erosion and thus the deflation of dust (Mulitza et

al., 2010). Oppositely, signs of human occupation and migration in the Sahara and Sahel regions are an indicator for changing climate conditions to drier or more humid periods throughout the Holocene (Kuper and Kröpelin, 2006). During humid periods, mega lakes existed in the Sahara Desert, of which palaeolake Megachad was the largest (Drake and Bristow, 2006). After this lake dried out, the fine sediments became available for airborne transport and the lakebed now makes the largest Saharan dust source, with the Bodélé Depression being part of it (Washington et al., 2006). The lithogenic fraction in offshore sediments generally consists of a mixture of dust particles and fluvial sediments, related to different climatic stages in the past. These different sedimentary inputs can be separated by endmember modelling on the basis of particle size (Weltje and Prins, 2003; Holz et al., 2007; McGregor et al., 2009; McGee et al., 2013) and used to identify different sources of dust (Prins et al., 2009).

Despite advances in dust research, our level of scientific understanding of the dust cycle is still low (IPCC, 2013). New and improved insights should not only aid the interpretation of geologic archives, but also improve dust forecasting and climate predictions (Bendetti et al., 2014). Global and regional climate models including a dust component rely on quantitative *in-situ* observations for validation. At present, most observations of Saharan dust transport and deposition are restricted to the west African coastal area (Honjo et al., 1982; Bory et al., 2002; Neuer et al., 2004; Brust et al., 2011; Fischer et al., 2016; Friese et al., 2016), the Caribbean (Prospero and Lamb, 2003; Prospero et al., 2014) and particular regions in North and South America (Prospero, 1999; Prospero et al., 2010; Yu et al., 2015b). Often these observations are indirect, using proxies for dust transport and deposition or satellite observations, and there is a great lack of knowledge on Saharan dust transport and deposition across the entire Atlantic Ocean.

1.8 TRAFFIC – Trans-Atlantic Fluxes of Saharan Dust

1.8.1 Project Set-Up

Several times sediment traps have been used to study Saharan dust deposition in the Atlantic Ocean. A more than 20-year long record of sediment trap-collected dust offshore northwest Africa reveals close relations between phytoplankton biomass production and dust deposition (Fischer et al., 2016), and highest dust deposition fluxes are received at this site in winter (Ratmeyer et al., 1999b). Offshore the Mauritanian coast, sediment traps were analysed for aluminium content, from which the dust fraction was estimated (Bory and Newton, 2000). Their results showed that the flux of biogenic matter influences dust export to the deep ocean (Bory et al., 2002). They found that model estimates of atmospheric dust input to the ocean are two to three times lower than actual deposition fluxes, and attributed that to the under-representation of large dust particles in models and surface redistributions of dust before settling.

Torres Valdés et al. (2014) inventoried numerous sediment-trap sampling studies from the Atlantic Ocean. Despite many studies along the northwest African coast,

hardly any have been carried out in the central North Atlantic Ocean. Consequently, observational data on dust deposition across the Atlantic Ocean are virtually non-existent. This lack of crucial data led to the deployment of a series of sediment traps along a trans-Atlantic transect (Stuut et al., 2012) directly underneath the main path of the Saharan dust plume (Mulitza et al., 2010; Adams et al., 2012; Yu et al., 2015a). Four moorings were spaced at regular intervals across this transect (M1 at 23°W, M2 at 38°W, M4 at 49°W, and M5 at 59°W), and a fifth (M2 at 39°W) was moored at 13°N (Fig. 1.3A). In addition, two moored dust-collecting surface buoys were added to two stations, M3 and M4 (Fig. 1.3A). This trans-Atlantic transect provides the opportunity to determine the seasonal and spatial fluxes in Saharan dust deposition, related to dust emissions and transport. The transect is located within the North Equatorial Current (NEC), flowing from the east Atlantic to the Caribbean Sea (Bub and Brown, 1996; Bourles et al., 1999) and linked to the Canary Current (CC) in the east, which flows southward along the northwest African coast (Mittelstaedt, 1991).

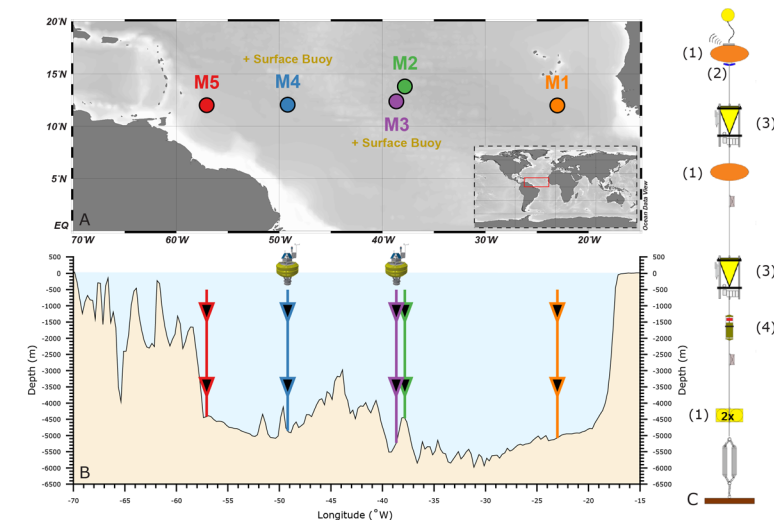


Figure 1.3 A: Map with sampling stations M1–M5 at 12°N. B: Bathymetry along 12°N with sediment traps at 1200 and 3500 m below sea level, and two dust-collecting surface buoys at M3 and M4. C: Set-up of mooring anchored to seafloor, containing floats (1), current profiler (ADCP; 2) sediment traps (3) and current meter (4).

1.8.2 Sediment-Trap Moorings

Each mooring was originally equipped with two sediment traps, located at approximately 1,200 and 3,500 meters below sea level (Fig. 1.3C). The lower traps were situated 880–1,300 meters above the ocean floor, and thus far away from sources of resuspended sediments. The traps are Technicap model PPS 5/2 and consist of a conical funnel (36°) with a collecting area of 1 m² (Fig. 1.4). On top is an 8 mm hexagonal baffle to maximize particle collection (Knauer and Asper, 1989), also preventing large swimmers entering the sediment trap. The 24 sampling

cups on each sediment trap are located on a rotating carousel underneath the funnel. All sediment traps were pre-programmed to sample synchronously at 8 to 22 day intervals, depending on the planning of the recovery cruise. In addition to the sediment traps, the moorings were equipped with current meters, acoustic Doppler current profilers (ADCPs), tilt meters and CTDs, to monitor current speeds and collection efficiency, and several floats to keep the mooring upright when deployed and to bring them back to the surface when serviced (Fig. 1.3C). The moorings were serviced on a yearly basis, however due to various technical failures and logistical problems the original set-up of a total of ten sediment traps on five moorings could not always be reproduced (Stuut et al., 2012; 2013; 2015; 2016).

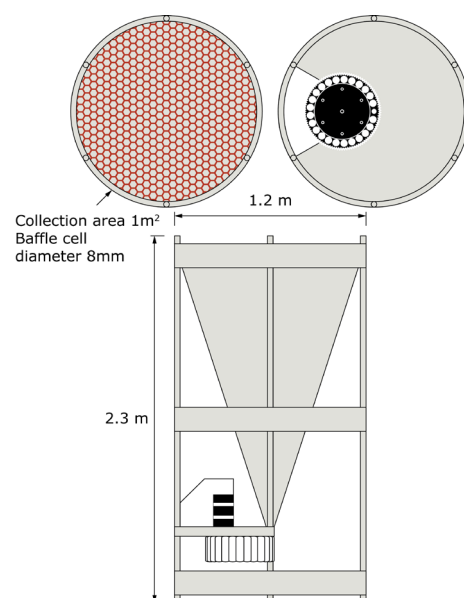


Figure 1.4 Top view, bottom view and side view of Technicap PPS 5/2 Sediment trap, with a 1 m² collection area and 24 sampling cups on a rotating carousel.

Prior to sediment-trap deployment, the sampling cups were filled with ambient seawater, to which a biocide (HgCl_2) and a pH buffer (Borax, $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$) were added, to create a density slightly in excess of the ambient seawater. After recovery of the sediment trap, 40 mL of the supernatant solution was taken from each bottle for nutrient analysis (ammonia, phosphate and silica), and for pH analysis. The height of the sediments in the collecting bottles was measured for a first-order estimate of the total mass flux. Subsequently the samples were stored at 4°C for transport to and storage at NIOZ, prior to further processing and analysis.

1.8.3 Moored Dust-Collecting Surface Buoys

Two moored dust-collecting surface buoys (nicknamed Michèle and Laura; Fig. 1.5) were added to the transect in 2013 at stations M3 and M4 (Fig. 1.3). They collected aerosols by pumping air through 47 mm polycarbonate filters with a pore size of 0.2 μm . These were placed on a carousel with a total of 24 filters, rotating synchronously with the sediment traps below at 8–22 day intervals. The buoy's sampler was linked to a meteorological sensor, that monitored wind speed, wind direction, temperature and humidity. The filters are placed one-by-one under the air inlet through which an air pump sampled for two hours (in 2014) and four hours (in 2015) under favourable weather conditions, between 10:00 UTC and 18:00 UTC. Favourable weather conditions were determined by the attached meteorological sensors, and are defined by no precipitation ($<0.2 \text{ mm min}^{-1}$) or strong winds ($<20 \text{ m s}^{-1}$). During unfavourable weather conditions the air inlet remained closed while the weather kept being monitored. Sampling was initiated when weather conditions changed to favourable conditions for at least one hour continuously, and was again aborted when conditions changed to unfavourable. The buoys were equipped with a wind vane to keep the air inlet facing the wind, which was mostly blowing from an easterly direction. They also contained a satellite communication system (iridium) that allowed two-way communication with the buoys, and a GPS sensor for an accurate position of the buoys. Solar panels and batteries powered the entire systems. Both buoys were deployed in 2013 (Stuut et al., 2013), and serviced and redeployed in 2014 (Stuut et al., 2015) and 2016 (Stuut et al., 2016). Buoy M3 was deployed an additional time in 2014 (Bale, 2014) as it had to be serviced due to a malfunctioning of the dust sampler and its meteorological station.

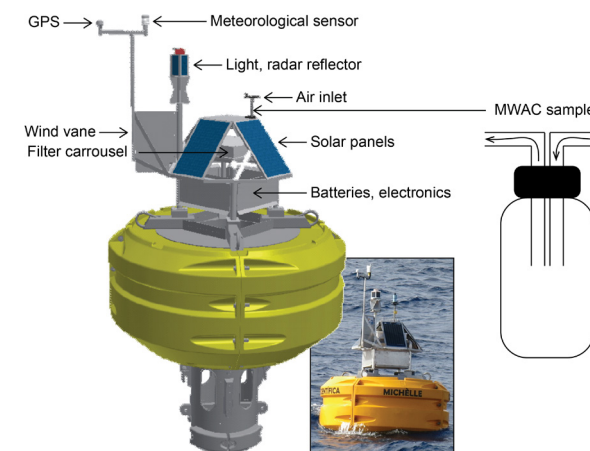


Figure 1.5 Drawing and photo of dust-collecting buoy (left) and MWAC sampler (right).

1.8.4 MWAC Samplers

The buoys were additionally equipped with an MWAC (Modified Wilson and Cooke; Goossens and Offer, 2000) passive air sampler (Fig. 1.5) that sampled continuously during the entire deployment period of the buoys. This sampler consisted of a plastic bottle acting as a settling chamber, connected to an inlet and outlet tube with a 7.5 mm diameter. These were installed vertically on the buoy's air inlet about 3 meters above sea level, while a wind vane ensured continuous windward orientation of the buoys and MWACs. The MWAC samplers have high sampling efficiencies between 75 and 105% (Goossens and Offer, 2000) for dust with a 30 μm median particle size, and slightly higher efficiencies (90–120%) for sand-sized particles with a median grain size between 132 and 287 μm (Goossens et al., 2000).

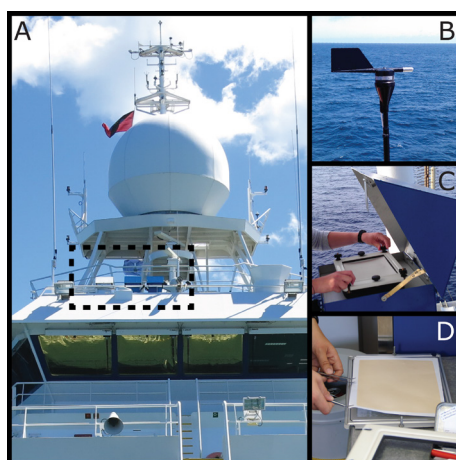


Figure 1.6 A: Dust collectors on top of the ship's bridge. B: Wind vane ensuring the sampling of frontal winds only. C: Under the hood of the dust collector is the sample filter. D: A filter filled with orange dust.

1.8.5 Shipboard-Collected Aerosol Samples

Aerosol samples were collected during all seagoing trans-Atlantic research expeditions with Anderson high-volume dust collectors, mounted on deck above the bridge of the ship (Fig. 1.6A). It used letter-sized Whatman Type 41 Cellulose Acetate filters, which are placed under a protective cover (Fig. 1.6C–D). The samplers were connected to a wind vane (Fig. 1.6B) which switched the samplers off when winds exceeded an angle of 90° from the ship's heading to prevent contamination from the ship's exhaust behind the sampler. A data logger monitored the volume of air pumped through the filters and increased pump power as the filter became loaded with particles to maintain a constant air flow. Samples were collected during regular intervals of 24 hours, but the sampling interval was increased under dusty conditions to obtain a higher temporal

resolution during dust-rich conditions. After removal of the filters from the dust sampler they were folded and stored in a plastic bag at room temperature.

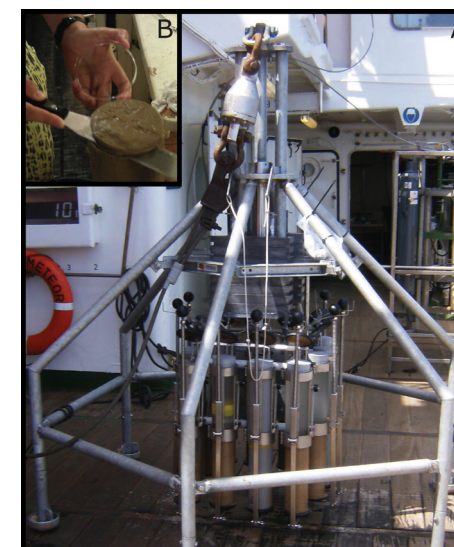


Figure 1.7 A: Multicorer after sampling, with tubes filled with sediments. B: Sampling of the top centimetre.

1.8.6 Seafloor Surface Sediments

Seafloor surface sediments were collected in 2012 using a multicorer at each of the five sediment-trap stations (Stuut et al., 2012; Van der Does et al., 2016), to recover the sediment-water interface and undisturbed sediments from the seafloor. The multicorer comprised 12 tubes with a length of 60 cm and a 10 cm diameter each (Fig. 1.7). Upon recovery this resulted in 20–33 cm-long sediment cores, some of which were sliced into 1 cm slices and stored in petri dishes. Others were stored intact at 4°C and -20°C for further analysis. For the analysis of seafloor surface sediments only the top centimetre of a core was used.

Chapter 2:

Particle-Size Analysis Methods

2.1 Sample Preparation and Pre-Treatment

2.1.1 Sediment-Trap Samples

The sediment-trap samples were first sieved through a 1 mm mesh to remove zooplankton swimmers, and subsequently wet-split in five aliquots using a WSD10 rotor splitter (McLane Laboratories, USA). For grain-size analysis, one of these aliquots was split into another five subsamples (1/25 of the original trap sample). The samples were then washed and centrifuged three times at $1800 \times g$ with Milli-Q water to remove HgCl_2 , borax and sea salts. Besides grain-size and dust-flux analysis, the sediment-trap samples were used for the analysis of total mass, biogenic contents (biogenic silicates, carbonates, organic matter), carbon and nitrogen isotopes, major element geochemistry using X-ray fluorescence (XRF), Sr-Nd-Hf isotopes and Rare Earth Elements (REE), biomarkers, pigments, (bacterial) DNA, pollen, chemical components by sequential leaching, zooplankton including pteropods and specific biomineral assemblages of foraminifera as well as coccolithophores (e.g. Guerreiro et al., 2017; Korte et al., 2017; Schreuder et al., 2018a; Schreuder et al., 2018b, Van der Does et al., 2018).

The sediment traps collected a wide range of settling particles besides mineral dust, including the skeletons of marine plankton (foraminifera, radiolarians, pteropods, diatoms, coccolithophores, etc.) and organic matter (marine and atmospheric from biomass-burning aerosols). To isolate the dust fraction, these biogenic components were chemically removed in three steps. First, organic matter was oxidized by adding 10 mL H_2O_2 (35%) to the wet sediment-trap sample, boiling the mixture until the reaction stopped, and by continuously adding demineralised water to prevent the sample to boil dry. Next, the sample volume was increased to 100 mL with demineralised water, and 10 mL HCl (10%) was added to dissolve biogenic carbonates. The sample was boiled for exactly 1 minute, filled up to 1000 mL and set aside to cool and let the particles settle in a few hours. When the overlying fluid was clear, the sample was decanted to 100 mL, filled up with demineralised water to 1000 mL once more and left to settle again

overnight. The following day the samples were decanted to 100 mL and 6g NaOH pellets were added to remove the biogenic silicates. The sample was boiled for 10 minutes, and filled to 1000 mL twice as in the preceding treatment step. After settling the sample was decanted in several steps, with several hours of settling in between, to a volume of approximately 5 mL before particle-size analysis.

2.1.2 Buoy Filters and MWAC Samples

Dust and sea salts were removed from the original 47 mm, 0.2 μm polycarbonate filters by ashing them in a low temperature asher and subsequent rinsing with Milli-Q water over pre-weighed filters (25 mm polycarbonate with a pore size of 0.4 μm). After determining the dry weight of the collected dust, these filters were ashed again to isolate the dust for particle-size analysis. The MWAC samples only required the rinsing of the dust from the sample bottle with Milli-Q water prior to grain-size analysis, at the same time dissolving any salts present in the samples.

2.1.3 Shipboard-Collected Aerosol Samples

The dust was removed from the filters of shipboard-collected dust in two ways. First, some filters were treated by simply washing off the dust from the filter. This was done by first cutting off a small strip from the letter-sized filter and placing this strip on a small microscope sample glass. The dust was subsequently rinsed off with Milli-Q water from a spray bottle with increased force, following the procedure of Stuut (2001). To ensure a more quantitative removal of dust particles from the filters, a second method was employed. A circular piece 4.4 cm in diameter was cut from each filter and ashed in a low temperature asher to remove the filter and isolate the dust. After grain-size analysis revealed there might still be some filter fibres in the dust samples, the samples were treated with a small amount of H_2O_2 to completely remove any remaining filter parts from the samples.

2.1.4 Seafloor Core Top Sediments

Of the top centimetre of each core a small sample (~2 g) was taken for particle-size analysis. For reproducible and representative results, sub-samples of different cores from the same multi-core deployment were taken. The preparation for particle-size analysis was carried out in the same way as for the sediment-trap samples, i.e. chemical removal of organic matter with H_2O_2 , biogenic carbonates with HCl, and biogenic silicates with NaOH.

2.2 Particle-Size Analysis with Coulter LS13 320

Particle-size analysis for this study was conducted with a Coulter laser diffraction particle sizer (*Beckman Coulter LS13 320*) in the setup with the micro liquid module (MLM) for the analysis of small-volume samples (12 mL). Particles in suspension move in front of a laser beam and scatter the light, which is then measured by the detectors. The angle at which the light is scattered is related to the particle's size, with a larger scattering angle for smaller particles.

Immediately prior to analysis a few drops of 1M sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$) were added to ensure complete disaggregation of particles. During analysis, samples were homogenized using a magnetic stirrer, and the influence of air bubbles was minimized by using degassed water. This method was developed to prevent a so-called “ghost peak” (McCave et al., 1995; Ratmeyer et al., 1999b) in the grain-size distributions, which result from air bubbles being detected as particles by the laser particle sizer. The system, called BEX (bubble extirminator, developed at NIOZ), exposes the water to a vacuum which removes the dissolved gases from the water. Analysis was run for 90 seconds, and the resulting particle-size distributions range between 0.4 and 2000 μm .

Reproducibility was checked regularly by J. Titschack at MARUM (Bremen, Germany), where the exact same equipment and setup, including the BEX system, is used. Replicate analyses with three internal glass-bead standards show that the reproducibility is better than $\pm 0.7 \mu\text{m}$ for the mean and $\pm 0.6 \mu\text{m}$ for the median particle size (1σ). The average standard deviation integrated over all size classes is better than $\pm 4 \text{ vol}\%$. In addition, analysis of three individually sampled (from different tubes of a multicorer) and pre-treated (isolation of the lithogenic fraction) core-top samples from the five stations M1–M5 revealed a high reproducibility, with the average standard deviation over all size classes being $\pm 7 \text{ vol}\%$ (Fig. 2.1).

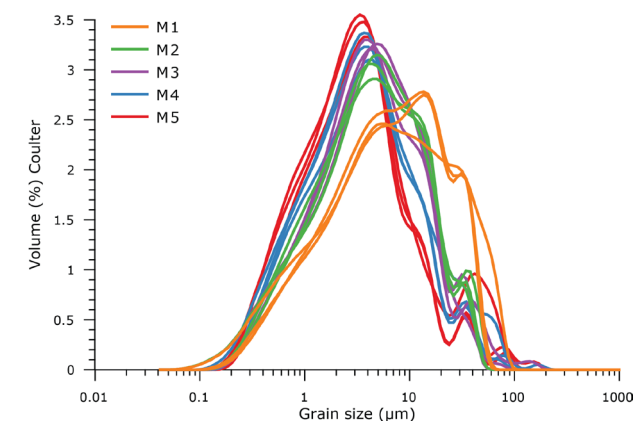


Figure 2.1 Grain-size distributions of three replicate samples from seafloor sediment core-tops sampled at the five mooring stations M1–M5, measured with the Coulter laser particle sizer at NIOZ.

2.3 Particle-Size Analysis with Sympatec QicPic

Besides analysis with the *Coulter LS13 320*, particle-size distributions as well as particle shape of five core-top samples were determined by a *Sympatec QicPic* at VU University in Amsterdam. This instrument uses image analysis, which primarily measures particle contours (Fig. 2.3). This way of measurement gives additional information on the two-dimensional shape of the particle. The particle size (1.95 to

2000 μm) is calculated using the mean of Feret diameters in all directions, which are derived from the distance of two tangents to the contour of the particle (Fig. 2.2C).

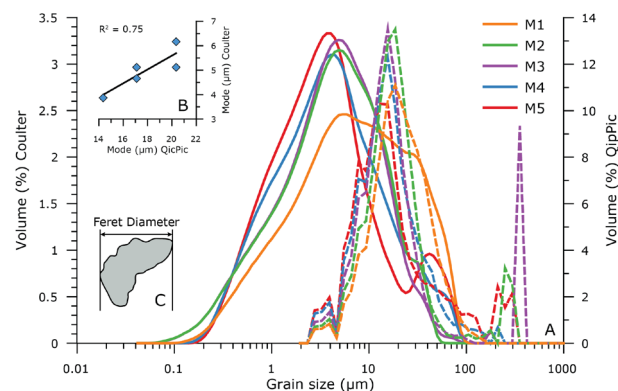


Figure 2.2 A: Grain-size distributions of seafloor sediment core-tops sampled at the five mooring stations M1–M5 as measured by the Coulter laser particle sizer and Sympatec QicPic. B: The linear relation between the modal grain sizes of both methods. C: Definition of the Feret diameter used by the Sympatec QicPic.

Comparison of the two methods of particle-size analysis reveals that the Coulter laser particle sizer is more appropriate for mineral dust samples, as the Sympatec QicPic is unable to analyse particles smaller than 2 μm , which make up a large part of the dust samples (Fig. 2.2A). However, also the Coulter laser particle sizer is slightly biased as it seems to overestimate the fine fraction (Konert and Vandenberghe, 1997). At the coarse end of the grain-size distribution the QicPic also performs less accurately, which is visible as large peaks at $>200 \mu\text{m}$. This is probably caused by contamination in the lab, seen as thread-like particles from the image analysis (Fig. 2.3A), as the QicPic measures the sample from a beaker which is in open contact with the air in the lab, whereas the Coulter laser particle sizer measures the sample from the MLM, which is in a closed sample chamber. To compare modal grain sizes of the two methods, the data was regrouped in similar size classes, which was done geometrically using the Folk and Ward graphical method (Folk and Ward, 1957), with the program *GRADISTAT* (Blott and Pye, 2001). Comparing the modal grain sizes of both methods shows a good linear relation ($R^2 = 0.75$; Fig. 2.2B) but differences can be expected since both methods use different size classes prior to regrouping. This systematic offset was also found by Shang et al. (2017), who employed the same method for analysis with QicPic. Both methods show a similar down-wind fining trend of dust particle size despite their absolute differences. A benefit of the QicPic method is that it also counts the number of particles in a specific size class, instead of a volume percentage that results from laser diffraction grain-size analysis. This can give additional statistical insights as particles of a specific size can be expressed as parts per million (ppm).

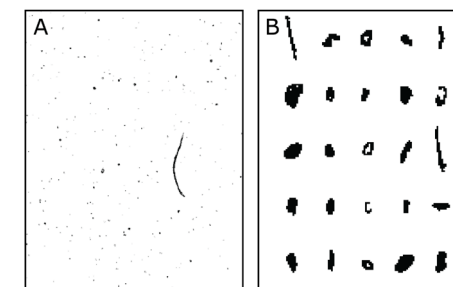


Figure 2.3 A: Particles as observed by the QicPic, with one large thread-like particle. B: Individual randomly-selected particles observed by the QicPic.

2.4 Particle-Size Analysis with Coulter Multisizer 3

Particle-size sediment standards were analysed on a Coulter “Counter” Multisizer 3 (MS3) at the Rosenstiel School for Marine and Atmospheric Science (RSMAS) at the University of Miami, and compared to measurements by the Coulter laser particle sizer (LS13 320) at NIOZ. During analysis, particles are suspended in an electrolyte and pass through an aperture between two electrodes. This causes a voltage pulse, which is proportional to the particle’s volume. In this way, the diameter of a particle can be estimated, assuming spherical particles.

Tests were performed with different stirring speeds and background measurements on the MS3 using a 100 μm aperture tube. Three standards were measured: one 15 μm garnet standard from Coulter and two glass-bead standards made at NIOZ; Ballotini A (median: 39.1 μm , mode: 41.7 μm) and Ballotini B (median: 61.5 μm , mode: 72.9 μm). Both methods show a good reproducibility for the 15 μm garnet standard (Fig. 2.4A). If an offset between different methods is consistent it can be used to normalise results from both methods to allow direct comparison. However, the MS3 was unable to properly measure the Ballotini A and B standards (Fig. 2.4B). In theory, the 100 μm aperture tube of the MS3 should be able to reliably measure particles up to 60 μm , although the results show that it is unable to measure particles larger than 42 μm (illustrated as the cut-off of the glass-bead standards at 42 μm in Fig. 2.3B). This makes the Coulter MS3 unsuitable for the particle-size analysis of the Saharan dust samples collected for this study. By comparing these three methods of particle-size analysis, the Coulter laser particle size analyser appears as the instrument best suited for the analysis of Saharan mineral dust particles.

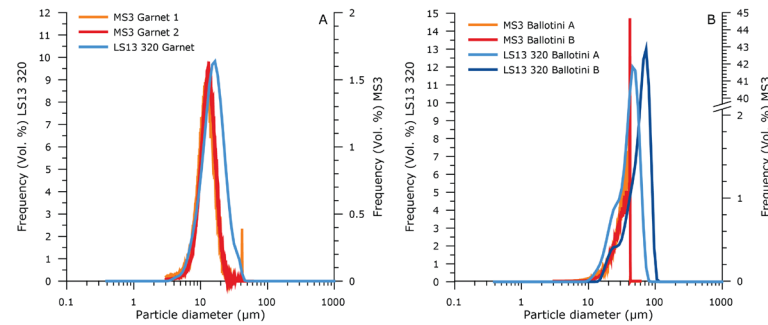


Figure 2.4. A: Particle-size distributions of 15 μm garnet Coulter-standard. B: Ballotini A & B NIOZ standards. Measurements performed on Coulter Counter Multisizer 3 (MS3, RSMAS; yellow and red lines) and Coulter laser particle sizer (LS13 320, NIOZ; blue lines).

Chapter 3:

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.

3.1 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., 2015a). 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; Wilcox et al., 2010; Shao et al., 2011b; Highwood and Ryder, 2014). Climatic effects are largely determined by particle characteristics, including particle size, particle shape, chemical and mineralogical composition, and cloud cover and the albedo of the underlying surface (Sokolik and Toon, 1999; Goudie and Middleton, 2001; Claquin et al., 2003; Goudie and Middleton, 2006; Otto et al., 2007; Shao et al., 2011b; Highwood and Ryder, 2014). Large particles in the lower atmosphere may have a warming effect on earth's climate by absorbing reflected (longwave) radiation (Otto et al., 2007; Mahowald et al., 2014). By contrast, small particles in the higher atmosphere may have a cooling effect, by reflecting incoming solar (shortwave) 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., 2011b). 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; Klaas and Archer, 2002; Fischer et al., 2007; Fischer and Karakas, 2009; Bressac et al., 2014). Both have the potential to reduce atmospheric $p\text{CO}_2$ 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; Stuut et al., 2005; Goudie and Middleton, 2006; Mahowald et al., 2014). Saharan dust is transported with the trade winds year-round, from the north-western 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; Prospero and Carlson, 1972; Adams et al., 2012; Tsamalis et al., 2013; Kanitz et al., 2014). 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 (Moulin et al., 1997; Holz et al., 2004; Adams et al., 2012; Yu et al., 2015a).

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, and density, as well as 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 (Schütz, 1980; Sarnthein et al., 1981; Holz et al., 2004; Mahowald et al., 2014) and generally do not exceed 20 μm when transported over long distances (Gillette, 1979; Tsoar and Pye, 1987). On the Cabo 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; Middleton et al., 2001; Goudie and Middleton, 2006; Mahowald et al., 2014). 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 but 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., 2015a). 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.

3.2 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. 3.1A). Each mooring is equipped with two sediment traps, at depths of 1,200 and 3,500 meters below sea level (BSL), or “upper” and “lower”, respectively (Fig. 3.1B, Table 3.1). 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 8 mm 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 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, 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 acoustic Doppler current profilers, 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 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 (1,200 m) sediment traps located at mooring stations M1, M2, and M4, and four lower (3,500 m) sediment traps at stations M2, M3, M4, and M5 (Fig. 3.1, Table 3.1). Three of the 10 sediment traps could not be recovered. In addition, seafloor sediments were collected by a multicorer at all five mooring stations, using the top centimetre for comparison with the sediment-trap samples.

Table 3.1. Locations and depths of the sampling stations M1–M5. BSL = below sea level.

Station	Latitude (°N)	Longitude (°W)	Trap depths (m BSL)	Bottom depth (m BSL)	Distance to African coast (km)
M1	12.00	23.00	1,150	5,000	700
M2	13.81	37.82	1,235 3,490	4,790	2,300
M3	12.39	38.63	3,540	4,640	2,400
M4	12.06	49.19	1,130 3,370	4,670	3,500
M5	12.02	57.04	3,520	4,400	4,400

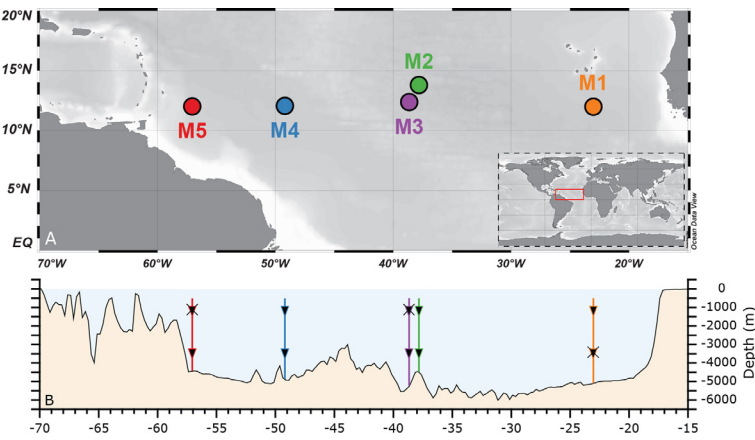


Figure 3.1. A: Map with sampling stations M1–M5 in the Atlantic Ocean at 12°N. B: Bathymetry along 12°N (from www.gebco.net) with sediment traps at 1,200m and 3,500m BSL. Crossed-out sediment traps could not be recovered.

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. 3.2). 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, 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 $>500 \mu\text{m}$ 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.

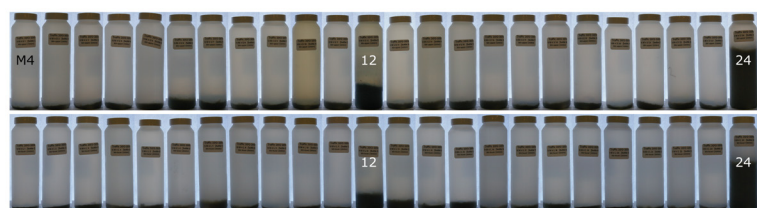


Figure 3.2. Sediment-trap bottles of the upper (1,200 m) and lower (3,500 m) traps at station M4 (12°N, 49°W).

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 880–1,300 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., 2015a), 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.

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), which were washed and centrifuged repeatedly at approximately $1800 \times g$ with Milli-Q water to remove the HgCl_2 , borax, and sea salts.

The sediment traps collect all particles settling down into the ocean, which 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). Briefly, 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. 3.5). 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 analysed, they can be compared directly to each other. Immediately prior to the grain-size measurements sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$) 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. In order to minimize the influence of air bubbles, the system uses degassed water. This resulted in particle-size distributions consisting of 92 size classes ranging from 0.375 to $2,000 \mu\text{m}$ 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 setup as in this study, by J. Titschack. This is performed by replicate analyses of three internal glass-bead standards, and the reproducibility is found to be better than $\pm 0.7 \mu\text{m}$ for the mean and $\pm 0.6 \mu\text{m}$ for the median particle size (1σ). The average standard deviation integrated over all the size classes is better than $\pm 4 \text{ vol } \%$.

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 the samples refer to are the middle date of each 16-day sampling period.

In order to determine the provenance of dust-carrying air layers, 4-day backward trajectories of air parcels were calculated with the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015), using the GDAS (0.5°) meteorological dataset (<http://www.ready.noaa.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).

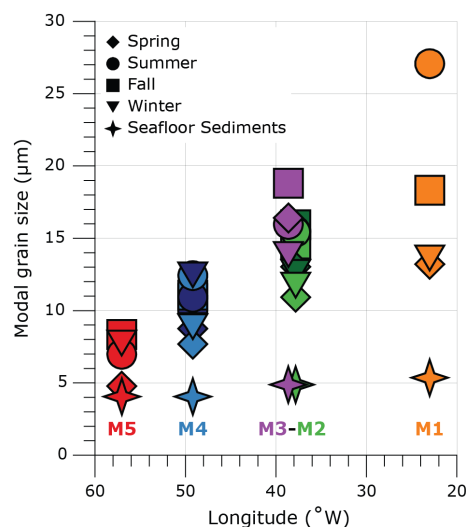


Figure 3.3. Downwind fining and seasonality in average modal grain size per season for all seven traps (an average of all modal values from the relative volume grain-size distributions, grouped per season), for October 2012–November 2013, and modal grain size of the seafloor sediments (from the grain-size distributions as show in Figure 3.4B), versus western longitude. For M2 (green) and M4 (blue), lighter colours indicate the upper (1,200 m) trap, and darker colours indicate the lower (3,500 m) trap.

3.3 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 in order to highlight the coarse second peak, or “shoulder”, of the grain-size distributions.

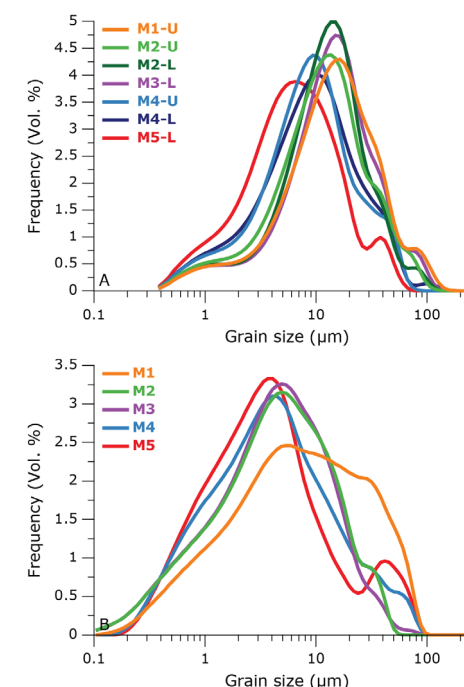


Figure 3.4. A: Average volume grain-size distributions of 24 samples from all seven sediment traps, where U = upper trap (1,200 m) and L = lower trap (3,500 m). Collected between October 2012 and November 2013. B: Grain-size distributions of seafloor sediments at the five mooring stations (M1–M5) along the trans-Atlantic transect.

3.3.1 Spatial Trends in Grain Size

Modal grain sizes of the sediment traps and seafloor sediments show a pronounced downwind fining (Fig. 3.3). 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. 3.3). 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. 3.4A). 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. 3.4B). They include “giant” particles of more than 100 μm , which are observed as far west as station M5 (57°W; approximately 4,400 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. 3.5). Furthermore, the average grain-size distributions show that the differences between stations are larger than between the upper (1,200 m) and lower (3,500 m) traps at stations M2 and M4 (Fig. 3.4A). 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. 3.4B).

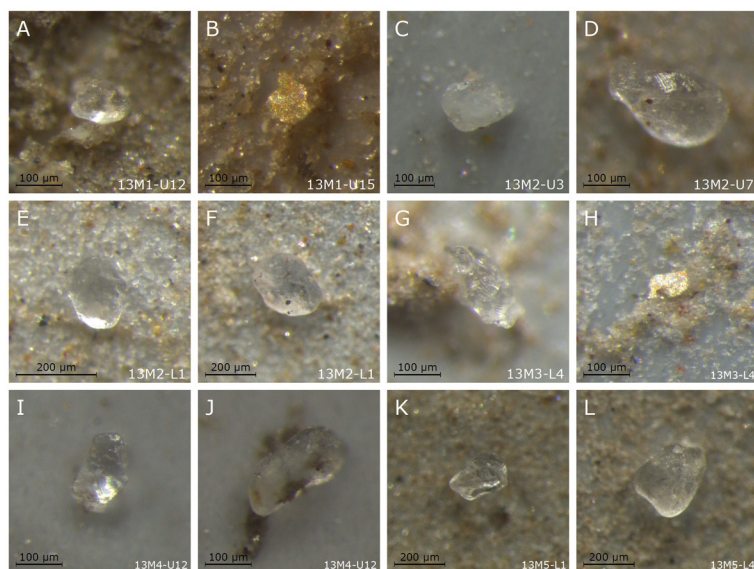


Figure 3.5. Light-microscope images of large dust particles from the sediment trap samples. Name in lower-right corners: 13Mx denotes the station (see Table 3.1 for the exact locations). U and L denotes Upper (1,200 m) and Lower (3,500 m) traps, respectively, followed by the sample number (out of a total 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).

3.3.2 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. 3.6A). During spring, a coarse shoulder is present in the grain-size distributions

(Fig. 3.6B), 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. 3.7A) 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.

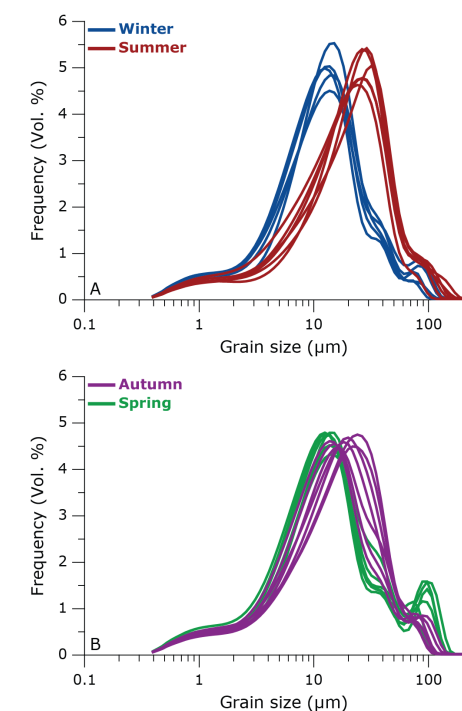


Figure 3.6. Seasonal grain-size distributions at station M1 (12°N, 23°W; approximately 700 km from the African coast) for October 2012–November 2013, of A: winter (blue), summer (red), B: fall (purple) and spring (green).

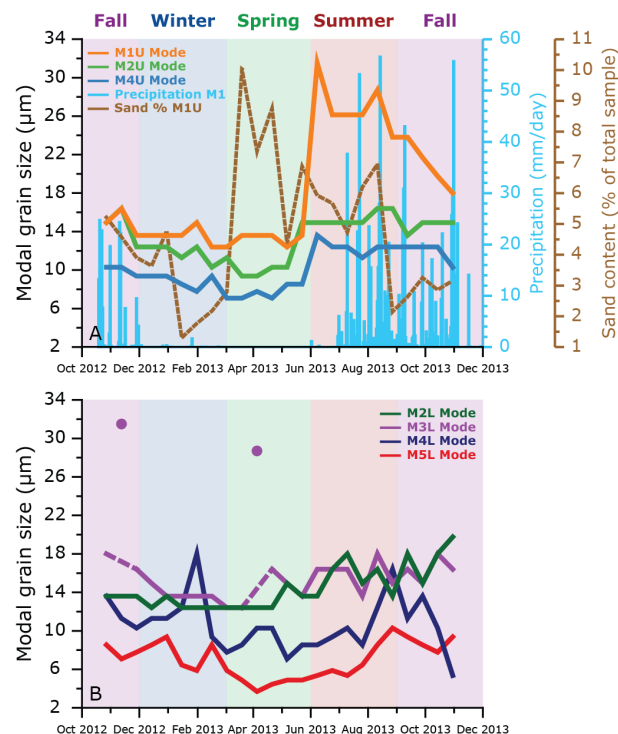


Figure 3.7. A: Modal particle diameter of dust collected by the three upper (1,200 m) sediment traps at stations M1, M2 and M4 (left axis), daily precipitation (right axis; TRMM) and percentage of sand particles ($>63 \mu\text{m}$) (far right axis) at station M1. B: Modal particle diameter of dust collected by the three lower (3,500 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. 3.7A). However, the difference in particle size between these seasons is greatest at M1, close to the source (Fig. 3.3). 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 1,200 m than in the four lower sediment traps at 3,500 m (Fig. 3.7B). 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. 3.7B). From summer 2013 onwards, the modal grain size of M2 and M3 converge, with synchronous fluctuations between 14 and $18 \mu\text{m}$. Seasonality at M4 is even weaker, with grain sizes varying between 5 and $18 \mu\text{m}$. At the westernmost station M5 modal particle size ranges between 4

and $10 \mu\text{m}$, with a decrease in spring 2013 and an increase in summer. In all seven traps, dust is finest during spring. Comparison of modal grain sizes found in the upper (1,200 m) and lower (3,500 m) traps from stations M2 and M4 shows that the lower sediment traps have slightly coarser dust than the upper traps (Fig. 3.7).

3.4 Discussion

The grain size of dust decreases with increased distance from the source (Glaccum and Prospero, 1980; Stuut et al., 2005; Goudie and Middleton, 2006; Mahowald et al., 2014): 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. 3.3). However, we observed giant particles ($\geq 100 \mu\text{m}$) as far west as station M5 (57°W ; approximately 4,400 km from the African coast; Fig. 3.5), 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, 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 $\text{PM}_{2.5}$, 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 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 $5.5 \mu\text{m}$. 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 19th century due to commercial agriculture (Mulitza et al., 2010). Increased human activity in the source region not only increases dust emissions but 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. 3.8). These aerosols can include sea salts, organic and black carbon, sulphates and mineral dust. However, the aerosols over our study area are mostly mineral dust originating from the African continent (Yu et al., 2015a), also since Adams et al. (2012) also 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. 3.8D). Aerosol concentrations are lowest during fall (Fig. 3.8A and E), and during winter the cloud is located in its southernmost position (Fig. 3.8B). 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.

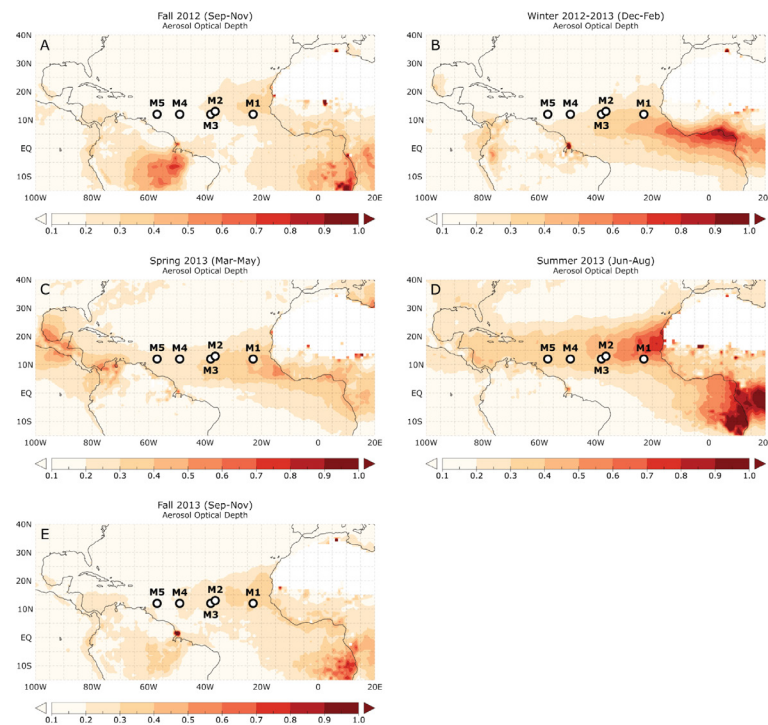


Figure 3.8. Three-month average aerosol optical depth (AOD) for the sampled seasons, from MODIS Terra. A: Fall (SON) 2012, B: Winter 2012–2013 (DJF), C: Spring 2013 (MAM), D: Summer 2013 (JJA) and E: Fall 2013 (SON). Stations M1–M5 are marked with black/white circles.

This seasonal, latitudinal shift of the dust cloud is reflected in the samples from stations M2 and M3, which are positioned at 1° 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 they are similar at both stations during summer (Fig. 3.7B). 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, determines the particle-size distributions of the entrained dust (Tsoar and Pye, 1987; Marticorena, 2014) and is further influenced during transport and deposition, creating different grain-size signatures for summer and winter dust.

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 Cabo Verde islands, which show the high mountain tops (highest point is Fogo at 2,829 m) piercing through the dust cloud, deflecting it around the islands (Fig. 3.9A). 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 Cabo Verde islands in a thick blanket of dust (Fig. 3.9B), meaning that the top of the cloud is at an elevation of at least 2,829 m. During summer, dust is transported in the high-altitude SAL (Carlson and Prospero, 1972; Prospero and Carlson, 1972; Tsamalis et al., 2013; Kanitz et al., 2014). 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) enable 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. 3.9C 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 3.9A and B, with the lowest (500 m) elevation representing winter dust transport and the highest (3,500 m) elevation representing summer dust. In winter (Fig. 3.9C), the higher trajectory is not

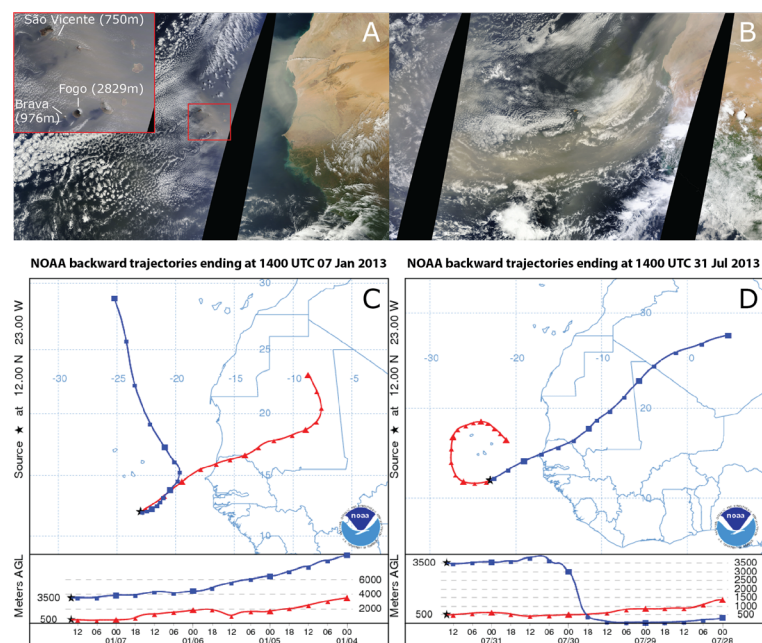


Figure 3.9. A: Satellite images of typical winter dust transport, with close-up of the Cape Verde islands (7 January 2013) and B: typical summer dust transport (31 July 2013) over the Cape Verde islands, at relatively low and high altitudes, respectively. Images from NASA Worldview, MODIS Terra satellite. 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 3,500 m (blue) AGL, showing trajectory maps (top) and elevation profiles (bottom). C: Ending at 7 January 2013, D: ending at 31 July 2013.

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. 3.9A), which is picked up from the surface and brought to higher altitudes. By contrast, in summer (Fig. 3.9D), 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 3,500 m AGL (Fig. 3.9D, bottom panel). This is in accordance with how the SAL is described: 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).

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 5 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. 3.7A). 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 a small amount of precipitation is already sufficient to wash out the suspended dust from the atmosphere by wet deposition.

At M1, the percentage of sand-sized particles ($>63 \mu\text{m}$) increases sharply in spring, while modal grain sizes increase in summer (Fig. 3.7A). This increase in coarse particles is related to coarse shoulders in the grain-size distributions of the spring samples (Fig. 3.6B) that are absent or less prominent in fall, winter, and summer (Fig. 3.6A and B). These coarse particles mostly include micas: due to their platy shape, these particles have a different aerodynamical behaviour 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, large ($\geq 100 \mu\text{m}$), more spherical particles were also observed in the samples, at very large distances from the source (Fig. 3.5). 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 (3,500 m) traps show less seasonality and are generally slightly coarser than the upper (1,200 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 3,500 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.

3.5 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 (3,500 m) show less seasonality than the upper sediment traps (1,200 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; and (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 kilometres 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, for example, 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 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 the three anonymous reviewers for their helpful comments.

Supplement

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.

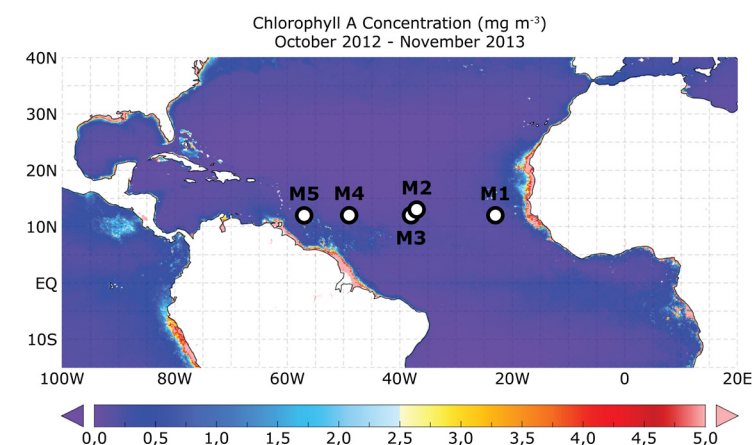


Figure S3.1. Average chlorophyll A concentrations over the Atlantic Ocean, from October 2012 to November 2013. Stations M1–M5 are marked with black/white circles. Source: MODIS Aqua, from Giovanni online data system (NASA GES DISC).

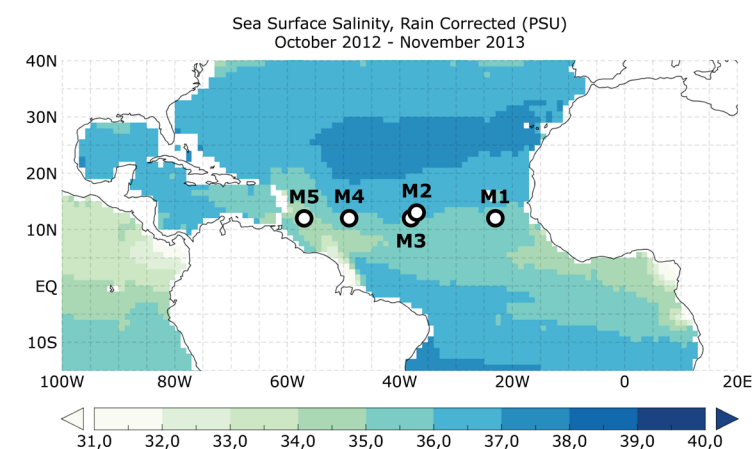


Figure S3.2. Average sea surface salinity over the Atlantic Ocean, from October 2012 to November 2013. Stations M1–M5 are marked with black/white circles. Source: Aquarius, from Giovanni online data system (NASA GES DISC).

Chapter 4:

Tropical Rains Control Saharan Dust Deposition and Bio-Availability of Nutrients

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Mineral dust plays an important role in the ocean's carbon cycle through the input of nutrients and metals which potentially fertilise phytoplankton, and by ballasting organic matter from the surface ocean to the deep sea floor (Pabortsava et al., 2017). Because open-ocean dust-flux measurements are either based on shipboard (Baker et al., 2010)- or sediment-trap data (Pabortsava et al., 2017), they are biased by interpolation and extrapolation of point observations in space and time. Alternatively, dust-flux estimations can be made using satellite observations (Yu et al., 2015a), but these are often hampered by the presence of clouds. Here, we present a two-year time series of sediment-trap dust-deposition fluxes directly below the core of the Saharan dust plume along a transatlantic array at 12°N, which shows the spatial and temporal evolution of Saharan-dust deposition across the Atlantic Ocean. We demonstrate that most of the dust deposited in the Atlantic Ocean is washed out of the atmosphere by summer rains. These field data are corroborated by comprehensive earth system model (CESM) dust-deposition data across the same transatlantic transect. In addition, nutrient-release bottle experiments in ambient sea water carried out along the same transect demonstrate how wet deposition of Saharan dust increases the release of both macro- (P, Si) and micronutrients (Fe) up to an order-of-magnitude as opposed to dry deposition. Rain-amplified bioavailability of these nutrients may well be the key to increased surface-ocean productivity in the remote and oligotrophic parts of the oceans and, potentially, also continental ecosystems (Yu et al., 2015b).

Large quantities of mineral dust are blown from the Sahara Desert across the Atlantic, impacting both the atmosphere and the ocean by altering the atmospheric radiation budget (Ryder et al., 2013b), serving as cloud condensation nuclei (Twohy et al., 2009) and ice nuclei (Atkinson et al., 2013), as well as influencing the ocean's carbon cycle through the delivery of nutrients which stimulate phytoplankton growth (Martin and Fitzwater, 1988; Pabortsava et al., 2017) and by ballasting organic particles to the ocean floor (Pabortsava et al., 2017; Van der Jagt et al., 2018). These effects depend on the size, shape, and mineralogy of the dust particles (Shao et al., 2011b), which in turn vary seasonally and along the downwind trajectory as a result of preferential settling of coarse particles (Van der Does et al., 2016; Korte et al., 2017) and atmospheric processing by cloud systems and chemical reactions (Shi et al., 2011). Transport of Saharan dust across the Atlantic Ocean was studied at sea during research expeditions (Stuut et al., 2005), estimated by remote sensing (Yu et al., 2015a), and monitored at stations on the eastern (Bory and Newton, 2000)- and western sides of the Atlantic (Carlson and Prospero, 1972). However, the general dust paradox “how are dust-derived nutrients available for phytoplankton?” remains unanswered (Blain et al., 2004).

Quantifications of actual nutrient inputs into the ocean are biased by estimates that rely on extrapolations of single depositional events encountered at sea during research expeditions. While deposition of dust into the ocean was recognised to be potentially affected by interaction with storm systems or wet deposition

(Duce et al., 1991), it remains elusive how the processing by storm systems would influence the bio-availability of nutrients carried by dust particles.

We monitored two years of Saharan dust deposition fluxes in-situ at four sites across the Atlantic Ocean using time-series of submarine sediment traps (M1–M4). The traps were moored at 1200 meters water depth, three positioned along a transect at 12°N and a fourth at 13°N (Fig. 4.1A), sampling from October 2012 to October 2014 (Supplement Table S4.1). This transect is complemented by two years (2013–2014) of Saharan dust concentrations measured on Barbados (Fig. 4.1B). We combine these observational data (Fig. 4.1C) with dry- and wet-dust deposition fluxes simulated with a coupled earth system model (Fig. 4.1D), and dust optical depth from the Moderate Resolution Imaging Spectroradiometer (MODIS, Fig. S4.1) as well as precipitation from the Tropical Rainfall Measuring Mission (TRMM, Fig. S4.1) along the transect (see Methods). In addition, we experimentally determined the release of nutrients from a Saharan dust source into Atlantic surface waters, contrasting dry- and wet deposition (Fig. 4.2). The wet-deposited dust was pre-treated with sulfuric acid of pH=2, mimicking chemical conditions that have been observed to occur in clouds (Zhu et al., 1992; Shi et al., 2011; see Methods), although in actual rainwater, the pH is obviously higher (Spokes and Jickells, 1996).

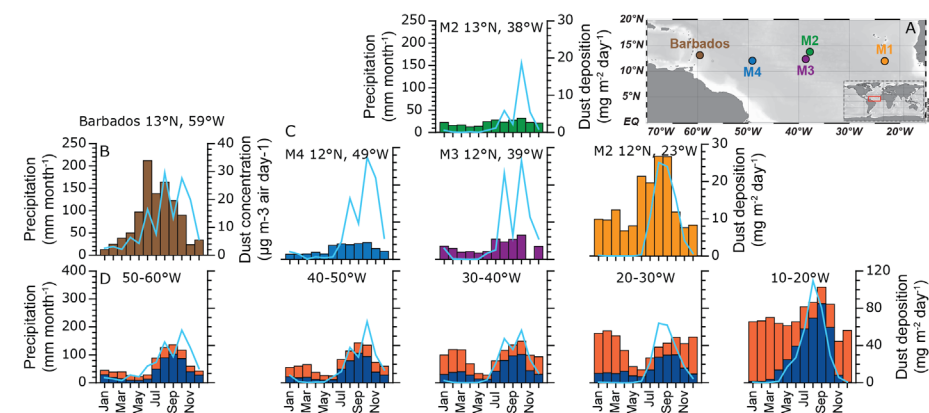


Figure 4.1. Setup of transatlantic dust-monitoring experiment with A: map projection of sediment-trap sampling stations M1–M4, and Barbados (Prospero et al., 1970). B: Average daily dust concentrations at Barbados (bars; right Y-axis) and averaged monthly precipitation (1°×1°, blue lines; left Y-axis), both averaged for 2013–2014. C: Daily dust deposition in the sediment traps M1–M4 (various longitudes across Atlantic, bars; right Y-axes), with monthly precipitation (1°×1°) both averaged per month for 2013–2014 (blue lines; left Y-axes). D: Stacked diagram of simulated transatlantic dust deposition fluxes averaged over 5°×10° latitude-longitude areas between 10°–15°N and 10°–60°W (dry deposition [red bars], stacked upon wet deposition [blue bars]); Y-axes on the right) and over a period of 15 model years. Monthly precipitation (5°×10° lat-lon) averaged per month for 2013–2014 (blue lines; left Y-axis).

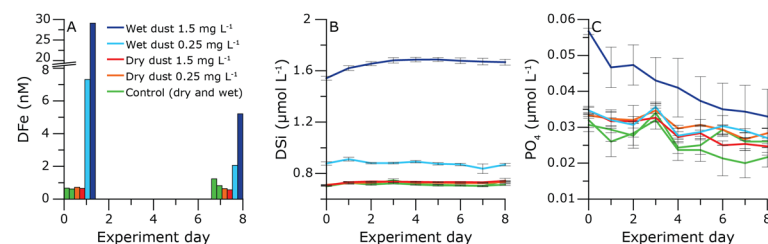


Figure 4.2. Development of A: total dissolved iron (DFe), B: silicate (DSi), and C: phosphate (PO_4) concentrations on each experiment day (DFe only measured on days 1 and 8) as a result of dry- and wet-dust additions of 0.25 and 1.5 mg L^{-1} Saharan dust to seawater sampled at station M1 ($12^\circ\text{N}, 23^\circ\text{W}$). Nutrient concentrations of dry-dust additions hardly differ from control runs, whereas wet-dust additions increase the amount of released nutrients significantly.

Observed dust mass fluxes were highest at station M1 and decreased sharply downwind to M4 (Fig. 4.1C. and S4.1A). Dust deposition showed distinct seasonality across the Atlantic, peaking in summer and autumn, and with lowest fluxes in winter and spring (Fig. 4.1C and S4.2B–E). Although station M2 is positioned only about 160 km north of station M3, it appears to have registered lower fluxes and less seasonality, which we hypothesize to be caused by its position away from the centre of the Saharan dust cloud. Overall, the fluxes' seasonal amplitude decreased from east to west, coinciding with seasonal particle-size trends along the transect (Van der Does et al., 2016), and with previously reported Saharan dust fluxes in the eastern Atlantic (Bory et al., 2002; Fischer et al., 2016).

The modelled dust deposition fluxes (Fig. 4.1D) should be interpreted in a general sense as they are the average across 15 years. As a result, they cannot be compared one-to-one to the observed fluxes, which may be subject to interannual variability. Yet, generally the modelled fluxes also generally reveal a strong decrease from east to west, albeit more gradual than the downwind decrease in fluxes observed in the sediment traps (Fig. 4.1C). More importantly, the modelled data show a sharp seasonal contrast between dry- and wet deposition, which is more pronounced than observed in the sediment traps. Modelled wet deposition is highest in summer and fall, following the meridional movement of precipitation along with the Intertropical Convergence Zone (ITCZ; Nicholson, 2000). From east to west, the modelled dominant deposition mode changes from winter/dry- to summer/wet deposition, which also match observations in Florida at the western side of the Atlantic, where wet dust deposition was found to dominate (Prospero et al., 2010). Dry deposition is highest in winter and spring in the east, and decreases westward in both magnitude and seasonal amplitude. These modelled deposition maxima in winter and spring (dry deposition) and late summer and fall (wet deposition) in the Atlantic Ocean are corroborated by the sediment-trap observations and the incidence of precipitation.

In concert with increased deposition fluxes observed in the traps, atmospheric dust loads as measured by DOD, which can only be quantified in clear-sky conditions,

were also highest in early summer, and decrease in mid-summer when precipitation washes the dust out of the atmosphere (Fig. S4.1). In late summer, increased dust deposition fluxes coincided further with intensified precipitation along the entire transect, although less clear at M2 (13°N) where precipitation is lower.

On Barbados, atmospheric dust loads increased in summer and autumn (Fig. 4.1B). Saharan dust arriving at Barbados is the residual of dust not being washed out by rains during its transatlantic crossing, and dust concentrations peak before precipitation peaks over the Atlantic. Reported Saharan dust concentrations in the western Atlantic peaked in spring (French Guyana) and summer (Barbados; Prospero et al., 2014), and previous studies have hypothesised that previous-year Sahel precipitation controls dust generation and emission (Prospero and Nees, 1986). In light of our current findings, we argue that North Atlantic precipitation in turn strongly influences the amounts of Saharan dust reaching the western Atlantic Ocean.

Sulfuric- and nitric acids are thought to be the most important acids for chemical processing of dust particles in the atmosphere that increase the solubility of, foremost, iron (Baker and Croot, 2010). Acid-processing of dust particles was shown to occur in clouds, where acidity may be as low as $\text{pH}=1$ and the solubility of ferric iron increases by three orders-of-magnitude for each unit decrease in pH (Zhu et al., 1992). Mesocosm experiments in the Mediterranean suggested that wet-deposition leads to increased concentrations of bio-available nutrients, particularly iron (Pulido-Villena et al., 2014), as opposed to dry-deposition (Ridame et al., 2014). Our nutrient-release experiments showed that deposition of pre-treated (wet) dust into sea water leads to an initial doubling of the release of phosphate and silicate and to an order-of-magnitude increase in dissolved iron, compared to dry deposition, which show no significant difference from the control runs (Fig. 4.2). In addition, concentrations of the leached nutrients remained significantly higher during the course of the nutrient-release bottle experiments.

In summary, our combined results of in-situ dust deposition fluxes, model simulations, remote-sensing analyses, and shipboard observations show that wet deposition is the main mechanism that deposits Saharan dust into the tropical Atlantic Ocean. Moreover, our shipboard experiments clearly showed wet-deposition events leading to a doubling in the release of macronutrients (P, Si) and up to an order-of-magnitude release of micro-nutrients (Fe). Consequently, and as was suggested from surface-ocean measurement (Schlosser et al., 2014), wet deposition of bio-available nutrients from mineral dust may well be key to increased surface-ocean productivity, particularly in oligotrophic parts of the world's oceans. Considering future changes in global and regional precipitation patterns and the many uncertainties regarding the dust cycle (IPCC, 2013), an improved understanding of these mechanisms is key to future climate predictions and the impact of mineral dust on global carbon budgets through ocean fertilisation.

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Supplement

Methods

This paper presents the particle fluxes from sediment traps moored along a transatlantic transect positioned directly below the main path of the Saharan dust cloud off northwest Africa. Deployment and recovery of the moorings and sediment traps, as well as specific information on the mooring equipment and sampling intervals, is described in the respective cruise reports (Stuut et al., 2012; 2013; 2015; 2016). In addition, the paper describes nutrient-release experiments carried out during an additional research cruise in the Atlantic Ocean (Stuut et al., 2016) in which we incubated ambient sea water with different kinds of dust to mimic cloud processing of dust resulting in wet deposition, contrasted with dry deposition. In this supplement, more details are provided about the applied methods.

Sediment Traps

The transect consists of four mooring stations (M1–M4), which sampled for two consecutive years (Table S4.1; for more details, see Van der Does et al. (2016). Three moorings were positioned at 12°N, and a fourth at 13°N. Here we present data from the upper sediment trap of each mooring at approximately 1,200 meters water depth, sampling continuously at 8 or 16-days intervals (Table S4.1). Sediment trap 2014-M1 only sampled the first nine intervals due to a technical failure.

Table S4.1. Details of the sediment traps at station M1–M4, for the two sampling years 2013 (2013Mx) and 2014 (2014Mx).

Sediment trap	Latitud (°N)	Longitude (°W)	Trap depth (m.b.s.l.)	Distance from African coast (km)	Sampling intervals (days)	Sampling period
2013-M1	12.00	23.00	1,150	700	16	19 Oct 2012 – 7 Nov 2013
2013-M2	13.81	37.82	1,235	2,300	16	19 Oct 2012 – 7 Nov 2013
2013-M4	12.06	49.19	1,130	3,500	16	19 Oct 2012 – 7 Nov 2013
2014-M1	12.00	23.01	1,200	700	8 & 16	23 Nov 2013 – 31 Mar 2014
2014-M2	13.81	37.82	1,190	2,300	8 & 16	1 Dec 2013 – 25 Oct 2014
2014-M3	12.40	38.63	1,362	2,400	8 & 16	1 Dec 2013 – 17 Oct 2014
2014-M4	12.04	49.22	1,100	3,500	8 & 16	9 Dec 2013 – 25 Oct 2014

Prior to deployment of the sediment traps, a biocide was added to the sampling cups (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} , $\text{pH} \approx 8.5$). As a result, the density inside the sampling cups was higher than the ambient seawater, to avoid water from leaking out. After recovery, the samples were sieved through a 1 mm mesh to remove zooplankton, and subsequently wet-split in five equal splits using a WSD10 rotor splitter (McLane Laboratories, USA). Samples were then washed with Milli-Q water and centrifuged (5 minutes at approximately $1800 \times g$) three times. Bulk-sample mass was determined by weighing two freeze-dried 1/5 aliquots of each sample, resulting in an average deviation of 2.67% ($\sigma = 2.38$) by mass between paired splits. For determining the dust content of the samples, 1/25 of the original sample was used.

Mineral Dust Fluxes

Next to Saharan dust, the sediment-trap samples contained numerous particles derived from the marine environment, including biomineral particles (e.g. foraminifera, radiolarians, diatoms, coccolithophores) and organic matter (marine and atmospheric from e.g. biomass burning). To isolate the mineral dust fraction, all biogenic constituents were removed chemically, following the procedure described by Filipsson et al. (2011). The samples were then filtered over a pre-weighed 25 mm, $0.4 \mu\text{m}$ polycarbonate filter and rinsed with Milli-Q water. By weighing the filter, the Saharan dust mass flux $>0.4 \mu\text{m}$ could be determined. The weight of the sediment fraction smaller than $0.4 \mu\text{m}$ is considered negligible (Rosenberg et al., 2014). The filtration over $0.4 \mu\text{m}$ filters also allows for a direct comparison with particle-size distributions of the same samples as presented by Van der Does et al. (2016), who describe particle sizes from 0.4 to $2,000 \mu\text{m}$. Samples were assigned a mid-date, which is the middle of their 8 or 16-day sampling period. A short delay between ocean surface and sediment traps must be considered as the organic aggregates that incorporate the dust particles and carry them downward, have settling velocities of about 200 m day^{-1} (Nowald et al., 2015; Van der Jagt et al., 2018), which is in line with previously determined settling velocities for our sediment-trap samples of at least 140 m day^{-1} (Van der Does et al., 2016; Korte et al., 2017).

Dust mass fluxes are highest at M1, with average values of $14.3 \text{ mg m}^{-2} \text{ day}^{-1}$ in 2013, and decrease sharply downwind to M4 with 3.3 and $2.3 \text{ mg m}^{-2} \text{ day}^{-1}$ in 2013 and 2014, respectively (Fig. S4.1A).

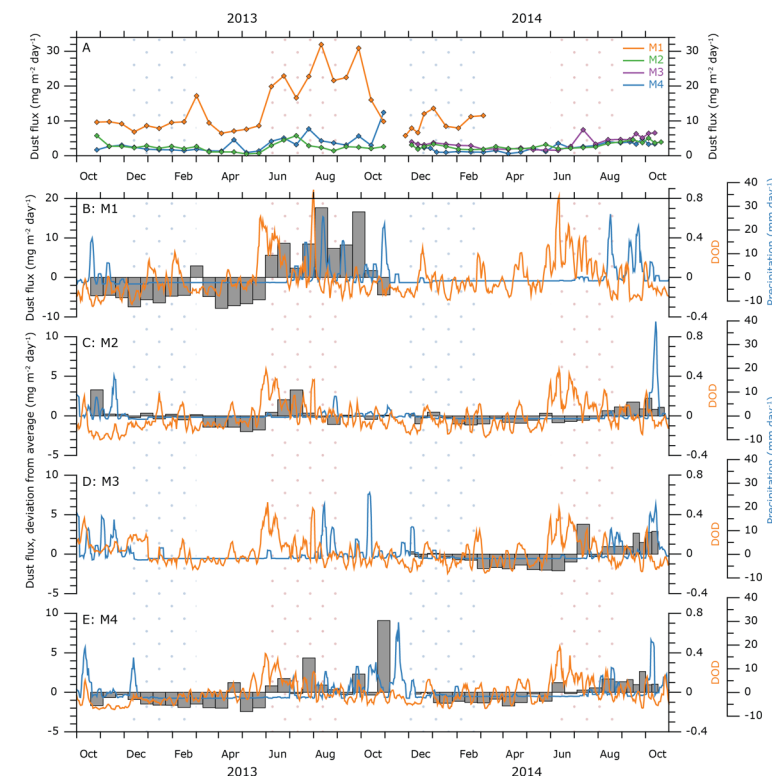


Figure S4.1. Time series of observed Saharan dust and precipitation across the Atlantic, A: Total mass fluxes of Saharan mineral dust (particles $>0.4 \mu\text{m}$) collected by sediment traps M1-M4 from autumn 2012 to autumn 2014. B-E: Dust mass fluxes from the sediment traps as deviation from the annual 24-sample average (bars), Dust Optical Depth (DOD, MODIS Terra; orange lines) and precipitation (TRMM; blue lines), as deviation from the yearly averages at station M1-M4. Note the different scale for dust fluxes at station M1 (B). Due to instrument failure after the ninth sample at 2014-M1, the average was calculated for those samples only ($N=9$). Stippled areas represent winter (DJF) and summer (JJA) seasons.

Dust fluxes derived from the total mass of the terrigenous fraction with the method described above appear smaller than so-called residual-mass fluxes, which are calculated by subtracting the weights of all biogenic constituents from the bulk-sample mass, and which are often assumed to represent the lithogenic or mineral dust fraction (Fischer et al., 2016). Residual mass fluxes were determined for the samples from October 2012 to November 2013 by Korte et al. (2017). Although the residual flux is highly correlated with the dust flux presented here ($R^2 = 0.69-0.80$), absolute values are 2 to 18 times higher (average 5.8 , $\sigma = 3.3$; Fig. S4.2). Korte et al. (2017) concluded that the residual fraction is most likely an overestimation, due to various uncertainties and assumptions in the mass of organic matter and biogenic opal, which is demonstrated by comparing these two representations of the dust fraction.

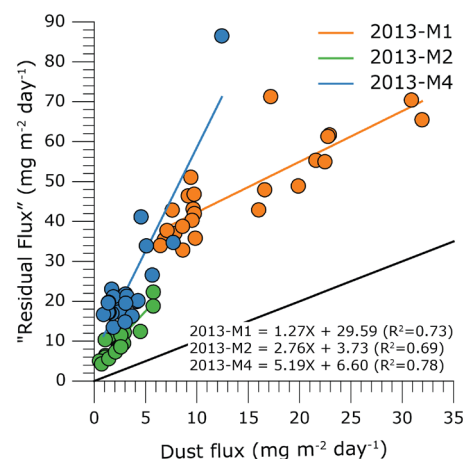


Figure S4.2. “Residual” flux (calculated after sequential leaching; Korte et al., 2017) versus dust flux (presented here) for 2013-M1–M4. Black line represents 1:1 relation.

Remote Sensing

Dust Optical Depth (DOD) was derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) Aerosol Optical Depth (AOD) and fine-mode fraction (FMF) Collection 6 data, from both Terra and Aqua satellites, following the method developed by Yu et al. (2009). This is based on the distinct particle sizes of mineral dust, marine aerosols and combustion aerosols. The characteristic FMF for individual aerosol types based on MODIS measurements (over ocean only) were determined, and AOD and FMF were used to derive the DOD (Yu et al., 2009). Daily DOD data were obtained for 2012–2014 (Fig. S4.1) and averaged for both Terra and Aqua, only when both AOD and FMF were retrieved.

Precipitation data from TRMM (Tropical Rainfall Measuring Mission) was obtained at daily resolution for $1^\circ \times 1^\circ$ boxes around the sampling sites from the Giovanni online data system, developed and maintained by the NASA Goddard Earth Sciences Data and Information Services Center (GES DISC). For smoothing purposes, 5-day running averages were computed for both DOD and precipitation data (Fig. S4.1).

Dust Modelling

The model results presented here (Fig. 4.1 in $5^\circ \times 10^\circ$ lat-lon resolution and Fig. S4.3 in $5^\circ \times 5^\circ$ lat-lon resolution) were obtained with the Community Earth System Model (CESM; Hurrell et al., 2013). Present-day boundary conditions were prescribed in a fully coupled control simulation including atmosphere, land surface, ocean, and sea ice components. The simulation uses a resolution of $0.9^\circ \times 1.25^\circ$ (lat×lon) for the atmosphere and a nominal resolution of 1° for the ocean and was conducted

for 20 model years. Results are presented as multi-year averages based on the last 15 years of the simulation, to obtain a robust climatological average. In our setup, the Community Atmosphere Model (CAM4; Neale et al., 2013) has been chosen as the atmospheric component including the bulk aerosol module (BAM) to simulate mineral dust prognostically in four size bins (particle diameters of 0.1 to 1.0 μm , 1.0 to 2.5, 2.5 to 5.0, and 5.0 to 10.0 μm). The model includes representations of dust emission, transport and deposition which comprises both dry deposition (including both turbulent and gravitational settling) and wet deposition (through in-cloud and below-cloud scavenging). More details on the representation of the dust cycle in the model can be found in Albani et al. (2014) and references therein. The main message that we take from the modelling data is the significant difference between summer (wet) and winter (dry) deposition. The aim of our manuscript is pertinently not to calibrate a model with robust *in-situ* measured time-series data, but rather the other way around: using the model's general outcome to provide a complementary framework to understand the processes that we infer from the *in-situ* observations.

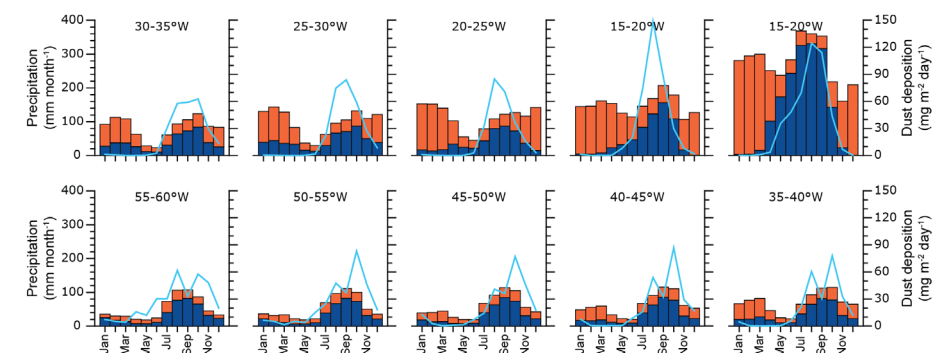


Figure S4.3. Dust-deposition modelling results presented in $5^\circ \times 5^\circ$ grid cells. Stacked diagram of simulated transatlantic dust deposition fluxes averaged over $5^\circ \times 5^\circ$ latitude-longitude areas between 10° – 15° N and 10° – 60° W (dry deposition [red bars], stacked upon wet deposition [blue bars]); Y-axes on the right). Monthly precipitation ($5^\circ \times 5^\circ$) averaged per month for 2013–2014 (blue lines; left Y-axis).

Nutrient-Release Experiments

Saharan dust nutrient-release experiments were conducted at station M1 (12° N, 23° W) in spring 2016 during research cruise JC134 (Stuut et al., 2016). Seawater was sampled from the mixed layer at 20 m with a Seabird CTD equipped with a rosette holding 24×10 L Niskin bottles. From the rosette, the seawater was tapped gently into the acid-cleaned (10% HCl) incubation bottles. Wet- and dry-deposition from a paleo lake Saharan dust source (21.08° N, 12.19° W) was mimicked in varying amounts (0.25 and 1.5 mg L^{-1}) by adding pre-leached and untreated dust to the incubation bottles, respectively. The paleo-lake is located in the foothills of the Atlas Mountains, in one potential source and transit area, from where Saharan dust is transported across the Atlantic Ocean (Scheuven et al., 2013). Before addition, the particle size of the dust was adjusted to be comparable to the dust that was

observed in the traps at site M1 (Van der Does et al., 2016) by sieving over a 32 μm sieve. For wet deposition, the dust was leached in 40 mL of artificial rain (consisting of Milli-Q + H_2SO_4) at $\text{pH}=2$, at an end concentration of 0.0051 M H_2SO_4 . It was kept in suspension for 24 hours, resembling the timespan used for cloud processing experiments (Spokes et al., 1994; Shi et al., 2009). Untreated dust was added in the same amounts directly to the seawater in the incubation bottles as a representative for dry-dust deposition. The experiments were run in triplicates for a total of eight days. After each day of the experiment, subsamples of the water from the incubation bottles were filtered and analysed for the macro-nutrients phosphorous (PO_4), silicate ($\text{Si}(\text{OH})_4$) and nitrate (NO_3) using colorimetric methods (Murphy and Riley, 1962; Strickland and Parsons, 1972; Hansen and Koroleff, 1983). Nitrate measurements did not show a different behaviour between dry- and wet-dust additions (Fig. S4.4), showing that nitrogen compounds leached from Saharan sources in sulfuric acid are negligible. The micro-nutrient iron (DFe) was analysed on the first and last day of the experiment using automated Flow Injection Analysis (Klunder et al., 2011).

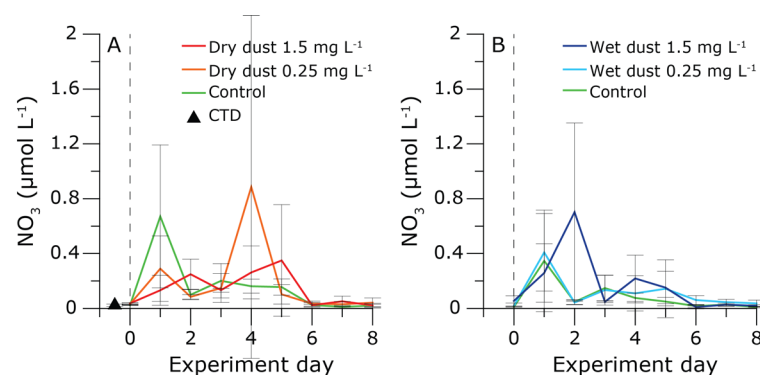


Figure S4.4. Development of nitrate (NO_3) concentrations of each experiment day as a result of dry- (A) and wet-dust (B) additions of 0.25 and 1.5 mg L^{-1} Saharan dust to seawater sampled at station M1 (12°N, 23°W). Results show no differences between controls and dry- and wet-dust additions. Dashed line represents start of the experiment.

Chapter 5:

The Mysterious Long-Range Transport of Giant Mineral Dust Particles

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Abstract

Giant mineral dust particles ($>75\ \mu\text{m}$ diameter) found far from their source have puzzled scientists for a long time. These wind-blown particles impact the atmosphere's radiation balance, clouds and the ocean carbon cycle but are generally ignored in models. Here we report new observations of individual giant Saharan dust particles of up to $450\ \mu\text{m}$ in diameter sampled in air over the Atlantic Ocean at 2,400 and 3,500 km from the west African coast. Past research points to fast horizontal transport, turbulence, uplift in convective systems and electrical levitation of particles as possible explanations for this fascinating phenomenon. We present a critical assessment of these mechanisms using order-of-magnitude estimates and backward trajectories. Results show that established concepts are merely able to explain our new observations. Therefore we propose several lines of research we deem promising to further advance our understanding and modelling.

5.1 Introduction

About 30 years ago, scientists first observed so-called giant ($>75\ \mu\text{m}$ diameter) wind-blown mineral dust particles at large ($>10,000\ \text{km}$) distances from their source (Betzer et al., 1988). These sand-sized mineral aerosols or dust particles, the largest of which ($>200\ \mu\text{m}$) were all individual quartz grains, were transported from Asia to the remote Pacific Ocean. In Europe, giant dust particles were found $>4,000\ \text{km}$ from their Saharan source (Middleton et al., 2001), and dust particles up to $300\ \mu\text{m}$ in diameter were sampled during aircraft campaigns over northwestern Africa (Ryder et al., 2013b). In marine sediment traps, positioned underneath the main Saharan dust plume in the Atlantic Ocean, giant particles are dominated by large quartz particles $>100\ \mu\text{m}$, found at distances up to 4,400 km from the west African coast (Van der Does et al., 2016).

The Sahara is currently the largest single source of wind-blown sediments. Transport of Saharan dust across the Atlantic Ocean is subject to seasonal atmospheric changes in wind systems, blowing at different altitudes. In winter, low-level dust is carried towards the Atlantic with the northeasterly trade winds, or Harmattan, at altitudes between 0 and 3 km (Tsamalis et al., 2013). In summer, the Saharan Air Layer (SAL) dominates dust transport. Upon reaching the west African coast, the SAL encounters a cool marine air mass that lifts the warm, dusty air to altitudes up to a maximum of 5–7 km (Adams et al., 2012; Tsamalis et al., 2013; Chouza et al., 2016). Wind velocities of up to $25\ \text{m s}^{-1}$ associated with the Atlantic extension of the African easterly jet can lead to fast westward transport, particularly around 4 km (Sarnthein et al., 1981; Chouza et al., 2016).

It is often assumed that the particle size of long-range transported mineral dust does not exceed 20–30 μm (Tsoar and Pye, 1987; Zender et al., 2003; Kok et al., 2012; Ryder et al., 2013a), and climate model simulations often limit particle diameters to only $<10\ \mu\text{m}$ (Huneus et al., 2011). However, the incorporation of coarse particles is important as the radiative effect of dust is especially sensitive

to the coarse dust mode. Coarse particles reduce the single scattering albedo of shortwave radiation, increasing radiative absorption (Ryder et al., 2013b), and enhance the absorption of longwave radiation (Otto et al., 2007), possibly causing a net atmospheric warming as shown by Kok et al. (2017). This latter study (Kok et al., 2017) also demonstrated a significant effect on the atmospheric radiative balance when larger particles up to $20\ \mu\text{m}$ are incorporated into climate models. If giant dust particles ($>75\ \mu\text{m}$) would be considered, the effects on atmospheric radiation budgets could be tremendous. In addition, the under- (or non-) representation of particles larger than $10\ \mu\text{m}$ in climate models and the distance these particles can travel affect total deposition fluxes over land and ocean. Giant mineral dust particles also play a role in the ocean carbon cycle, as they have a large ballasting potential for marine organic aggregates, making these aggregates denser and therefore aiding the transport of organic matter to the deep ocean (Van der Jagt et al., 2018). In addition, they influence cloud microphysics by acting as giant cloud condensation nuclei, which can accelerate the hydrological cycle through increasing precipitation rates (Posselt and Lohmann, 2008). This demonstrates why a mechanism explaining the long-range transport of giant dust particles is urgently needed.

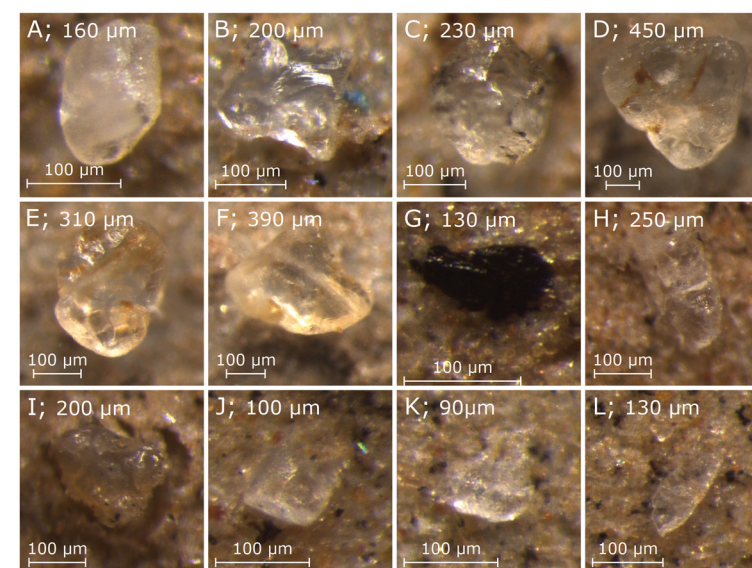


Figure 5.1. Giant mineral dust particles sampled by the MWAC samplers at M3 (12°N , 38°W) and M4 (12°N , 49°W) in 2014 and 2015, with their approximate diameters. A–C: 2014–M3; D–F: 2014–M4; G–I: 2015–M3; J–L: 2015–M4.

5.2 Results

5.2.1 New Evidence for Long-Range Transport of Giant Dust Particles

Here we present new data from the same trans-Atlantic transect as Van der Does et al. (2016), this time collected directly from the atmosphere by Modified Wilson and Cooke (MWAC) samplers (see Methods), mounted on moored dust-collecting surface buoys at two stations in the tropical North Atlantic Ocean. The passive air-samplers collected one discrete sample during periods between 2013 and 2016, comprising 281–432 days (Table 5.2) at approximately three meters above sea level. These samples show giant dust particles ($>75\text{ }\mu\text{m}$; Fig. 5.1) that were collected at 2,400 and 3,500 km, respectively, from the west African coast at sampling stations M3 and M4 (Fig. 5.2). These are mostly well-rounded quartz particles up to $450\text{ }\mu\text{m}$ in diameter, with what appears to be high aspect ratios. As atmospheric samples, these sand-sized dust particles can only have been carried there by the wind. This observation adds further evidence to the ability of the atmosphere to transport giant particles over very long distances.

The main reason why climate models do not incorporate dust particles $>10\text{ }\mu\text{m}$, despite the growing evidence for their existence far away from sources suggesting substantial residence times in the atmosphere, is related to the physical laws on which the models are based. The settling speed of small particles in air is usually calculated using equations based on Stokes’ Law (Maring et al., 2003; Zender et al., 2003). Settling velocities for particles $>20\text{ }\mu\text{m}$ are overestimated by Stokes’ Law due to turbulence created by the falling of larger particles and are therefore determined empirically (Bagnold, 1941; Zender et al., 2003). Using the formula from Bagnold (1941), we compare the settling velocities of giant mineral dust particles of 100, 200, and $300\text{ }\mu\text{m}$ (Table 5.1). These data show that with such rapid settling velocities, it is not possible for giant particles to reach the sampling sites at 2,400 and 3,500 km from the west African coast, even at high wind velocities of 25 m s^{-1} . Clearly some additional mechanisms are needed to keep these dust particles aloft.

Table 5.1. Settling velocities after Bagnold (1941), and estimates of travelled distance based on favorable summer (strong winds, elevated dust) and winter (lower wind speeds and elevation) conditions.

Particle size (μm)	Settling velocity (mm s^{-1})	Summer: Travelled distance at 25 m s^{-1} winds from 7 km altitude	Winter: Travelled distance at 10 m s^{-1} winds from 3 km altitude
100	400	438 km	75 km
200	1,000	175 km	30 km
300	1,500	117 km	20 km

5.2.2 Potential Mechanisms

Several studies suggest such mechanisms including atmospheric vertical mixing (Maring et al., 2003; Denjean et al., 2016; Gasteiger et al., 2017) or large dust storms and turbulence (Tsoar and Pye, 1987; Betzer et al., 1988; Ryder et al., 2013a), but the capacity of these mechanisms for long-range transport of giant dust particles over the ocean has not been explored. In addition, the shape of the mineral dust particles can influence their deposition, with aspherical particles having lower settling velocities than more spherical particles (Stuut et al., 2005; Yang et al., 2013). However, this does not seem to have a large effect on the giant dust particles observed at our Atlantic sampling locations, as these are almost exclusively spherical quartz minerals (Fig. 5.1).

Here we provide an integral discussion of the potential of four different mechanisms that can facilitate long-range transport of giant particles. First, strong winds causing fast horizontal transport greatly enhance the distance over which the dust travels. Second, transport of individual large dust particles is further aided by strong turbulence, keeping them in suspension for a longer time (Garcia-Carreras et al., 2015), although this could also have a negative effect on the dust particles, causing them to settle even quicker. Third, particle charge affects their dynamics, and, for negatively-charged particles, can offset a particle’s weight in a downward directed electric field, so keeping it aloft for longer (Nicoll et al., 2011; Renard et al., 2018). Finally, thunderstorms or tropical cyclones can carry dust particles to great heights, strongly increasing their horizontal travel distance if the particles can leave the storm through the anvil or upper-level outflow region without being rained out. This mechanism will be particularly effective when multiple uplift cycles are encountered along active areas such as the intertropical convergence zone (ITCZ).

In the following we provide a plausibility analysis of these four factors to test whether they can in fact explain our new observations of giant dust particles. We will discuss winter and summer situations separately.

5.2.3 Winter Scenario

In boreal winter, dust transport occurs at lower altitudes and at lower wind speeds (Table 5.1). There is insufficient convection over the sampling area to aid the long-range transport of dust particles, as the ITCZ is shifted southward (Žagar et al., 2011). Backward-trajectory calculations for February (see Methods) show that simple horizontal particle transport (no settling) within the boundary layer, where horizontal winds are fastest, would still take at least 48 hours to reach M3, and at least 72 hours to reach M4 (Fig. 5.2A). Transport at higher levels is unlikely, as winds become increasingly more westerly with height (Tomas and Webster, 1994). Assuming the sedimentation velocities for particles of $100\text{ }\mu\text{m}$ given in Table 5.1, the shortest travel time to M3 and M4 would correspond to a total vertical sedimentation of ~ 70 and 100 km , respectively, strongly suggesting that other mechanisms must be involved.

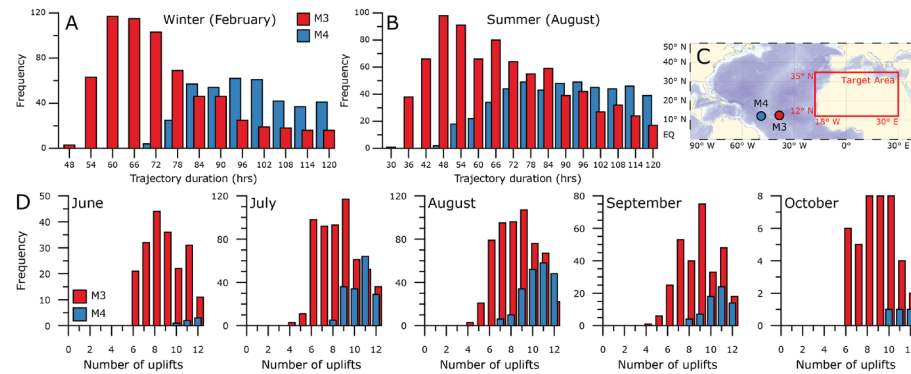


Figure 5.2. Distribution of travel times for backward trajectories from buoys M3 (red) and M4 (blue) to the target area (panel C), for February (A) and August (B). D: Frequency of the minimum number of deep convective uplift cycles needed for a 100 μm particle to travel from the target area (panel C) to the sampling buoys M3 (red) and M4 (blue), assuming a constant sedimentation velocity of 400 mm s⁻¹ (Table 5.1), for June–October. All computations are based on ERA-Interim data during the ten-year period 2006–2015.

Fast horizontal transport events will be characterized by strong winds over the ocean but often also over land, as these tend to be associated with synoptic-scale subtropical highs (Knippertz et al., 2011), leading to large dust emissions. Given that transport is usually restricted to the boundary layer, we can assume highly turbulent conditions, stirred mechanically by high wind shear, even in the absence of buoyancy generation over the ocean. However, it is difficult to quantify the effect of turbulence on the likelihood of individual giant dust particles to stay suspended without any direct observations of this process.

The third mechanism which may contribute to keeping giant particles aloft is via electrical forces. Many studies have found that atmospheric charging affects particle dynamics, with a vertical electrical force being able to potentially compensate a particle's weight (Nicoll et al., 2011; Renard et al., 2018). Renard et al. (2018) found large particles (>40 μm) persisting over long distances over the Mediterranean region, without significant downwind trends in size. They speculated this was due to particle charge, counteracting gravitational settling. Electric charge has also been shown to increase dust emission from source areas up to tenfold (Kok and Renno, 2006; Esposito et al., 2016). Whether or not the electric field hinders or assists a particle in staying aloft depends on the relative polarity of the particle and the atmospheric electric field encountered. The downward-directed electric field always present in fair weather will drive positively-charged particles downwards, but the field direction can reverse in disturbed weather or during saltation events (Harrison et al., 2016) and particles can readily carry both polarities of charge, so an upwards electrical force is possible. The initial charge generated at dust emission is lost within hours (Nicoll et al., 2011) but charged particles have still been detected far away from sources. For instance, during Hurricane Ophelia of October 2017, which brought large amounts of dust and soot particles to the

southern UK, appreciable negative charge was detected in the dust plume after a transport time of tens of hours (Harrison et al., 2018), showing that charge is also generated during transport. The most likely reason is that particles are more or less continuously charging through collisions, a process called tribo-electrification (Farrell et al., 2004; Harrison et al., 2016). This effect is facilitated in an atmospheric layer that is characterized both by a high dust concentration and strong turbulence, indeed exactly the kind of strong-wind situation described above, which would sustain electric fields sufficiently to reduce the fall speeds of highly charged particles. A further consequence of a system of particles is to reduce the air's electrical conductivity through removal of cluster ions, allowing the charge on the particle assembly to be sustained for longer than for an isolated charged particle.

Two factors together determine the electrical effect on a particle of a given size: the local atmospheric electric field and the particle charge. The atmospheric electric field varies appreciably between fair weather conditions ($E = -10^2 \text{ V m}^{-1}$), and disturbed weather conditions such as convective clouds and thunderstorms in which substantial charge separation occurs meteorologically to generate strong electric fields of different polarities ($E = \pm \sim 10^4 \text{ V m}^{-1}$). An individual particle's charge aloft can also cover a wide range, resulting from its interactions with other particles and cosmic ray-generated ions, or, exceptionally, its internal radioactivity. Figure 5.3A shows the number of particle charges necessary for the electric force on the particle to have the same magnitude as a particle's weight in a range of typical atmospheric electric fields, i.e. under which conditions it could become levitated. In the weak fair weather field (10^2 V m^{-1}), only the smallest particles are affected, with ~ 2 elementary charges (e) required. For larger particles, the number of charges and field required increases. In electric fields characteristic of disturbed weather ($\sim 10^4 \text{ V m}^{-1}$), particles of 100 μm typically require 10^7 or $10^8 e$ to offset their weight and reduce the particle's fall speed (Fig. 5.3B). In-situ measurements of individual particle charges aloft are not available for comparison and related quantities near the surface are only poorly-known, but charges found in dust devils are of $\sim 10^6 e \text{ cm}^{-3}$ (Farrell et al., 2004) and re-suspended dust on the order of 10^3 to $10^4 e / \text{particle}$ (or $10^{12} e \text{ cm}^{-3}$) (Harrison et al., 2016). Larger charges are likely for giant particles, scaling with particle area, but at very large charges and fields, charge emission would prevent further electrification (Whitby and Liu, 1966). One highly relevant factor is particle composition (Esposito et al., 2016; Harrison et al., 2016), as it is known that mineralogy affects the charging of particles. The giant dust particles found at the buoys are mostly quartz (Fig. 5.1) and it is found experimentally that quartz particles may charge more easily than clay minerals (Harrison et al., 2016, and references therein). Any electrically-assisted transport would require the sustained or fortuitous presence of strongly electrified clouds and particle charges of the appropriate relative polarities. Clearly, more lab, field and numerical studies are needed to quantify this effect in a fully turbulent dust layer with interacting particles of different sizes.

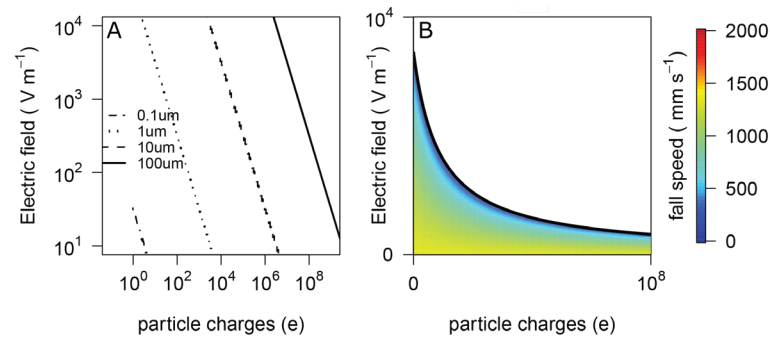


Figure 5.3. Influence of charge and electric field on the net force on a particle. A: Combination of particle charge and local electric field required for the magnitude of the electric force experienced by a particle to equal the particle's weight, for particles having diameters of 0.1, 1, 10 and 100 μm . B: Fall speed for a 100 μm quartz particle for increasing electric field and particle charge (density of quartz = 2648 kg m^{-3} , drag coefficient $C_D = 1.5$).

5.2.4 Summer Scenario

A fourth potential mechanism occurs during summer in the presence of ITCZ convection. Fastest horizontal transport (based on 6-hourly ERA-Interim 3-dimensional wind fields (Dee et al., 2011); see Methods) at the latitude of the buoys (i.e., $\sim 12^\circ\text{N}$) occurs within the African easterly jet (Fink et al., 2004), reducing the minimum transport time in August to M3 at 600 hPa to 30 hours, and to M4 to 48 hours (Fig. 5.2B). This is significantly less than in winter, generally increasing the probability of giant dust particles reaching the sampling sites, consistent with the higher number of larger dust particles found in submarine sediment traps at our sampling sites in summer (Van der Does et al., 2016). As in winter, we would generally assume giant particles to only reach the buoys during the strongest wind situations as reflected in our trajectory computations. Due to the SAL being elevated, surface friction cannot help create turbulence as in winter. Therefore, the question arises whether the shear above or below the wind maximum is strong enough to mechanically stir the atmosphere. Dust radiative heating in the layer could potentially modify vertical stability to sustain turbulence and thus vertical motion. Given the high dust loadings typical for summer and assuming some level of turbulence, tribo-electrification may also play a role, as indicated by balloon measurements from Cape Verde (Nicoll et al., 2011). In contrast to winter, however, the decoupling from the surface suggests overall lower levels of turbulence. Therefore, it is conceivable that vertical transport in convective systems is needed in addition to support long-range transport.

In summer the buoys are close to the Atlantic ITCZ (Žagar et al., 2011), where convection is frequent. In August the tropical cyclone season also intensifies (e.g. Kimball and Mulekar, 2004). Convective cells can uplift individual particles to the high tropical tropopause at altitudes of about 15 km (Fink et al., 2004; Takemi, 2005; Xu et al., 2018). Satellite observations have shown that dust

particles can escape areas of deep convection and exit the systems from upper layers, although significant amounts are washed or rained out (Sauter and L'Ecuyer, 2017). Such transports have the disadvantage of lifting particles out of the strongest horizontal winds at mid-levels, which would increase the overall travel time, although a little closer to the equator a second wind maximum, the so-called tropical easterly jet, exists near the tropopause (Fink et al., 2004).

To test this effect, a simple experiment was conducted. Five-day backward trajectories were calculated (see Methods) that incorporate a forced, immediate uplift to the upper troposphere (at 150 hPa) simulating deep convection. Using a settling velocity of 400 mm s^{-1} for a 100 μm dust particle (Table 5.1; Bagnold, 1941), we calculated the number of times such a dust particle would have to be lifted to the tropopause and subsequently settle back to sea level to arrive at the mid-ocean sampling sites. The results reveal that summer conditions require the fewest uplift cycles to cover the source-to-sink distance: in July and August, a dust particle of 100 μm has a 1/400 chance of reaching sampling site M3 with only four repeated uplifts (Fig. 5.2D). Chances increase greatly with increasing amount of uplift cycles along the trajectory. For the same particle to reach M4, a minimum of seven uplift cycles would be required, which is reached in August on only a few occasions (Fig. 5.2D).

According to satellite estimates, every uplift cycle reduces the mass of dust by about a factor of 60, as 1.6% ($\pm 0.7\%$) of the Saharan dust layer mass can escape the deep-convective cloud between 8 and 12 km (Sauter and L'Ecuyer, 2017). This would result in a minimum dilution of dust particles of nearly $1.3 \cdot 10^7$ to station M3 and $2.8 \cdot 10^{12}$ to M4, as most of the dust uplifted by convection is removed by wet deposition (Zender et al., 2003; Sauter and L'Ecuyer, 2017). As initial giant (37.5–300 μm) dust particle concentrations within 12 hours of uplift are estimated by Ryder et al. (2013a) to be up to $12,000 \text{ particles m}^{-3}$, these estimates suggest that this mechanism alone would unlikely explain the observed long-range transport, as the minimum number of uplift cycles would result in concentrations of $7.9 \cdot 10^{-4}$ and $3.2 \cdot 10^{-9}$ dust particles per cubic meter of air, respectively. Therefore, the convection would either need to be combined with effects of turbulence and charge, or several convective uplifts would need to occur within the same long-lived convective system.

Possible scenarios for this re-entrainment in a convective cell are long-lived squall lines from West Africa that transport particles out through the stratiform region to the east and re-enter through the rear inflow jet (Peters and Tetzlaff, 1988). Such a scenario would also benefit from a strong African easterly jet and associated shear. An alternative mechanism is lifting in tropical cyclones, e.g. up in the eyewall, out to the west of the system near the tropopause and re-integration after sedimentation to lower levels. This scenario would be more likely in areas of less shear, as shear tends to be detrimental to tropical cyclone development, but would benefit from the very long lifetime of tropical cyclones. However, both the convective uplift and the electric charge are affected by precipitation processes, as charge promotes the removal of dust by cloud droplets (Nicoll et al., 2011), and most dust uplifted by convection

is deposited by wet deposition (Zender et al., 2003; Sauter and L'Ecuyer, 2017). Therefore, convection and charging could also potentially work against each other.

5.3 Discussion

We presented evidence that giant mineral dust particles are transported through the atmosphere across the Atlantic Ocean, thousands of kilometres from their north African sources. We have evaluated four possible mechanisms that could aid this long-range transport. The best option for dust particles to be transported over great distances is within the turbulent SAL in summer, as this elevated atmospheric layer facilitates conditions optimal for the proposed mechanisms. Firstly, this air layer is characterized by strong winds (Adams et al., 2012; Tsamalis et al., 2013; Chouza et al., 2016), and turbulence created in this layer in combination with high particle concentrations allows for tribo-electrification of dust particles, compensating the particles' weight. In summer, long-range transport can be further facilitated by deep convective clouds, lifting dust particles to the upper troposphere, also due to the more northern position of the intertropical convergence zone. Long-range transport of giant mineral dust particles in winter seems less likely, although data suggests that it does occur, albeit in a lower amount.

Our analysis has shown that highly optimal conditions need to be met to make the transport we observe possible, and many details about the mechanisms we investigated are still unquantified. More theoretical, laboratory, field, and modelling work is required to substantiate our estimates, and a more detailed study of dust collected at long distances from the source should give more information on the seasonality of giant particle transport and particle concentrations. We would like to propose several future research directions to further investigate the possibilities and constraints of the discusses mechanisms.

1. The dust-collecting buoys, which actively sample dusty air through a carousel of filters, will produce a time-series of dust concentrations and giant particle counts, parallel to meteorological observations such as wind speed and wind direction, which will be crucial for the understanding of the magnitude of occurrence of long-range transport of giant particles.
2. In-situ measurements of particle charge and electric fields in combination with dust particle size and concentrations would help the quantification of the effect that charge has on the horizontal transport distance of giant dust particles.
3. A more detailed meteorological analysis of e.g. the occurrence of large convective cells along the Saharan dust-plume trajectory could quantify the possibility of the giant dust particles being transported by convective cells, and whether some kind of re-entrainment into moving convective systems takes place that could enable particles to be uplifted several times in the same long-lived system.

4. More theoretical, field, lab and modelling work on quantifying the effect of turbulence on the survival of giant particles could help to understand this process, and the effect on the number of particles being transported over long distances.

5. If there would be a better understanding of the occurrence and effect of these mechanisms, these should ultimately be incorporated into climate models and allowing the long-range transport of giant mineral dust particles, rather than a priori restricting to the transport of particles <10 µm. As a result of incorporating giant particles into earth-system models, many processes will be improved, in particular estimations of the atmospheric radiative balance, and in turn the atmosphere's global energy budget, which are highly impacted by the coarse dust fraction.

Table 5.2. Sampling duration of MWAC samplers on the dust-collecting buoys at M3 and M4 (Fig. 5.2) together with statistics on the co-located wind measurements.

Sampler	Sampling start	Sampling end	Days sampled	Min. wind velocity (m s ⁻¹)	Max. wind velocity (m s ⁻¹)	Average wind velocity (m s ⁻¹)
2014-M3	24-11-2013	01-09-2014	281	1.9	13.6	8.7 ± 2.0
2014-M4	28-11-2013	27-01-2015	425	4.5	14.2	9.1 ± 1.8
2015-M3	22-01-2015	29-03-2016	432	0.9	12.7	6.7 ± 1.7
2015-M4	28-01-2015	25-03-2016	422	0.4	14.2	8.2 ± 2.2

5.4 Materials and Methods

5.4.1 MWAC Samplers

In conjunction with the sediment traps described in Van der Does et al. (2016), two moored dust-collecting surface buoys were deployed at two of the sampling stations, M3 (12.39°N, 38.63°W) and M4 (12.06°N, 49.19°W; Fig. 5.2), for two consecutive years (Table 5.2). These buoys were equipped with one MWAC (Modified Wilson and Cooke; Goossens and Offer, 2000) sampler each, a passive air sampler that sampled continuously over the time the buoys were deployed. Our MWAC samplers consisted of a plastic bottle with an inlet and outlet tube of 7.5 mm diameter. They were installed vertically about three meters above sea level, while a wind vane ensured windward orientation. The MWAC samplers have sampling efficiencies between 75% and 105% for dust with a median particle size of 30 µm, meaning that in some occasions an oversampling would occur. However, Goossens and Offer (2000) conclude that the MWACs are the least inefficient samplers. Sampling efficiency varies slightly with different wind speeds, but without apparent trends (Goossens and Offer, 2000). For sand-sized particles with median grain sizes between 132 and 287 µm the samplers have slightly higher efficiencies of 90% to 120%, which are constant and independent of wind speed for velocities between 6.6 and 14.4 m s⁻¹ (Goossens et al., 2000). Maximum wind velocities at the sampling stations approximated 14 m s⁻¹ (Table 5.2). We found these samplers to

be best suited for our sampling purposes, as their sampling efficiencies are good, the mechanism is extremely simple, and the samplers are very inexpensive.

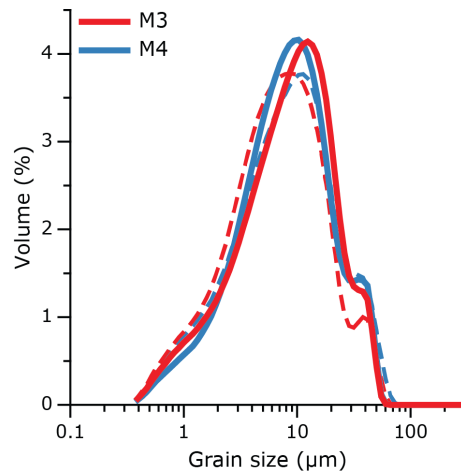


Figure 5.4. Grain-size distributions of MWAC samples collected in 2014 (solid lines) and 2015 (dashed lines).

The MWAC samplers collected a discrete Saharan dust sample over the entire sampling period. Besides mineral dust, the samples inevitably also contained sea salts. These were removed by rinsing the sample bottle with Milli-Q water, and subsequently filtrating over a 25 mm polycarbonate filter with a pore size of 0.4 μm (2014 samples), and 47 mm polycarbonate filters with a pore size of 0.2 μm (2015 samples). These filters were then qualitatively analysed with a light microscope for the presence of giant mineral dust particles and photographed. Grain-size distributions of the complete MWAC samples were obtained using a laser particle size analyzer *Coulter LS13 320*, using the method described by Van der Does et al. (2016) (Fig. 5.4). This figure shows the limitations of this method of obtaining information on giant particles, which are present in numbers which are below the detection limit of the laser particle sizer, and thus not registered in the grain-size distributions, which shows particles up to only 80 μm (Fig. 5.4).

We have tried to eliminate any possible contamination of giant particles to our dust samples. Microscope analysis (Fig. 5.1) confirms the giant particles to be mineral dust, and not e.g. plastic fragments from the sampling bottle, not salt crystals (which have a very typical cubic mineral shape), nor glass shards from glass beakers and petridishes (which would have very angular shape, with typical glass-fracture features). In addition, we can see that some of these particles have some sort of iron coatings, typical for Saharan dust particles. Other sources of sediment can be excluded since these samples were collected

in the middle of the Atlantic Ocean, directly from the air. Therefore, the possibility of contributions from the ocean or riverine input can be excluded.

The moored dust-collecting surface buoys are also equipped with a carousel of 24 filters through which a pump actively samples air, and as a result the dust can be collected on these filters on a much higher resolution (Van der Does et al., in preparation). Unfortunately, this active sampling proved to be unsuccessful as all the filters were retrieved ruptured, and future sampling campaigns will allow for a much more detailed study of the occurrence of giant mineral dust particles (seasonality, giant particle concentration in air, etc). The sampling is done in parallel with meteorological observations such as wind speed and wind direction. Also, additional upgrades of the buoys will include wet-deposition samplers.

5.4.2 Estimation of Travel Paths Based on Stokes' Law

Typically, dust is lifted to a maximum altitude of about 7 km within the SAL (Tsamalis et al., 2013). If an individual 100 μm dust particle was uplifted to 6.5 km at wind speeds of 25 m s^{-1} , and subject to a settling velocity of 0.4 m s^{-1} based on Stokes' Law (Bagnold, 1941), it would be transported ~400 km horizontally before reaching sea level. This is only about halfway to the most eastern sampling station M1 (12°N, 23°W; Van der Does et al., 2016), 700 km from the west African coast, demonstrating that horizontal transport alone is insufficient by far for the long-range transport of even larger dust particles, such that some additional mechanisms are needed to keep these dust particles aloft.

5.4.3 Trajectory Calculation for Giant Mineral Dust Particles

Five-day backward trajectories were calculated using Lagranto (Wernli and Davies, 1997; Sprenger and Wernli, 2015) for the ten-year period between 2006 and 2015 in order to obtain a robust climatological estimate. Lagranto is a Lagrangian trajectory analysis tool, which solves the following trajectory equation:

$$\frac{d\vec{x}}{dt} = \vec{u}(\vec{x}) \quad (1)$$

With $\vec{x} = (\lambda, \phi, p)$ being the position vector in geographical coordinates (longitude, latitude, height in pressure coordinates) and $\vec{u} = (u, v, w)$ being the 3-D wind vector (zonal, meridional, vertical). Lagranto is driven by 6-hourly ERA-Interim (Dee et al., 2011) 3-dimensional wind fields. The trajectories were started every six hours at the locations of buoys M3 and M4 (see Fig. 5.2), resulting in a total of about 1,200 trajectories per month. Trajectories that potentially carry dust are determined on the basis of their passage through a target region defined here as the area between 12°N and 35°N, and between 18°W and 30°E (Fig. 5.2B).

Three types of experiments were conducted:

1. Transport within the boundary layer: In winter time, dust usually remains in the lowest 1.5 km (Stuut et al., 2005; Tsamalis et al., 2013; Friese et al., 2016). In this layer, the atmosphere is characterized by the near-surface northeasterly trade winds over the ocean. Fastest transport from Africa to M3 and M4 can be expected in this layer, as winds become increasingly westerly with height in winter (Tomas and Webster, 1994). Therefore, in order to estimate the shortest possible traveling time (assuming no sedimentation), trajectories were started at 50 hPa above M3 and M4 and computed with the 3-dimensional ERA-Interim wind fields. This was only done for February when wintertime dust reaches its westernmost extension over the ocean (Klose et al., 2010).

2. Transport within the SAL: In summer, fastest transport occurs within the SAL associated with the western extension of the African easterly jet with its core around 600 hPa. To test shortest possible travel times, we therefore computed trajectories starting from this level over M3 and M4 during August, when the jet reaches its northernmost position and is fully developed (Afiesimama, 2007).

3. Transport affected by convective lifting: In summer dust travels in the vicinity of the ITCZ and could therefore be lifted within convective updrafts. To estimate the effect of this on travel time, also taking into account sedimentation, a simple thought-experiment was conducted. The vertical velocity (ω) field in ERA-Interim was modified to reflect the settling velocities of dust particles. A constant value of 400 mm s⁻¹ (1.44 km hr⁻¹), typical for particles of 100 µm in diameter (Bagnold, 1941) was added globally. This settling velocity was converted into pressure coordinates, as ERA-Interim defines vertical velocities in Pa s⁻¹ using equation 2:

$$\omega = -\rho g w \quad (2),$$

where ω is vertical velocity in pressure coordinates, ρ density, g the gravitational constant, and w the vertical velocity in height coordinates. The density ρ was calculated using equation 3:

$$\rho = \frac{p}{RT} \quad (3),$$

where p is pressure, R the ideal gas constant for dry air ($R_d = 287 \text{ J kg}^{-1} \text{ K}^{-1}$), and T temperature from the U.S. Standard Atmosphere (1976).

When calculating backward trajectories, air parcels ascended rapidly due to the effect of sedimentation. Upon crossing the tropopause (defined here as 150 hPa), parcels were immediately set down to 950 hPa, mimicking the (backwards) effect of a convective updraft. The number of convective updrafts needed for the

parcel to reach the target region were counted. The winter months did not yield any significant results due to the predominance of westerlies at upper levels.

5.4.4 Charge Effects

The effect of an electric force on the fall speed of a particle has been considered using a simple model of a spherical particle of radius r in a vertical electric field E , with weight W , an Archimedian upthrust U , an electric force F_E and a drag force F_D . If the particle is moving downwards but experiencing an upward electric force (due to carrying a negative charge in a down-ward directed field, such as that in fair weather), the balance of forces in equilibrium can be represented as

$$U + F_E + F_D = W \quad (4).$$

The ratio of U to W is given by the ratio of the densities of air ρ_a and the particle ρ_p , which is $\sim 1:1000$, hence $U \ll W$, and U can be neglected. The drag force depends on the particle's projected area $A (= \pi r^2)$, its fluid-relative speed v and the drag coefficient C_D . Assuming a spherical particle carrying a charge q , the electric force can be written as qE , the drag force parameterized as $\frac{4}{3}\pi r^3 \rho_p g$, hence the equilibrium description of equation (4) becomes:

$$qE + C_D \frac{1}{2} A \rho_a v^2 = \frac{4}{3} \pi r^3 \rho_p g \quad (5).$$

For calculation of v , C_D depends on the flow and the associated Reynolds number and typically varies from 0.5 to 1.5 for a smooth sphere.

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Chapter 6:

Seasonality in Saharan Dust Particle Characteristics Across the Atlantic Ocean: Transport Versus Deposition

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Ready to be submitted to Aeolian Research

Abstract

Saharan dust is transported in great quantities from the North African continent every year, most of which is deposited in the North Atlantic Ocean. This dust impacts regional and global climate, by affecting the atmospheric radiation balance and altering ocean carbon budgets. Nonetheless, little research has been done on Saharan dust collected in-situ across the open Atlantic. Here we present a time series of dust collected along a trans-Atlantic transect, by both subsurface sediment traps and moored dust-collecting surface buoys. The results show a good correlation between the atmospheric dust and the lithogenic particles settled in the sediment traps, confirming the aeolian origin of the lithogenic particles intercepted by the sediment traps, even in the western part of the Atlantic Ocean. The sediment traps, collecting dust deposited by both dry and wet processes, show increased deposition fluxes in summer, coinciding with increased precipitation at the sampling sites. In contrast, the buoys that collect transported dust at sea level, show little seasonal variation in both dust concentration and particle size, as the large amounts of dust transported in summer at high altitudes are outside their sampling range. This implies that the large amounts of wet deposition in summer determines the seasonality of both the deposition flux as well as its particle size in the sediment traps.

6.1 Introduction

Saharan dust is exported in large quantities from the African continent and transported across the Atlantic Ocean towards the Caribbean, about 182 Tg annually (Yu et al., 2015a). This atmospheric dust can alter radiation budgets (Ryder et al., 2013b) and can act as cloud condensation and ice nuclei (Wilcox et al., 2010; Atkinson et al., 2013). When deposited in the ocean, nutrients carried by the dust enhance the ocean's carbon cycle as they stimulate phytoplankton growth, and enhance carbon export to the sea floor by acting as mineral ballast particles that accelerate settling of organic matter aggregates (Martin and Fitzwater, 1988; Armstrong et al., 2002; Bressac et al., 2014; Pabortsava et al., 2017; Van der Jagt et al., 2018; Chapter 4). Most studies focused on sampling Saharan dust on land (Kandler et al., 2009; Marticorena et al., 2010; Skonieczny et al., 2013; Friese et al., 2017), in the eastern part of the North Atlantic Ocean (Ratmeyer et al., 1999b; Bory et al., 2002; Brust et al., 2011; Fischer et al., 2016; Friese et al., 2016), or in the Caribbean and North and South America (Prospero et al., 2010; Prospero et al., 2014; Yu et al., 2015b). Atmospheric dust has been sampled over and across the Atlantic Ocean by airplane campaigns (Weinzierl et al., 2017) and shipboard sampling (Stuut et al., 2005). Especially the field campaigns are limited in temporal resolution, whereas the studies at monitoring stations are inherently limited on a spatial scale. By combining results from these studies, we conclude that in the east, close to the northwest African sources of the dust, deposition fluxes peak in winter, and in the west, at distal sites in the Caribbean, deposition maxima occur in summer. This seasonality is attributed to the latitudinal movement of the Intertropical Convergence Zone (ITCZ), influencing the prevailing dust-transporting wind systems (Nicholson, 2000; Adams et al., 2012). The Saharan Air Layer (SAL) transports the dust

between 5 and 7 km and with high wind velocities in summer, as opposed to the Harmattan winds that transport the dust at lower altitudes (0–3 km) in winter (Stuut et al., 2005; Adams et al., 2012; Tsamalis et al., 2013; Kanitz et al., 2014).

Dust emissions from northwest Africa show large inter-annual variability, and the amount of dust deposition greatly depends on the distance to the source (Schütz, 1980; Sarnthein et al., 1981; Goudie and Middleton, 2001). The particle size of deposited dust is also related to the distance to the source, with dust deposited at proximal locations being coarser-grained than dust deposited at distal locations (Holz et al., 2004; Van der Does et al., 2016). This is in turn related to particle shape and density, due to the larger settling velocities of more solid quartz particles compared to platy clay minerals and micas, affecting both the vertical and horizontal distance over which the particles are transported (Glaccum and Prospero, 1980; Stuut et al., 2005; Mahowald et al., 2014). Furthermore, conditions in the source, such as wind velocity, erosion threshold, and soil particle size, determine the size of the emitted dust (Marticorena, 2014).

In-situ observations of dust deposition across the Atlantic Ocean are sparse. A unique long-term sampling campaign, with subsurface sediment traps along a transect at 12°N, directly underneath the main path of the Saharan dust plume, was started in 2012. Results from the first two years of sampling show a significantly decreasing dust deposition flux down-wind of the North African sources (Chapter 4). At the same time, dust particle size decreases downwind (Van der Does et al., 2016), related to a decreased contribution of quartz particles and a relative increase of clay minerals to the west (Korte et al., 2017). In addition, the data revealed a distinct seasonal pattern of coarser dust particles deposited in summer (Van der Does et al., 2016) and increased fluxes as a result of wet deposition (Chapter 4). Furthermore, dust deposited in the Atlantic can provide fertilizing nutrients enhancing productivity of calcifying phytoplankton (coccolithophores) as found in the sediment traps (Guerreiro et al., 2017). Here we present results from the second year of sampling by the sediment traps along the Atlantic transect of dust particle size, and combine these with the dust deposition fluxes (Chapter 4). In addition, we present two years of atmospheric dust concentrations and dust particle size at two of the stations along the transect, collected by moored surface buoys.

6.2 Material and Methods

Saharan mineral dust was sampled between October 2012 and March 2016 along a longitudinal transect crossing the Atlantic Ocean using three sampling methods. At five sites (M1–M5) along a trans-Atlantic transect at 12°N, dust was collected by subsurface sediment traps moored at 1,200 and 3,500 m below sea level (BSL, Fig. 6.1). For the second year of sampling the transect was expanded with two surface buoys moored at two stations in the middle of the transect (M3 and M4), which sampled dust directly from the atmosphere. These buoys were equipped with a dust collector holding 24 sample filters

actively filtering the air, and additionally one passive air sampler (MWAC – Modified Wilson and Cooke sampler; Goossens and Offer, 2000) each. Combining these different sampling methods yields important information about depositional processes, while the multi-year record allows for identification of seasonal and inter-annual variability of dust transport and deposition.

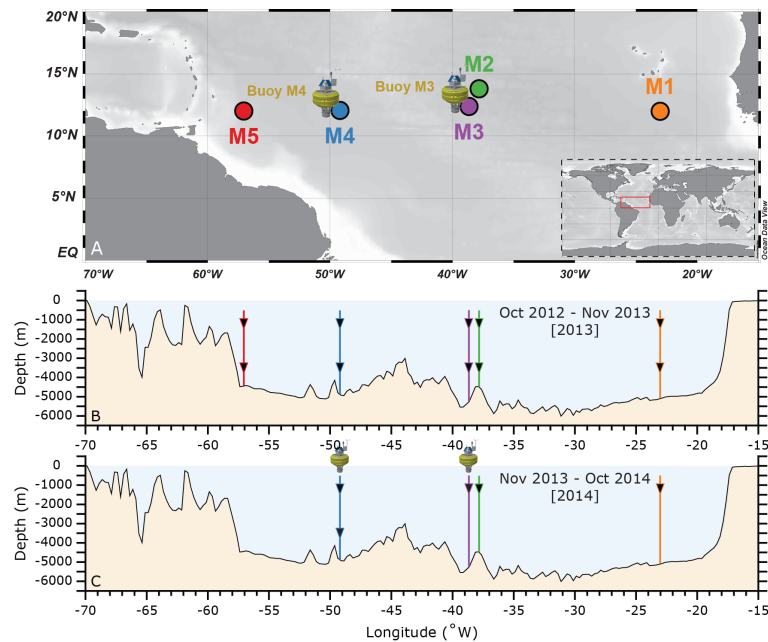


Figure 6.1. A: Map with locations of sediment-trap sampling stations M1–M5, with dust-collecting moored surface buoys at M3 and M4, in the Atlantic Ocean. B,C: Bathymetry along 12°N (source: gebco.net) with successful sediment trap sampling of the 2013 and 2014 sampling series.

6.2.1 Sediment Traps

Sediment traps were moored at five stations along a transect across the Atlantic Ocean, as described by Van der Does et al. (2016). Four moorings were located at 12°N, and a fifth at 13°N (Fig. 6.1A). Sediment traps were located at 1,200 (upper, or U) and 3,500 (lower, or L) meter below sea level (BSL) on each mooring, and consisted of 24 sampling bottles, pre-programmed to synchronously sample over 8 and 16-day intervals (Table 6.1). During the first year the traps sampled from 19 October 2012 to 7 November 2013 at all five sites (henceforth called 13M1–13M5) at 16-day intervals. Dust particle sizes were presented by Van der Does et al. (2016), and dust fluxes in Chapter 4. During the second sampling year the traps sampled from 23 November 2013 until 25 October 2014 at M1–M4 (henceforth called 14M1–14M4), at 8 (N=6–9) and 16 (N=15–18) day intervals. This variable sampling resolution was applied to minimize the time without sampling, and to synchronise sediment-trap

sampling at all stations. The set up was slightly different for the two sampling years, with seven sediment traps recovered after the first sampling year, and five after the second sampling year (Fig. 6.1A & B). Detailed information on the mooring equipment and sampling intervals is given in the cruise reports (Stuut et al., 2012; Stuut et al., 2013; Stuut et al., 2015). For 14M1-U only the first nine samples out of 24 could be recovered due to rotation malfunction of the trap carousel. Sediment-trap sample preparation is described in more detail by Van der Does et al. (2016).

Table 6.1. Sampling positions, intervals, average particle size and dust content (deposition flux or dust concentration, if available) of sediment traps of 2013 (13M1–M5) and 2014 (14M1–M4); buoys of 2014 (M3-A and -B, M4-A) and 2015 (M3-C and M4-C); and MWAC samplers of 2014 and 2015. BSL = below sea level.

Sampler	Lat. (° N)	Lon. (° W)	Trap depth (m BSL)	Km to African coast	Sampling intervals	Sampling period	Avg. modal grain size	Avg. dust content
13M1	12.00	23.00	1,150	700	16 days	19 Oct 2012 – 7 Nov 2013	18.25 μm^b	14.27 $\text{mg m}^{-2} \text{d}^{-1c}$
13M2	13.81	37.82	1,235; 3,490	2,300	16 days	19 Oct 2012 – 7 Nov 2013	13.85 μm^b	3.11 $\text{mg m}^{-2} \text{d}^{-1c}$
13M3	12.39	38.63	3,540	2,400	16 days	19 Oct 2012 – 7 Nov 2013	16.47 μm^b	4.64 $\text{mg m}^{-2} \text{d}^{-1c}$
13M4	12.06	49.19	1,130; 3,370	3,500	16 days	19 Oct 2012 – 7 Nov 2013	10.39 μm^b	3.72 $\text{mg m}^{-2} \text{d}^{-1c}$
13M5							6.99 μm^b	8.69 $\text{mg m}^{-2} \text{d}^{-1}$
14M1	12.00	23.01	1,200	700	8 & 16 days	23 Nov 2013 – 31 Mar 2014	15.20 μm^a	9.46 $\text{mg m}^{-2} \text{d}^{-1ac}$
14M2	13.81	37.82	1,190	2,300	8 & 16 days	1 Dec 2013 – 25 Oct 2014	13.35 μm	2.86 $\text{mg m}^{-2} \text{d}^{-1c}$
14M3	12.40	38.63	1,362	2,400	8 & 16 days	1 Dec 2013 – 17 Oct 2014	12.85 μm	3.66 $\text{mg m}^{-2} \text{d}^{-1c}$
14M4	12.04	49.22	1,110; 3,316	3,500	8 & 16 days	9 Dec 2013 – 25 Oct 2014	11.10 μm	2.40 $\text{mg m}^{-2} \text{d}^{-1c}$
Buoy 2014 M3-A	~12.3	~38.7	-	2,400	8 & 16 days	1 Dec 2013 – 20 Apr 2014	-	-
Buoy 2014 M3-B	~12.3	~38.7	-	2,400	8 days	7 Sep 2014 – 21 Jan 2015	-	-
Buoy 2015 M3-C	~12.3	~38.7	-	2,400	11 & 22 days	26 Jan 2015 – 29 Mar 2016	19.9 μm	0.0198 $\mu\text{g L air}^{-1}$
Buoy 2014 M4-A	~12.0	~49.1	-	3,500	15-16 days	9 Dec 2013 – 30 Mar 2014	-	-

Sampler	Lat. (° N)	Lon. (° W)	Trap depth (m BSL)	Km to African coast	Sampling intervals	Sampling period	Avg. modal grain size	Avg. dust content
Buoy 2015 M4-C	~12.0	~49.1	-	3,500	11 & 22 days	6 Feb 2015 – 21 Mar 2016	24.2 μm	0.0082 μg L air^{-1}
MWAC 2014 M3	~12.3	~38.8	-	2,400	Continuously (281 days)	24 Nov 2013 – 1 Sep 2014	8.54 μm	0.05 mg day^{-1}
MWAC 2014 M4	~12.0	~49.1	-	3,500	Continuously (425 days)	28 Nov 2013 – 27 Jan 2015	11.29 μm	0.04 mg d^{-1}
MWAC 2015 M3	~12.3	~38.8	-	2,400	Continuously (434 days)	22 Jan 2015 – 31 Mar 2016	12.4 μm	0.13 mg d^{-1}
MWAC 2015 M4	~12.0	~49.1	-	3,500	Continuously (422 days)	28 Jan 2015 – 25 Mar 2016	10.29 μm	0.07 mg d^{-1}

^a Average of nine samples

^b Published by Van der Does et al. (2016)

^c Upper traps (1,200 m) as in Chapter 4.

A mid-date was assigned to each sample interval, which is the middle of their 8 or 16-day sampling periods. The samples were grouped into seasons: autumn (September, October, November), winter (December, January, February), spring (March, April, May), and summer (June, July, August). Organic aggregates in which dust particles are incorporated have average settling velocities of about 200 m day^{-1} (Nowald et al., 2015), although much higher settling velocities up to 400 m day^{-1} can be reached during high dust deposition conditions (Van der Jagt et al., 2018). Therefore, we assume an average delay of 6 days for the aggregates to settle from the ocean's surface to the sediment traps at 1,200 m depth, and another 11.5 days to the sediment traps at 3,500 m depth. This is in agreement with previously determined settling velocities for these sediment-trap samples of $>140 \text{ m day}^{-1}$ (Van der Does et al., 2016; Korte et al., 2017).

6.2.2 Buoys and MWAC Samplers

In conjunction with the sediment traps, two dust-collecting surface buoys were moored at sampling stations M3 and M4 (Fig. 6.1), nicknamed buoy Michèle and buoy Laura, respectively. These were initially deployed in November 2013 (Buoys M3-A and M4-A; Stuut et al., 2013), subsequently re-deployed in 2014 (Buoy M3-B; Bale, 2014; and Buoys M3-C and M4-C; Stuut et al., 2015), and recovered in 2016 (Stuut et al., 2016). The buoys were built at NIOZ based on the aerosol-collecting buoys designed by E. Sholkovitz (e.g. Sholkovitz et al., 2001; Sholkovitz and Sedwick, 2006) and are still developing. The buoys consist of a carousel with 24 filters which rotate under the air inlet (Fig. 6.2), synchronously with the subsurface sediment traps at 8 to 22 day sample intervals (Table 6.1). They collected aerosols by pumping air through the 47 mm polycarbonate filters of 0.2 μm pore size. These aerosols predominantly consisted of mineral dust but also inevitably contained variable amounts of sea salts from spray. The volume of air filtered by the sampler

was monitored, and the buoys were linked to a meteorological sensor that monitored wind speed, wind direction, temperature, precipitation and relative humidity, which allowed the buoys to only sample under favourable weather conditions, i.e. no precipitation ($<0.2 \text{ mm min}^{-1}$) or strong winds ($<20 \text{ m s}^{-1}$). Under these conditions, sampling was carried out once daily starting at 10:00 UTC for two hours (in 2014) and four hours (in 2015). If weather conditions were not favourable, the air inlet did not open while the weather kept being monitored for a period of eight hours. When weather conditions then changed to favourable for at least one hour, the sampling scheme was initiated. When during sampling the weather conditions changed to unfavourable conditions the schedule was aborted for another hour until another period of favourable conditions, limited to 18:00 UTC. The buoys were also equipped with a wind vane to keep the air inlet facing the wind, which was mostly blowing from an easterly direction, and with a satellite communications system (Iridium) that allowed two-way communication with the buoys.

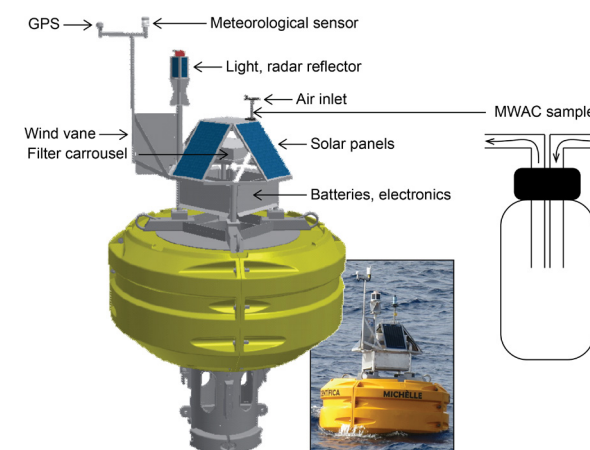


Figure 6.2. Drawing and photo of dust-collecting buoy (left) and MWAC sampler (right).

For each filter, an average of 2,288 and 5,914 L of air per day was sampled by buoys M3-C and M4-C, respectively, and each session the volume decreased as the filter was becoming more clogged by dust and sea salt. Due to technical problems with the power supplies of the buoys, the entire filter routine was not completed for every deployment. In 2014, all buoy filters were recovered broken. Consequently, the dust collectors were modified for the second deployment in 2015, which included an additional support filter underneath the sample filters, and a buffer chamber in the air-flow line to reduce oscillations of the filter caused by the membrane pump (Stuut et al., 2015). Also, the volume of air sampling was increased for the second year, as the first year revealed that the solar panels supplying power to the pumps worked very efficiently. The next year (2015), all filters returned intact, and dust particle size and dust concentration could be determined. As a consequence of the

broken filters, the dust concentration for the 2014 filters could not be determined, and not all broken filters contained sufficient material for particle-size analysis.

In addition to the filters, both buoys were equipped with an MWAC sampler, a passive air sampler that sampled continuously over the time the buoys were deployed (Fig. 6.2). The MWAC sampler consists of a plastic bottle which acts as settling chamber, and is connected to an inlet and outlet tube with a 7.5 mm diameter. The samplers were installed vertically on the buoy's air inlet. The buoy's wind vane ensured continuous windward orientation. For Buoy M3, the MWAC sampler was deployed from 24 November 2013 until 1 September 2014, comprising winter 2013-2014, spring and summer 2014. The MWAC sampler on buoy M4 sampled from 28 November 2013 until 27 January 2015, comprising winter 2013-2014, spring, summer and autumn 2014, and most of winter 2014-2015 (Table 6.1). The samplers were located about 3 meters above sea level (Fig. 6.2). These MWAC samplers have very high sampling efficiencies, between 75 and 105% for dust with a median particle size of 30 μm , at wind velocities between 1 and 5 m s^{-1} (Goossens and Offer, 2000). Average wind velocity as recorded by buoy M3 (A-C) ranged between 4.1 and 9.9 m s^{-1} per sampling interval, with maximum velocities up to 18.3 m s^{-1} , well below the pre-set upper wind limit of 20 m s^{-1} . For buoy M4 wind-velocity data are largely unavailable due to a technical failure of the electronics: for buoy M4-A wind velocities were recorded only for the first seven filters with large parts of the data missing, and for buoy M4-C no data were recorded at all. Sampling efficiency varies slightly with different wind speeds, but without apparent trends. Also for sand-sized particles (median grain size of 132 – 287 μm), the efficiency reported in literature is slightly higher (90–120%) but constant and independent of wind speed, for velocities between 6.6 and 14.4 m s^{-1} (Goossens et al., 2000).

6.2.3 Grain-Size Analysis and Dust Masses

For grain-size analysis of the sediment-trap samples, 1/25 of the original sample was used. Prior to grain-size and flux analysis, all biogenic constituents—comprising organic matter, carbonates, and biogenic silica—were chemically removed from the sediment trap samples to isolate the dust fraction, following the procedure described by McGregor et al. (2009). After particle-size analysis, the sediment-trap samples were filtered over 25 mm diameter, 0.4 μm pore size polycarbonate filters and rinsed with Milli-Q water and subsequently weighed to obtain the dust mass and therefore deposition flux (Chapter 4).

Besides mineral dust, the buoy filters contained a significant amount of sea salt, which had to be removed from the buoy filters before analysis. The original 47 mm diameter, 0.2 μm pore size polycarbonate buoy filters were ashed in a low-temperature ashers and subsequently washed with Milli-Q water onto pre-weighed filters (25 mm diameter, 0.4 μm pore size polycarbonate), dried and post-weighed to obtain the mass of the dust fraction $>0.4 \mu\text{m}$. Next, the filters containing the dust were ashed again in order to retrieve the dust particles for particle-size

analysis. Some of the buoy's samples collected in 2013 did not contain sufficient material for reliable particle-size analyses. This resulted in six (M3-A), three (M3-B and M4-A), 14 (M3-C) and 20 (M4-C) reliable grain-size distributions.

The MWAC samples only required the rinsing of the dust from the sample bottle with Milli-Q water prior to grain-size analysis, at the same time dissolving the present sea salts. After particle-size analyses these samples were also filtered and weighed on 47 mm diameter, 0.4 μm pore size (2014-MWACs) and 0.2 μm pore size (2015-MWACs) polycarbonate filters to establish long-term-averaged mass.

Immediately prior to grain-size analysis of all samples, sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$) was added to disaggregate the particles. Particle-size distributions were obtained using a Coulter laser diffraction particle sizer (LS13 320) equipped with a micro liquid module (MLM), as described by Van der Does et al. (2016). A magnetic stirrer homogenized the sample during analysis, and degassed water minimized the influence of air bubbles on the measurements. This resulted in particle-size distributions ranging from 0.375 to 2000 μm , described by 92 size classes. This way both the particle-size analyses and deposition fluxes are of the same mineral-dust size fraction $>0.4 \mu\text{m}$.

6.2.4 Aerosol Optical Depth (AOD)

Aerosol optical depth (AOD) at the buoy sampling sites M3 and M4 ($1 \times 1^\circ$) was obtained at a daily resolution from MODIS Terra, through the Giovanni online data system, developed and maintained by the NASA GES DISC. From this dataset a 10-day running average was calculated to smoothen the data.

6.3 Results

6.3.1 Sediment-Trap Samples

Particle sizes of the first year of sampling (October 2012–November 2013) were described by Van der Does et al. (2016), who showed a distinct down-wind fining as well as seasonal differences, with coarser-grained dust deposition in summer, and finer-grained in winter and spring. For the second year of sampling (November 2013–October 2014) a similar down-wind fining is observed, as coarse particles are deposited closer to the source at the easternmost sediment trap at M1 (23°W), and finer-grained particles are transported over greater distances (Fig. 6.3). This downwind fining was also demonstrated by Korte et al. (2017), who showed for samples from the first year that quartz particles, which are generally coarser and heavier than clay minerals, are predominantly deposited at proximal sites. Also reflected in the second year of sampling is the seasonal signal of modal grain sizes with coarser dust particles deposited in summer, and finer particles in winter, with especially finer dust observed in spring. Overall, dust particle sizes from the lower (3,500 m) sediment traps show less seasonality for both years than the upper (1,200 m) sediment traps (Fig. 6.3B).

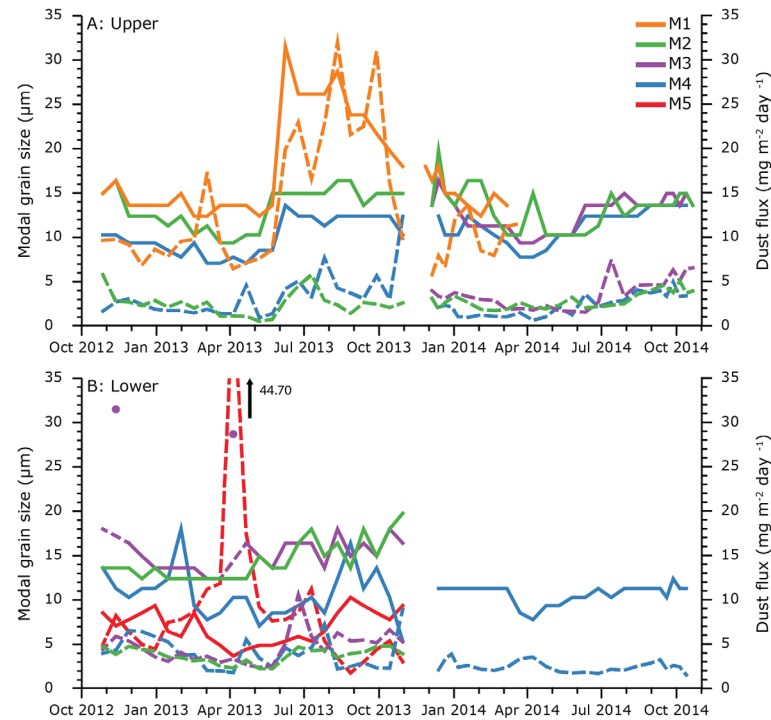


Figure 6.3. Modal grain sizes (solid lines) and dust deposition fluxes (dashed lines) of 2013–2014, for the upper traps at 1,200 m (A) and lower traps at 3,500 m (B).

Seasonal variations of dust deposition fluxes in the sediment traps match the particle-size trends, with higher deposition and coarser particles in summer and autumn, as opposed to lower deposition and finer particles in winter (Fig. 6.3). Dust deposition along the transect is also characterized by a downwind decrease, as most dust is deposited at the easternmost station M1, closest to the west African coast and the source of the dust (Chapter 4). Similar to the grain size of the dust, the lower sediment traps show a less clear seasonal signal in dust deposition flux than in the upper traps. In addition, the lower traps tend to show higher deposition fluxes than their upper counterpart during the same year (13M2, 13M4 and 14M4; Fig. 6.3). The deposition flux at 13M5-lower is much higher than expected for the down-wind decreasing trend, and shows a large outlier in April 2013 (Fig. 6.3B), which Korte et al. (2018a) attributed to downslope transport of sediment re-suspended from the nearby Barbados margin. At the same time, geochemical analyses revealed that input of Amazon-River sediments to the upper trap at M4 is minimal, if not absent (Van der Does et al., 2018).

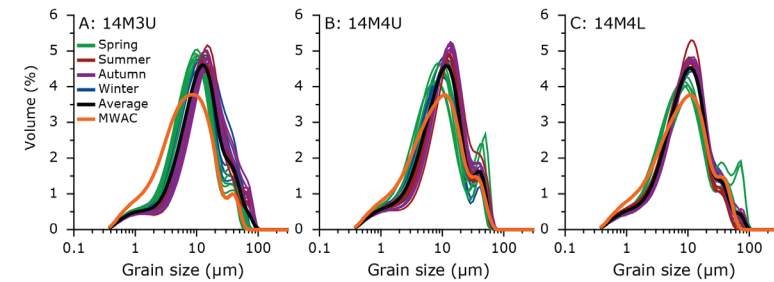


Figure 6.4. Grain-size distributions of 14M3-U / MWAC 2014-M3 (left; A), 14M4-U / MWAC 2014-M4 (middle; B), and 14M4-L / MWAC 2014-M4 (right; C).

6.3.2 MWAC Samplers

The MWAC samplers on both buoys collected dust in parallel with the sediment traps at stations M3 and M4 in 2014, only as a single discrete sample during their entire mooring period. Dust collected using the two different sampling methods show almost identical particle-size distributions (Fig. 6.4), although at station M3 the MWAC-sampler shows slightly finer dust than the sediment-trap average (Fig. 6.4A). The secondary mode or “shoulder” of the grain-size distributions is represented in both the MWAC samples and sediment trap samples, demonstrating the good agreement between the two different sampling methods. Not only the upper traps but also the dust in the lower trap at M4 resembles the atmospheric dust very well (Fig. 6.4C). This demonstrates that although modal grain sizes of the dust collected by both the upper and lower traps show differences in seasonality and absolute grain sizes (Fig. 6.2), the average annual grain-size distributions are very similar to the dust collected from the atmosphere directly above. Dust collected by the MWAC samplers in 2015 show the expected downwind-fining trend, with dust collected by buoy M3 being coarser-grained than dust collected further downwind by buoy M4 (Fig. 6.6).

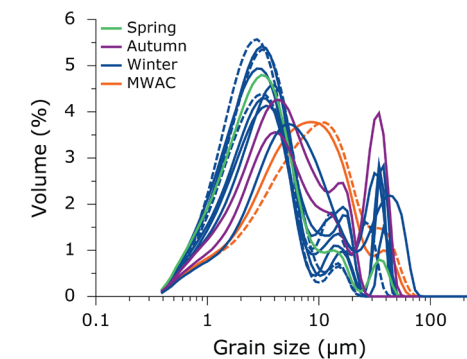


Figure 6.5. Grain-size distributions of 2014 buoy filters of M3-A & B (solid lines) and M4-A (dashed lines), and MWAC samples (orange).

Dust accumulation in the MWAC samplers is very low, ranging from 0.04 to 0.13 mg day⁻¹ (Table 6.1), which is not surprising, given the small opening of the samplers (7.5 mm diameter). In 2014 the difference in flux between the two buoys is very small, however during both sampling years the dust mass at the more proximal buoy M3 is larger than at the more distal buoy M4.

6.3.3 Buoy Filters

The (broken) buoy filters from 2014 (M3-A,B; M4-A) that had collected enough dust for reliable grain-size analyses were mostly collected during winter (Fig. 6.5), although some suffered from insufficient particles at the coarse end of the grain-size distributions resulting in less reliable peaks. Modal grain sizes range between 2.8 and 5.4 μm (first mode), and although fine-grained dust is expected in winter, they are much finer-grained than the finest-grained sediment-trap samples at M3 and M4 (Fig. 6.3). All samples show a secondary mode at approximately 15 μm , which could be the result of platy mica particles that have similar aerodynamic properties as smaller, spherical particles (Stuut et al., 2005). The buoy grain-size distributions show finer-grained dust than that collected by the MWAC samplers in 2014, which collected dust for an entire year or even longer (Fig. 6.5).

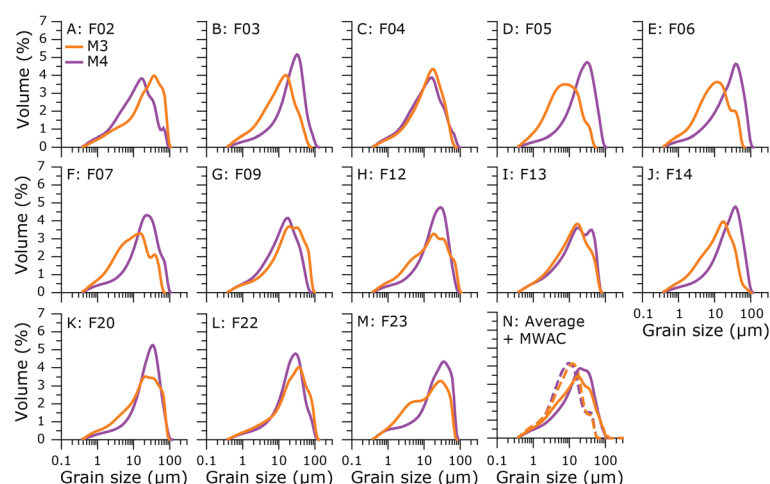


Figure 6.6. A–M: Grain-size distributions of buoy filters of M3-C (orange), and M4-C (purple) in 2015. N: Average grain-size distributions of buoys M3 (N=14) and M4 (N=20) and the two MWAC samples (dashed lines).

In 2015 most filters had collected enough dust for grain-size analysis (Fig. 6.6). Modal grain sizes range between 7.8 and 38.0 μm (average 19.9 μm) for eastern buoy M3, and between 14.9 and 38.0 μm (average 24.2 μm) for the more western buoy M4. Secondary shoulders in the grain-size distributions are mostly absent and buoy samples are coarser-grained than the MWAC samples (Fig. 6.6N). For many

samples, dust collected by the more downwind buoy M4 appears to be coarser-grained, but their average grain-size distribution is very similar (Fig. 6.6N).

In contrast to the subsurface sediment traps, the dust-collecting buoys measure the concentration of dust in the atmosphere, which is a measure for dust *transport*, as opposed to dust *deposition* over the ocean. In 2015, an average of 2,288 and 5,914 L of air were pumped through each filter per day for buoy M3-C and M4-C, respectively. Dust concentrations vary between 6.6 and 51.7 $\mu\text{g m}^{-3}$ (average 19.8 $\mu\text{g m}^{-3}$) for buoy M3-C, and between 2.5 and 33.0 $\mu\text{g m}^{-3}$ (average 8.2 $\mu\text{g m}^{-3}$) for buoy M4-C (Fig. 6.7). This downwind decrease in concentration is much more evident than the accompanying grain-size distributions and modal grain sizes (Fig. 6.6). Although there is no clear seasonal signal in the dust amounts over the year for either buoy, the individual peaks of increased dust concentrations match well between the two buoys, with a smaller amplitude and absolute concentrations at buoy M4. The dust concentrations also match the atmospheric dust load, represented by the aerosol optical depth (AOD) for most of the year, except for summer, when most dust is deposited by wet deposition (Chapter 4).

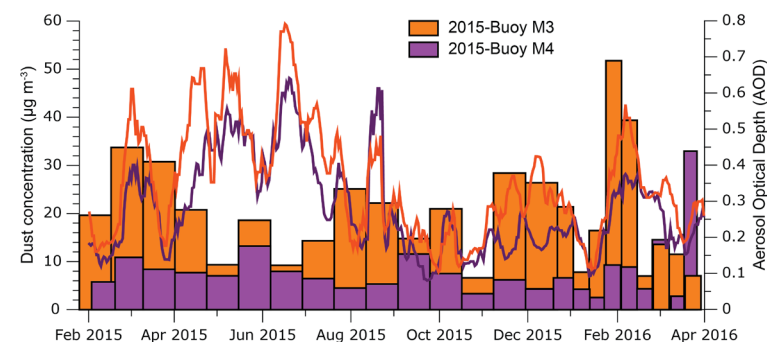


Figure 6.7. Atmospheric dust concentrations (bars) and aerosol optical depth (AOD, from MODIS Terra; lines) at buoys M3-C and M4-C (in 2015).

6.4 Discussion

Dust sampling for this study was carried out using three different methods. First, dust was directly sampled from the atmosphere by the dust-collecting buoys over regular time intervals, revealing dust concentrations during atmospheric transport. Second, passive MWAC samplers attached to these buoys took one discrete sample of atmospheric dust during the entire deployment period of the buoys. Dust entering these samplers by raindrops cannot be excluded entirely since the glass tubes of the samplers do allow rainwater to enter. Third, the subsurface sediment traps collected both dry- and wet-deposited dust continuously at 1,200 m and 3,500 m depth along the full transect, at synchronous time intervals with the dust-collecting buoys at the surface.

Combined, these methods show the relationship between dust concentrations during atmospheric transport, and dust fluxes after dry and wet deposition.

6.4.1 Atmospheric Dust: Transport Versus Deposition

There appears no clear seasonal signal for neither the atmospheric concentrations of dust nor the particle sizes during atmospheric transport, as sampled by the buoys. Contrastingly, the deposited dust in the subsurface sediment traps shows a clear seasonal trend of increased fluxes in summer with coarser-grained dust, and lower deposition fluxes with finer-grained dust in winter. In the summer season, generally more and coarser dust is transported across the Atlantic Ocean with stronger winds at higher altitudes (Van der Does et al., 2016; Friese et al., 2017), with a maximum altitude of 5 to 7 km across the Atlantic Ocean (Muhs, 2013; Fig. 6.8). These larger amounts of atmospheric dust are reflected by high AOD values (Fig. 6.7), but most of this dust is outside the sampling range of the surface buoys due to the altitude at which the SAL occurs. In winter, dust is transported at lower altitudes (Fig. 6.8), between 0 and 3 km over the eastern Atlantic and between 0 and 2 km over the western Atlantic (Muhs, 2013; Tsamalis et al., 2013). This dust is therefore preferentially collected by the surface buoys at sea level.

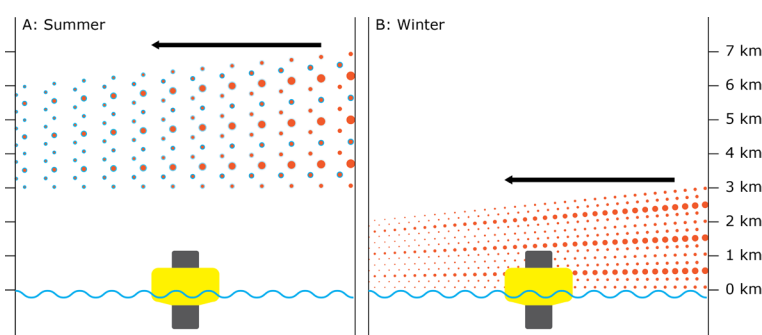


Figure 6.8. Schematic overview of summer and winter dust transport over the Atlantic Ocean. A: Summer scenario depicting greater amounts and coarse-grained dust by both dry and wet deposition in the east, and lower amounts and finer-grained dust by predominantly wet deposition in the east. Dust transport occurs at high altitudes, up to 7 km. B: same as the summer scenario, except the main mechanism over the full transect is by dry deposition. Dust transport occurs at lower altitudes, between 0 and 3 km.

A comparison of dust deposition fluxes in the sediment traps combined with precipitation data from satellites and with model simulations of dust deposition using a comprehensive earth system model (CESM) revealed that the main mechanism of dust deposition over the Atlantic Ocean is by wet deposition, mostly occurring in summer (Chapter 4). Over the eastern Atlantic, close to the source, dry deposition still plays a significant role in winter, but along the downwind transect, wet deposition progressively turns to be nearly the sole depositional process (Chapter 4), as illustrated by Figure 6.8. These precipitation

events in summer wash out large amounts of dust from the atmosphere, which are not sampled by the buoys as they stop sampling as soon as rain is detected by their weather sensors. This results in high (wet) deposition fluxes in the sediment traps, while at the same time low dust concentrations for the buoys at sea level. This wet deposition therefore seems to determine the seasonal signal observed in the sediment traps of both the dust particle size and the deposition flux.

Although the dust collected by the buoys lack a clear seasonal signal, inter-annual variations are large, with very fine-grained dust collected in winter 2014 and much coarser-grained dust throughout the entire year 2015 (Fig. 6.5 and 6.6), although the first sample set may be biased by the selective loss of (coarse) particles due to the broken filters. The downwind decrease in dust concentrations measured by the buoys is very clear, with larger amounts of dust transported at the more eastern station M3, and lower dust concentrations at M4 as a result of deposition between the stations. The accompanying grain-size distributions appear much more similar for the two stations, for some individual samples even coarser-grained for the more downwind buoy at M4 (Fig. 6.6). This could be related to the larger volume of air sampled by the buoy at M4, in turn related to the power at which the engine sucks air through the filter, that could influence the amount of large particles sampled. Atmospheric dust concentrations are corrected for the increased air volume, but the grain-size distributions are not. As the design of the buoys is still improving, further testing of the influence of the air volume sampled by the buoy on the grain-size distribution of the collected dust is necessary. Alternatively, the dust collected by the buoy at M4 could be composed of more minerals with a platy shape, that have a smaller “aerodynamical size” than more spherical particles of the same diameter (Stuut et al., 2005), but are measured by the laser particle sizer as larger particles.

The particle size of the dust collected by the MWAC samplers shows many similarities to the dust collected by the buoys and the subsurface sediment traps. Only the 2014-MWAC sample from buoy M3 is finer-grained than the synchronous sediment-trap samples, and also finer-grained than the MWAC-sample collected more downwind at M4. This could be due to the fact that the sampling periods were not equal for both MWACs (Table 6.1). Especially the MWAC-sampler at M4 (Fig. 6.4B-C) sampled for a longer period (14 months) than the sediment traps (10,5 months) at the same site. Also, the MWAC sampler at buoy M3 collected until end of August and may have missed most of the coarser-grained dust that is washed down by late-summer rains, although it is not clear how much wet deposited dust the samplers receive. Also, the different collection method—atmospheric dust transport collected by the MWAC samplers with a possible small contribution from raindrops, and both dry- and wet-deposited dust collected by the sediment traps—can result in differences in particle-size distributions. In 2015 the MWACs show the expected down-wind fining, with coarser-grained dust sampled at M3 and finer-grained dust at M4 (Fig. 6.6). Because of the similarities between the grain-size distributions of the MWAC samplers and the sediment traps, we can conclude

that the dust sampled directly from the atmosphere is the same as the lithogenic particles captured by sediment traps below, confirming their common aeolian origin.

6.4.2 Dust Deposition (Sediment Traps)

Seasonal differences in deposition flux (dry and wet deposition combined) and particle size in the sediment traps are largest at M1, as most dust and coarsest-grained particles are deposited closest to the source (Van der Does et al., 2016). This results in a good correlation between dust flux and particle size at this upwind site, which implies that the coarse-grained dust determines the dust flux. The downwind decrease in particle size is also reflected by a decreased seasonal amplitude, and therefore a reduced correlation between deposition flux and particle size.

The lower sediment traps at 3,500 m depth show a smaller seasonal trend than the upper traps at 1,200 m depth, for which we propose a number of hypotheses. First, the observation could be due to the larger catchment area of the deeper traps. As downward particle fluxes may be displaced by lateral ocean currents, the deeper traps may sample particles which settled out from different or additional regions than the upper traps (Siegel and Deuser, 1997; Waniek et al., 2000). Not only will this potentially change the particle flux in the lower traps, it may also smoothen the seasonal signal. In addition, the lower ocean-current velocities at the depth of the lower traps could have resulted in a higher collecting efficiency of the traps and thus higher mass fluxes, although current velocities at both depths are well below critical levels (Korte et al., 2017). Second, there is a time lag between particle collection of larger versus smaller sizes by the upper and the lower traps, which is related to the differences in sinking velocity between small and larger particles: Clemens (1998) argued that the reason for a lacking relation between grain size and particle flux in sediment traps is the larger settling velocities of coarse-grained particles compared to finer-grained ones. This could in turn result in an expression of coarser-grained particles during large dust storm events, while the associated high dust flux lags behind this signal as finer-grained particles have not yet settled to the sediment trap depth (Clemens, 1998). This effect would be larger at greater depths. In our sediment-trap samples we observed this time-lag effect as the particle size seems to increase before the deposition fluxes (Fig. 6.3). Third, the deep sediment trap at M5 could have received a seasonal contribution from laterally-transported material from the nearby Barbados margin (Korte et al., 2018a), either by nepheloid layers (Inthorn et al., 2006a; Inthorn et al., 2006b) or lateral ocean currents. This was observed by Honjo et al. (1982), who found increased particle fluxes with depth, and attributed this to possible resuspension of particles or lateral particle transport. The sediment traps used in the present study, however, are still at a generous distance (880–1,320 m) from the seafloor. At M5 the strong negative correlation between the deposition flux and particle size in the lower sediment trap (3,500 m BSL) implies that a larger input of lithogenic material coincided with smaller modal grain sizes. Although the particle size at this station matches the down-wind fining trend, riverine or hemipelagic particles are also characterized by smaller particle sizes

(Stuut et al., 2002), and the strong negative relation matches the scenario of increased input of fine-grained riverine sediment. The sediment traps at stations M1–M4 are located further from continental margins than station M5, and the input of laterally transported riverine sediments is shown to be negligible (Van der Does et al., 2018).

6.5 Conclusions

Here we have shown a unique time series of Saharan dust sampled along a transect in the tropical North Atlantic Ocean. We compared dust during transport over the ocean, collected directly from the atmosphere with dust-collecting buoys, to dust deposition, collected by subsurface sediment traps at two depths in the ocean. Results show a high similarity between the atmospheric dust in transport and the deposited lithogenic particles collected while sinking to the ocean floor, demonstrating their common aeolian origin. However, there are also differences between the transported and deposited dust. The subsurface sediment traps collect both dry- and wet-deposited dust, and deposition fluxes are increased in summer, coinciding with coarser-grained dust deposition, due to large amounts of dust being deposited by summer rains. In contrast, the buoys are restricted to dust in transport (dry), and transport at sea level, and these samples lack a clear seasonal signal. This implies that the large amounts of dust transported at high altitudes in summer, brought down by wet deposition, determines the seasonality of both the deposition flux as well as the particle size of the deposited dust.

In conclusion, our results demonstrate a large difference between atmospheric transport and deposition of aeolian dust over the ocean. This can have important implications for satellite observations of atmospheric dust, as observations of atmospheric dust cannot be extrapolated to oceanic dust deposition fluxes. The processes involved in dust transport and (dry and wet) deposition and their relation have shown to be complex. This study provides important new insights into these processes, that can be used for climate model simulations that incorporate dust transport and deposition, and for the interpretation of long-term paleo dust records of deposited dust.

Acknowledgements

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Chapter 7:

North African Mineral Dust Across the Tropical Atlantic Ocean: Insights from Dust Particle Size, Radiogenic Sr- Nd-Hf Isotopes and Rare Earth Elements (REE)

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Abstract

Large amounts of mineral dust are exported from North Africa across the Atlantic Ocean, impacting the atmosphere and ocean during transport and after deposition through biogeochemical processes. In order to characterize the isotopic signature of dust from different seasons and years, in relation to their bulk particle size, and to obtain a general idea of its provenance, Saharan dust was collected using subsurface sediment traps moored in the tropical North Atlantic Ocean in 2012–2013, and by shipboard aerosol collection during three trans-Atlantic research cruises in 2005, 2012 and 2015. The samples were analysed for radiogenic Strontium (Sr), Neodymium (Nd), and Hafnium (Hf) isotopes, rare earth element (REE; La-Lu) abundances and particle size. In addition, soil sediments from Mauritania, a potential source area, were analysed and compared to the Atlantic dust samples. The results indicate no relation between Sr and Nd isotopic compositions and dust particle size. In contrast, Hf isotopic compositions show a strong relation with particle size, associated to the so-called zircon effect. We explored alternative sources of lithogenic particles to the sediment traps such as Amazon River sediments. Our results reveal that the sediment-trap samples bear distinctly different geochemical signatures from sediments from the Amazon Basin and Amazon River tributaries, and confirm that the primary source of lithogenic particles is northern Africa. The collected dust samples show close relations to African dust aerosols collected at Barbados and samples from the Bodélé Depression, although differences between seasons are observed, which we relate to differences in source areas.

7.1 Introduction

The Sahara Desert in northern Africa is the world's largest dust source (Muhs, 2013), contributing up to 70% of all global annual dust emissions (Maher et al., 2010; Huneus et al., 2011). In particular, the Bodélé Depression, located in the region of the Lake Chad Basin, is currently the largest single dust source, with the biggest dust export in the world (Koren et al., 2006; Washington et al., 2006), and appears to have been active for at least several hundred to thousands of years (Prospero et al., 2002; Armitage et al., 2015). Nevertheless, the contribution from the Bodélé Depression aerosols transported over the Atlantic basin arriving at Barbados in the Caribbean appears to be small (Pourmand et al., 2014; Bozlaker et al., 2018; Kumar et al., 2018). Most dust emitted from North African sources is transported westward across the Atlantic Ocean, with an estimated amount of 182 Tg between 30°N and 10°S every year (Yu et al., 2015a). The emitted dust impacts both the atmosphere and the ocean, from affecting the atmospheric radiation budget (Ryder et al., 2013b) and acting as cloud condensation nuclei and ice nuclei (Wilcox et al., 2010; Atkinson et al., 2013), to the enhancement of the ocean's carbon cycle by delivering nutrients stimulating phytoplankton growth (Martin and Fitzwater, 1988), and by mineral ballasting of organic particles in the ocean (Armstrong et al., 2002; Bressac et al., 2014; Van der Jagt et al., 2018). Saharan dust can also transport viable spores, pathogens and microbes over great distances (Griffin, 2007), which can impact marine and

terrestrial ecosystems (De Deckker et al., 2008), and be harmful to human health and increase mortality rates (Morman and Plumlee, 2014; Schweitzer et al., 2018).

Dust particle size and composition vary seasonally, and with the distance over which the dust is transported. Particle sizes decrease downwind as a result of more rapid gravitational settling of coarse-grained particles (Bagnold, 1941; Van der Does et al., 2016), while the mineralogical and isotopic composition of the deposited dust changes downwind by the preferential settling of heavier quartz and zircon particles closer to Africa (Pourmand et al., 2014; Korte et al., 2017). At Barbados in the Caribbean, daily dust samples have been collected for over 50 years (Prospero et al., 1970; Prospero and Nees, 1977; Prospero and Lamb, 2003), making it one of the longest present-day dust records. Dust concentrations at this island peak during the summer months, related to the latitudinal movement of the Intertropical Convergence Zone (ITCZ). This leads to a northern displacement of the dust cloud over the Atlantic Ocean in summer, followed by a southward migration in winter (Nicholson, 2000). In addition, mineral dust is typically transported at low altitudes (< 3000 m) by the trade winds during the winter season, whereas the Saharan Air Layer (SAL) carries the dust at higher altitudes (>5000 m) across the Atlantic during the summer (Stuut et al., 2005; Friese et al., 2016; Van der Does et al., 2016). The latitudinal and altitudinal seasonal shift of the dust-carrying atmospheric systems, in turn, results in a change of dust transport from different source areas and thus a seasonal change of dust composition (Pourmand et al., 2014; Friese et al., 2017).

Compositional characterization of trace elements and different isotope systems of mineral dust particles can yield information about the source area of the dust (Scheuvens et al., 2013), and selective processes during transport and deposition such as the sorting based on particle size and shape. Dust provenance identification can be aided by determining the mineralogical composition (Caqueneau et al., 1998; Kandler et al., 2009; Kandler et al., 2011; Friese et al., 2017), geochemical composition (Moreno et al., 2006; Muhs et al., 2007; Castillo et al., 2008; Bozlaker et al., 2018), identification of biomarkers (Schreuder et al., 2018b), and radiogenic isotopes (Grousset and Biscaye, 2005; Meyer et al., 2011; Abouchami et al., 2013; Pourmand et al., 2014). The radiogenic Sr, Nd, Hf isotopes and the REE composition of aerosols at emission and deposition sites are shown to be particularly useful proxies for understanding changes in dust provenances and weathering regimes (Scheuvens et al., 2013; Pourmand et al., 2014). Most rare earth elements (REEs) show comparable values relative to the average composition of the upper crust, however the depletion or enrichment of several elements as a result of natural (e.g., chemical and physical weathering processes that lead to enrichment or depletion of heavy minerals such as zircon) or anthropogenic processes can be used as a tracer for source regions (Meyer et al., 2011; Meyer et al., 2013; Scheuvens et al., 2013).

Previously, Pourmand et al. (2014) analysed Sr-Nd-Hf isotopes and REEs of Saharan dust collected in the Caribbean at Barbados between 2003 and 2011, and demonstrated seasonal shifts of dust sources and transport from the Sahara, as well

as a systematic shift in nearly all geochemical proxies as a function of temporally variable dust plumes. Zhao et al. (2018) compared these distal dust samples to surface sediments from the Sahara and Sahel, and also demonstrated the seasonal variation of dust provenance. Here, we present the radiogenic Sr-Nd-Hf isotope and REE composition of Saharan dust collected during three trans-Atlantic research cruises and from submarine sediment traps moored in the tropical North Atlantic Ocean. Their geochemical characteristics will provide a better understanding of dust transported and deposited over the open Atlantic Ocean. Alternative sources of lithogenic particles to the sediment traps from Amazon River sediments are investigated. By analysing the geochemical composition and combining them with particle-size data, we aim to shed light on the provenance of the lithogenic fraction in the sediment-trap samples of anomalously high depositional events. The data are compared to soil samples collected from Mauritania and to data from literature, to gain insights into the general provenance of the dust, whether that is North African or South American. Moreover, this dataset also allows for comparing the composition of dust particles collected from the atmosphere with those collected after sinking through the water column.

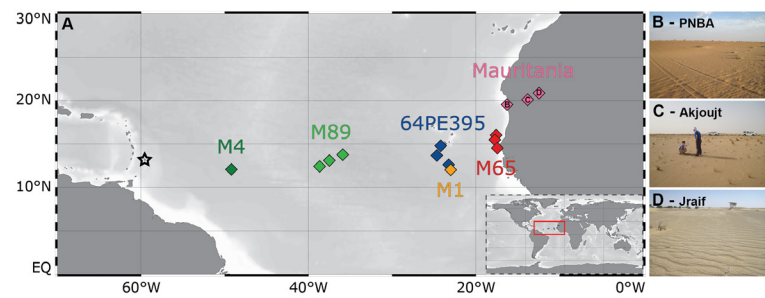


Figure 7.1. A: Location of the samples used in this study in the North Atlantic Ocean: Sediment-traps M1 (12°N, 23°W) and M4 (12°N, 49°W); Soil samples from Mauritania; and average locations of aerosol samples collected during three research cruises: M65, M89 and 64PE395. The Caribbean island Barbados is indicated with a star. Photos of sample locations PNBA (B), Akjoujt (C) and Jraif (D) in Mauritania.

7.2 Material and Methods

7.2.1 Sample Collection

Saharan dust was collected at five mooring stations along a trans-Atlantic transect, as described by Van der Does et al. (2016) and Korte et al. (2017). The transect consisted of four stations along 12°N, and a fifth station at 13°N, with two sediment traps moored at each station at 1,200 m and 3,500 m below sea level (BSL). The sediment traps consisted of 24 sampling bottles which were pre-programmed to sample at synchronous intervals of 16 days, from 19 October 2012 to 7 November 2013 (Stuut et al., 2012; Stuut et al., 2013). For this study, five samples from three different time intervals were analysed from two sediment traps, moored at 1,200m

below sea level at stations M1 (23°W) and M4 (49°W) (Fig 7.1, Table 7.1). These are synchronous sample intervals #12 (13-04-2013 – 29-04-2013) and #24 (22-10-2013 – 07-11-2013) from both stations, which showed anomalously high deposition fluxes at M4 (Van der Does et al., 2016), and #9 (24-02-2013 – 12-03-2013) from M1, which is the sample with the largest dust flux from that station.

Table 7.1. Sample intervals, positions of sampling, and modal particle size of sediment-trap samples, shipboard-collected aerosol samples and Mauritanian soil sediments.

Sample ID	Sample type	Sampling interval				Location		Modal grain size (µm)
		From		To		Lat (°N)	Lon (°W)	
		Date	Time (UTC)	Date	Time (UTC)			
M1-U9	Sediment trap	24-2-2013		12-3-2013		12.00	23.00	12.40
M1-U12		13-4-2013		29-4-2013				13.61
M1-U24		22-10-2013		7-11-2013				18.00
M4-U12	Sediment trap	13-4-2013		29-4-2013		12.06	49.19	7.08
M4-U24		22-10-2013		7-11-2013				10.29
M65-D3	Aerosol	13-6-2005	07:30	14-6-2005	13:30	15.95	17.54°	26.14
M65-D4		14-6-2005	15:30	16-6-2005	21:45	15.44	17.71°	28.70
M65-D5		16-6-2005	22:00	18-6-2005	13:00	14.52	17.45°	28.70
M89-F09	Aerosol	10-10-2012	09:09	11-10-2012	14:53	13.71	35.82°	37.97
M89-F10		11-10-2012	14:53	12-10-2012	10:00	13.09	37.45°	37.97
M89-F11		12-10-2012	10:00	12-10-2012	20:30	12.41	38.63°	41.68
64PE395-F04	Aerosol	11-1-2015	21:30	12-1-2015	09:00	14.79	24.17°	4.05
64PE395-F06		12-1-2015	11:25	12-1-2015	17:20	13.68	23.67°	4.05
64PE395-F08		12-1-2015	22:43	13-1-2015	09:46	12.57	23.23°	4.05
PNBA	Soil sediment	18-11-2009				19.47	16.27	
Akjoujt		19-11-2009				20.07	13.79	
Jraif		20-11-2009				20.86	12.43	

* Average location of sampling at sea during transits

** Sampled during stationary interval

Aerosol samples were collected during three different research cruises in the tropical North Atlantic Ocean: FS Meteor Expedition M65 in June 2005 (Mulitza, 2005), FS Meteor Expedition M89 in October 2012 (Stuut et al., 2012), and RV Pelagia Expedition 64PE395 in January 2015 (Stuut et al., 2015)(Fig 7.1, Table 7.1). Samples from these three cruises were picked to represent three different seasons (summer, autumn and winter) of three different years, to shed light on possible differences in provenance of the dust in various years, and to be

compared to the dust found in the sediment traps. During all cruises, aerosol sampling was performed with an Anderson high-volume dust collector, mounted on the deck above the bridge of the ship. Dust was collected on letter-sized Whatman Type 41 filters under a protective cover. The samplers were equipped with a wind vane which switched off the sampler during winds blowing at an angle $>90^\circ$ from the ship's heading. This was done to prevent contamination from the ship's exhaust behind the sampler (see e.g. Stuut et al., 2012).

Soil samples were collected at various sites in Mauritania, one potential source and transit area of mineral dust, during a field campaign in 2009. These are named PNBA, Akjoujt and Jraif (Fig. 7.1), representing potential dust sources. The PNBA sample was taken from large (10–20 m) yellow dunes in Parc National de Banc D'Arguin (PNBA; Fig. 7.1B). The Akjoujt sample was taken from a relatively flat landscape with small (10–40 cm) vegetated dunes, about 70 km northeast of Akjoujt (Fig. 7.1C). The Jraif sample was taken from a dried-up lake bed in an area with mostly dunes close to Jraif (Fig. 7.1D). Exact sampling locations are given in Table 7.1.

7.2.2 Particle-Size Analysis

The results of dust particle-size analysis of the samples from the sediment traps were presented by Van der Does et al. (2016) and cited here for comparison. The dust was isolated from the filters of the shipboard-collected aerosol samples collected during cruises M65 and M89 by rinsing with Milli-Q water to remove the dust. The three filters collected during cruise 64PE395 were ashed in a low-temperature asher ($\sim 100^\circ\text{C}$) to remove the dust from the filters. Immediately prior to grain-size analysis, a few drops of 0.1 M sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$) was added to the sample to ensure complete disaggregation of the particles. Particle-size distributions were obtained with a Coulter laser diffraction particle size analyser (LS13 320), equipped with a micro liquid module (MLM) as described by Van der Does et al. (2016). The samples were homogenized during analysis by a magnetic stirrer, and the influence of air bubbles was minimized using degassed water. This resulted in particle sizes ranging from 0.375 to 2000 μm , distributed over 92 size classes.

7.2.3 Radiogenic Sr-Nd-Hf Systematics and REE Composition

Prior to radiogenic analysis, the samples were ashed in a quartz crucible at 750°C to remove all the organics, and were subsequently homogenized by grinding in an opal mortar and pestle. The two largest sediment-trap samples (M4-U12 and M4-U24) were split in two, of which one half the bulk sample and the other half the lithogenic fraction was analysed. To isolate the lithogenic fraction ($<10\%$ of bulk sample), the marine sediment-trap samples were treated to remove all biogenic constituents. This was performed by a sequential leaching procedure in several steps, based on the protocol described by Handley et al. (2013). First, the sample was rinsed with Milli-Q water and centrifuged. Next, 1M sodium acetate (NaAc) in 25% acetic acid (HAc) were added to the sample to remove carbonates. Amorphous and crystalline Fe- and Mn-oxides, which can adsorb dissolved

REEs from the ocean (Tachikawa et al., 1997), were removed by adding 0.04M hydrolaxamine hydrochloride ($\text{NH}_2\text{OAH} \cdot \text{HCl}$) and nitric acid (HNO_3). Finally, biogenic silica was removed by adding 2M sodium carbonate (Na_2CO_3), and samples were placed in an oven at 85°C for 5 hours. Every leaching step was followed by several rinsing and centrifuging steps. The samples were subsequently dried in an oven at 80°C . No specific size fractions were isolated before geochemical analysis.

All samples were subsequently prepared following the method described in detail in Pourmand and Dauphas (2010), Pourmand et al. (2012) and Pourmand et al. (2014). Briefly, the samples were fused with LiBO_2 alkali flux at 1070°C and added to 6M HNO_3 to ensure complete dissolution of refractory minerals. Radiogenic Sr, Nd, Hf isotopes and REEs were separated by a three-stage extraction chromatography scheme devised for high-precision isotope and elemental analysis on a Thermofisher Scientific Neptune Plus multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS) at the Neptune Isotope Lab, University of Miami. To check for possible contamination, procedural blanks were processed and analysed. Unless otherwise stated, the uncertainties on isotope ratios reported herein are at the 95% confidence interval. Variations in $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ are reported as ϵNd and ϵHf , expressed as deviations from the chondritic uniform reservoir (CHUR) values of 0.512630 ± 0.000011 and 0.282785 ± 0.000011 , respectively (Bouvier et al., 2008). The abundances of REEs were normalized to the mean of Post-Archean Australian Shale (PAAS), as described by Pourmand et al. (2012). The ratio of Lanthanum over Lutetium (La/Lu) is given, to represent the extent of light REE (LREE) over heavy REE (HREE) fractionation, which is related to the source rock composition as a function of its geochemical evolution through time. LREE are less compatible than HREE during partial melt, leaving the melt enriched in LREE and the residue in HREE, which can be used to discern source rocks of mantle and crustal origin. Sm-Nd model ages are also used for this purpose, and represent the crustal residence age of the source rocks. The rock ages are the result of the weakly radioactive ^{147}Sm , which decays into ^{143}Nd , and the crustal ratios of $^{143}\text{Nd}/^{144}\text{Nd}$ (Elderfield and Greaves, 1982; Elderfield, 1988). The $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of continental crust is generally lower than oceanic crust (Elderfield, 1988), and the difference becomes larger for rocks with younger ages. The Sm-Nd model ages are based on nominal value of 0.51316 for $^{143}\text{Nd}/^{144}\text{Nd}$ of depleted mantle and modern $^{147}\text{Sm}/^{144}\text{Nd}$ of 0.2137 (Goldstein et al., 1984; Grousset et al., 1988). The anomalies for Cerium and Europium are calculated according to the following relationships, normalized to the mean values of La-Lu for Post Archean Australian Shale (PAAS; Dauphas and Pourmand, 2015):

$$\text{Ce} / \text{Ce}^* = \text{Ce}_N / (\text{La}_N^{0.48} \times \text{Pr}_N^{0.52}) \quad (1)$$

$$\text{Eu} / \text{Eu}^* = \text{Eu}_N / (\text{Sm}_N^{0.45} \times \text{Gd}_N^{0.55}) \quad (2)$$

Positive anomalies (>1) mean Cerium and Europium are enriched, while negative anomalies ($0-1$) indicate they are depleted relative to their trivalent REE neighbours. Ce and Eu anomalies of 1 means that all three REEs have a crust-like composition (Sholkovitz, 1993). As Ce and Eu can carry different charges than their normal trivalent charge (+4 and +2, respectively), this can lead to distribution anomalies relative to the trivalent REEs (Elderfield and Greaves, 1982; De Baar et al., 1985; Elderfield, 1988). Ce can be preferentially incorporated in zircon minerals (Thomas et al., 2003) and Eu in plagioclase minerals (Weill and Drake, 1973; Elderfield, 1988), leading to enrichments of these elements if these minerals are more abundant. Ce anomalies can also be affected by oceanic redox conditions, and Eu anomalies by hydrothermal input (Elderfield, 1988), in the case of the sediment-trap samples.

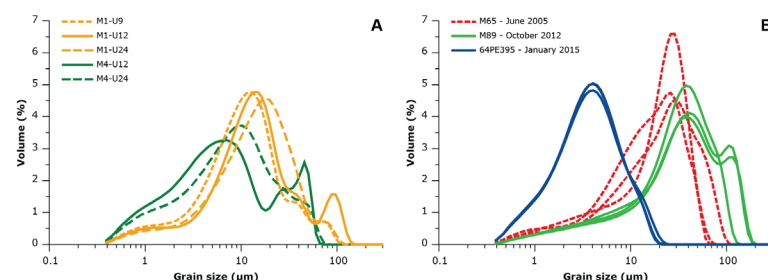


Figure 7.2. Grain-size distributions of the A: Sediment-trap samples (Van der Does et al., 2016), and B: Shipboard-collected aerosol samples.

7.3 Results

7.3.1 Dust Particle Size

The sample information, sampling day and time as well as samples' modal grain size (μm) data are presented in Table 7.1. Particle-size distributions of the sediment-trap samples show that in general, particles deposited at M1 close to the east coast of North Africa are coarser-grained compared to samples from M4 at the more distal location (Fig. 7.2A; Table 7.1). This finding is intuitively logical as coarser-grained wind-blown particles are deposited closer to the source (Van der Does et al., 2016). In addition, Van der Does et al. (2016) showed that seasonal changes in dust export exert control over the particle-size distribution of the dust, with coarser-grained particles in summer compared to winter. The sediment-trap samples collected in autumn (sample interval #24) are coarser-grained than the samples collected in spring (#9 and #12). In contrast, however, the shipboard-collected aerosol samples do not show the typical downwind decrease in particle size as expected. Samples from M65 and M89 are similar in particle size, but very different in sampling location. The samples from M89 were not expected to be coarsest as they were collected farthest from their north-African source (Fig. 7.1). Samples collected during 64PE395 have the smallest dust particles, and show the highest symmetry. The relative enrichment of coarse particles to the distributions of M89 and M65 could be due to the strength of the winds and dust events. Indeed,

these samples were collected during summer (M65) and autumn (M89), when wind strengths are higher, and the dust is transported at greater altitudes within the SAL (e.g. Chouza et al., 2016). For the samples collected during 64PE395, the ratio median/modal grain size—reflecting the difference between the most occurring value (mode) and the middle value (median)—is highest (~ 0.90), and thus the most similar, followed by the samples collected during M89 (~ 0.85) and M65 (~ 0.69). Both the samples collected in spring (#12) by the sediment traps at M1 and M4 (solid lines) and the shipboard-collected aerosols from M89 show a “shoulder” on the coarse end of the grain-size distribution (Fig. 7.2). These coarse-grained particles may mostly include mica particles, which have a different aerodynamical behaviour due to their platy shape than more spherical quartz particles of similar diameter (Stuut et al., 2005; Van der Does et al., 2016), although the latter are also clearly present in the samples (Van der Does et al., 2016).

7.3.2 Radiogenic Isotopes

Isotopic values for radiogenic Strontium, Neodymium and Hafnium are reported in Supplementary Supplementary Table 7.2. $^{87}\text{Sr}/^{86}\text{Sr}$ values for the sediment-trap samples range between 0.718942 ± 0.000011 (M4-24) and 0.723324 ± 0.000013 (M1-24; Fig. 7.3A). The shipboard-collected aerosol samples have lower $^{87}\text{Sr}/^{86}\text{Sr}$ values, averaging 0.713449 ± 0.0032 , with the samples from M65 being the least radiogenic, and collected closest to the source. The Mauritanian sediments have Sr isotopic values similar to the sediment-trap samples except for the sample collected near Akjoujt, which is considerably more radiogenic than any of the samples or the source regions (Fig. 7.3B; $^{87}\text{Sr}/^{86}\text{Sr} = 0.745105 \pm 0.000011$).

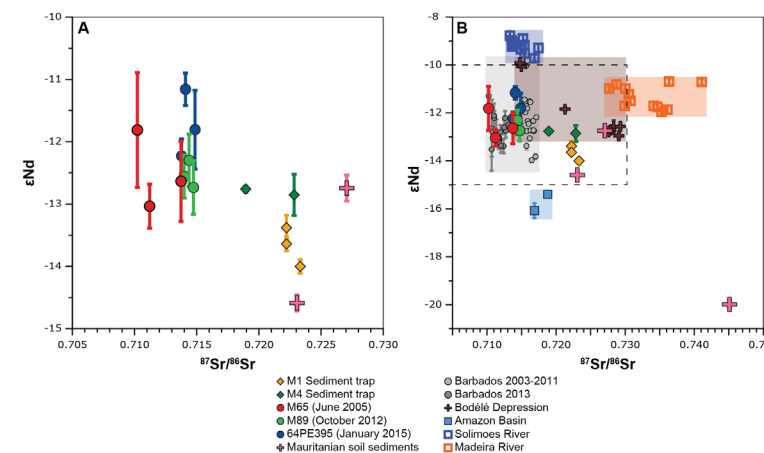


Figure 7.3. Radiogenic Sr and Nd isotopes. A: Samples from sediment traps, shipboard-collected aerosols and Mauritanian sediments. B: The data are compared to published data: Aerosols collected on Barbados (Pourmand et al., 2014), sediments from the Amazon Basin, the Bodélé Depression in Chad (Abouchami et al., 2013), and suspended sediment load from the Solimoes and Madeira Rivers (Viers et al., 2008). Error bars define 95% confidence intervals, which for Sr is smaller than the size of the symbols.

The ϵ_{Nd} values of the sediment-trap samples and the shipboard-collected aerosol samples cluster between -11.2 ± 0.26 (64PE395-F04) and -14.0 ± 0.11 (M1-U24), with mostly overlapping uncertainties (Fig. 7.3A). The sediment-trap samples collected at M1 have slightly lower ϵ_{Nd} values (average -13.7 ± 0.62) than samples collected at M4 (average -12.8 ± 0.13). The Mauritanian sample Akjoujt also has a considerably lower ϵ_{Nd} value (-20.0 ± 0.17) and falls well outside any source-region ranges previously reported (Fig. 7.3B).

The Hf isotopic composition of the samples shows higher ϵ_{Hf} values for finer-grained dust samples, and decreasing values for coarser-grained samples (Fig. 7.4A; Supplementary Table 7.2). Sediment-trap samples from the most proximal station M1 have less radiogenic ϵ_{Hf} values ranging from -17.2 ± 0.41 (M1-U24) to -15.3 ± 0.3 (M1-U9), with the coarsest sample having the lowest ϵ_{Hf} value (M1-24) and the finest sample the highest (M1-09). These samples plot along the igneous rock array (Fig. 7.4A). Samples collected at M4 have relatively more radiogenic ϵ_{Hf} values, with the coarsest sample (M4-24) having the lowest ϵ_{Hf} value (-12.8 ± 0.98) and the finest sample (M4-12) the highest (-9.3 ± 0.68). These data plot between the igneous rock array and the zircon-depleted sediment array (Fig. 7.4A). Shipboard-collected aerosols of M65 have the least radiogenic ϵ_{Hf} values and plot closest to the zircon-bearing sediment array. These samples are among the coarsest aerosol samples and are collected closest to the source of all the samples. Sample M65-D3 is an exception, with a highly radiogenic ϵ_{Hf} value of -1.7 ± 5.00 , albeit with a large uncertainty. Aerosol samples of M89 and 64PE395 plot close to the zircon-depleted sediment array, in the same range as the M4 sediment-trap samples. The Mauritanian sediments have values similar to the most proximal sediment-trap samples at M1, and the shipboard-collected aerosol samples of M65 (Fig. 7.4A).

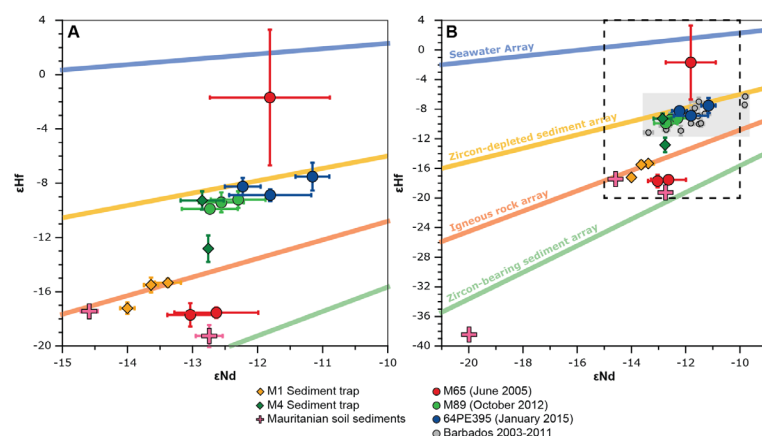


Figure 7.4. Radiogenic Hf and Nd isotopes. A: Samples from sediment traps, shipboard-collected aerosols and Mauritanian sediments. B: The data are compared to aerosols collected on Barbados (Pourmand et al., 2014). Seawater, sediment and rock arrays are defined by (Bayon et al., 2009). Error bars define 95% confidence intervals.

7.3.3 Rare Earth Elements

The REE patterns for the sediment-trap samples show an enrichment for Eu (Fig. 7.5A), with Eu anomalies ranging between 1.12 and 1.16 (Fig. 7.6A; Supplementary Table 7.3). These samples are also slightly enriched in heavy REEs (HREE, Gd-Lu), with ΣHREE values ranging between 6.4 and 7.9. The shipboard-collected aerosol samples have similar REE patterns as the sediment-trap samples, but lack the HREE enrichment (Fig. 7.5B). These samples also show larger Ce anomalies, averaging 1.06, 1.04 and 1.10 for M65, M89 and 64PE395 samples, respectively (Fig. 7.6A). Eu-anomalies are very similar for both the sediment-trap samples (average 1.14) and the shipboard-collected aerosol samples (average 1.17), but Eu anomalies for two of the Mauritanian sediments are elevated; 1.18 and 1.56. Contrastingly, the REE patterns of the Mauritanian samples look very similar to the sediment-trap samples (Fig. 7.5A), with ΣHREE values ranging between 1.79 and 2.82, and Eu anomalies ranging between 1.39 and 1.56. (Fig. 7.6B).

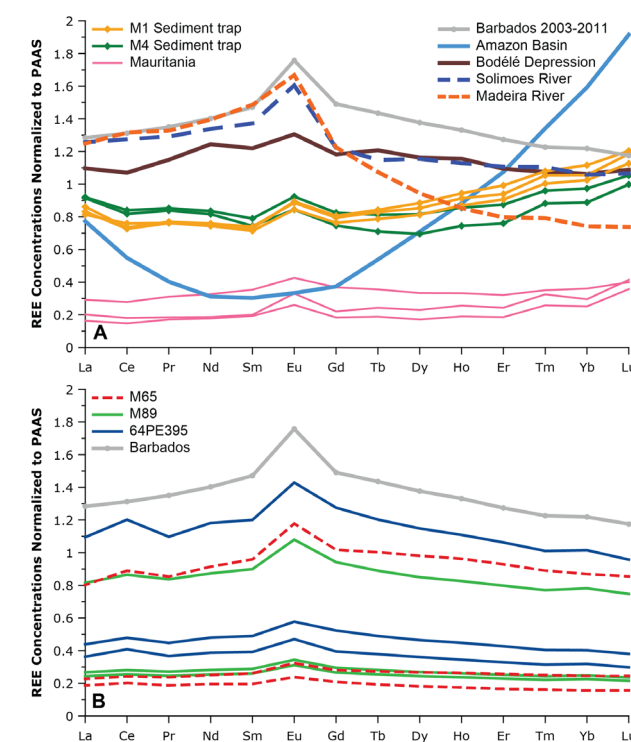


Figure 7.5. PAAS-normalized REE concentrations of samples from sediment traps and Mauritanian sediments (A) and shipboard-collected aerosols (B). The data are compared to published data: Aerosols collected on Barbados (Pourmand et al., 2014), sediments from the Amazon Basin, the Bodélé Depression in Chad (Abouchami et al., 2013), and suspended sediment load from the Solimoes and Madeira Rivers (Viers et al., 2008), also normalized to PAAS.

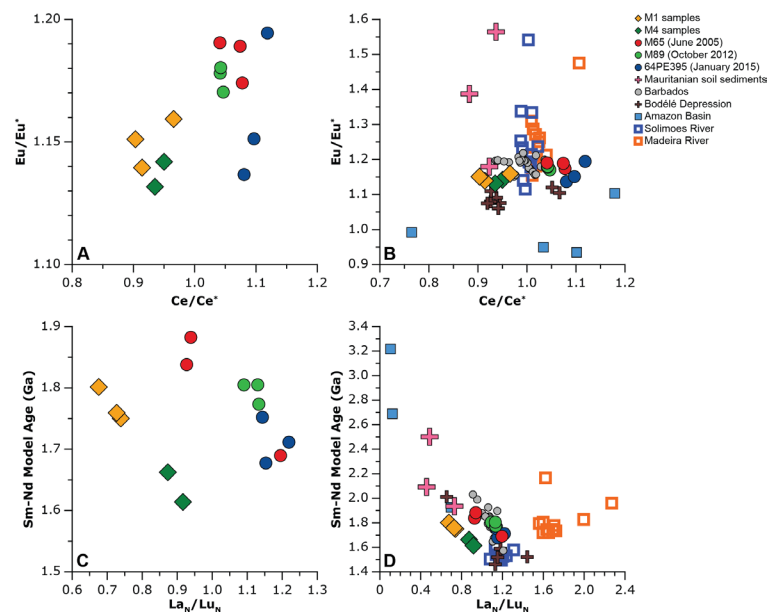


Figure 7.6. A: Eu- and Ce-anomalies of samples from sediment traps and shipboard collected aerosols. B: The data including Mauritanian sediments, and compared to published data: Aerosols collected on Barbados (Pourmand et al., 2014), sediments from the Amazon Basin, the Bodélé Depression in Chad (Abouchami et al., 2013), and suspended sediment load from the Solimoes and Madeira Rivers (Viers et al., 2008). C and D: Sm-Nd model age and La_N/Lu_N of the same samples.

Sm-Nd model ages are highest for the Mauritanian sediments (1.9–2.5 Ga), and similar for the sediment-trap samples (1.6–1.8 Ga) and shipboard-collected aerosol samples (1.7–1.9 Ga) (Fig. 7.6D; Supplementary Table 7.3), indicating that the age of the source rock is the same for the latter two. However, these are model ages and their relative values are more meaningful than the absolute ages. Their distinction is clearer in combination with La_N/Lu_N values, which is higher for the shipboard-collected aerosol samples (up to 1.22) than for the sediment-trap samples (0.68–0.92; Fig. 7.6C). The sediment-traps show similar values, close to the aerosol samples. The sediments collected at Jraif from Mauritania most closely resembles the dust samples.

7.4 Discussion

7.4.1 Relation Between Strontium and Particle Size

Several studies have reported that Sr isotopes in particular may be prone to grain-size dependence in their isotope composition (Dasch, 1969; Meyer et al., 2011). This is in turn related to the mineralogy, given that elemental Sr is preferentially enriched in minerals of low density and high buoyancy. Specifically, fine fractions are expected to have more radiogenic Sr isotopic values as these are usually composed of particles that are more prone to weathering such as clay minerals

and micas (Feng et al., 2009), which have higher Rb/Sr ratios, resulting in more radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ compositions for fine-grained particles (Feng et al., 2009; Meyer et al., 2011; Aarons et al., 2013). Contrary to this general expectation, however, the dust samples that were measured in this study do not exhibit a notable grain-size dependence between samples with different modal particle sizes with respect to Sr isotopic values (Fig. 7.7A). If particle size was the only variable affecting the Sr isotopic composition of the samples, one would expect more radiogenic values at M4 with finer-grained particles (7.1–10.3 μm). However this is not the case for the dust of different sizes at the different stations, nor between the individual samples at M1 (12.4–18.0 μm). Only at M4 does the finer-grained dust sample show more radiogenic Sr values than the coarser-grained sample. In the case of the samples studied here, the Sr isotopic values are therefore likely the result of a provenance signal that appears to dominate over the influence of the grain-size effect. As the Sr isotopic values of the sediment-trap samples are mostly >0.720 , a source in the west of northern Africa can be expected (Scheuven et al., 2013).

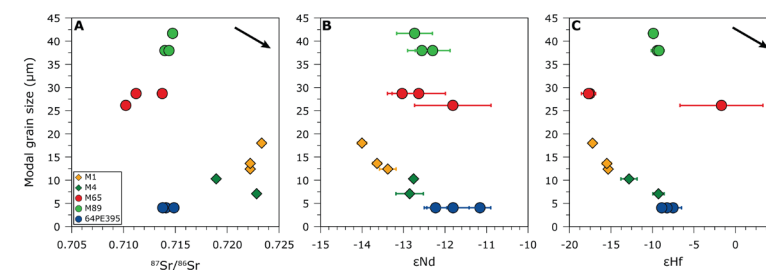


Figure 7.7. Relation between Sr-Nd-Hf isotopes and dust modal grain size. Arrow indicates the expected relation between size and isotopic composition.

For the shipboard-collected aerosols, the lack of relation between particle size and distance to the source may be related to the influence of seasonality, inter-annual variability and specific dust events, which can be related to differences in wind speed and changes in the provenance of the dust particles. Indeed, samples collected during 64PE395 have the smallest dust particles, related to finer-grained dust transport during winter, while coarser-grained dust is observed in summer and fall, in agreement with previous findings from sediment-trap samples along the sampled transect (Van der Does et al., 2016). The shipboard-collected aerosol samples do not show significant variation in Sr isotopic values, despite the particle size being very different (Fig. 7.7A), also among the individual samples of the different sampling campaigns. This is contrary to previous findings of a strong relation between particle size and Sr isotopic composition, although this was established for size-separated fractions of dust (Dasch, 1969; Meyer et al., 2011). Two of the shipboard-collected aerosol samples of M65, closest to the African coast, are exceptions and have the lowest Sr isotopic values (Fig. 7.3A). Although these samples are not the coarsest, they may have relatively more quartz particles than clay particles due to their proximal location to the source, influencing

radiogenic Sr values. Alternatively, a relatively larger contribution of less radiogenic carbonates could have influenced the radiogenic Sr values, although we would expect the three samples of M65 to be similar in composition, as these were collected very close to each other, both geographically as well as temporally. The “shoulder” at the coarse end of the grain-size distributions, possibly related to a larger amount of platy mica particles, can also influence the isotopic signature, although this is not clear for the samples presented here. In addition, an aspect of the transport distance that may be overlooked is that the samples closer to the west-African coast are seemingly closer to their possible source areas, but the dust found in these samples could originate from sources much further inland.

7.4.2 Relation Between Neodymium and Particle Size

In contrast to Sr isotopes, Nd isotopes are not expected relate to the dust's particle size (Feng et al., 2009; Meyer et al., 2011), related to the different geochemical behaviour of elemental ^{87}Sr and its parent isotope ^{87}Rb , and a more similar behaviour between ^{143}Nd and its parent isotope ^{147}Sm , respectively. Sr is more easily lost from minerals during weathering processes than Rb, and Sm and Nd show similar responses to weathering (Feng et al., 2009). Indeed, no relation between the Nd isotopic values and the dust's modal grain size is observed (Fig. 7.7B). The low Nd isotopic values of the shipboard-collected aerosols and sediment-trap samples also point to a source area of the dust in a more western region of northern Africa (Scheuven et al., 2013). The Mauritanian sediments show similarities to the sediment-trap samples, except for the sample from Akjoujt, which is different in all isotope systems and which may therefore not be representative of the average composition of the source soil. Nevertheless, it must be noted that highly radiogenic Sr values in combination with less radiogenic Nd isotopic values have been reported for Mauritania (Scheuven et al., 2013). However, the interpretation of these soil samples as long-range mineral dust should be done with caution, considering potential aerosol samples might be substantially different than the soil sediments (Pourmand et al., 2014). We also note that these particles can inherently have very different particle-size distributions, and mineral and chemical composition as a result from sorting during dust generation, transport and deposition. This sorting occurs as the dust is transported in different ways during different seasons, at different altitudes, which results in the falling out of different minerals affecting the total composition of the deposited dust and the remaining airborne dust (Scheuven et al., 2013). These findings place more emphasis on the need to characterize erodible soil with high potential for generating airborne particles in potential source regions to allow for a consistent comparison of dust particles between source and sink, based on their geochemistry, as Zhao et al. (2018) did for four potential source regions in northern Africa.

7.4.3 Relation Between Hafnium and Particle Size

The grain-size dependence of Hf isotopes is the result of gravitational settling of zircons during transport, and is known as the “zircon effect” (Patchett et al., 1984;

Aarons et al., 2013). Zircon minerals typically have low $^{176}\text{Hf}/^{177}\text{Hf}$ isotopic ratios due to their extremely low Lu/Hf ratios, which results from Lu exclusion and Hf incorporation in the crystal structure of zircons (Hoskin and Schaltegger, 2003). Heavier zircon particles are deposited close to the source during atmospheric transport, changing the Hf isotopic composition of dust aerosols with increasing transport distance towards more radiogenic Hf isotopic values (Patchett et al., 1984; Aarons et al., 2013). The more radiogenic Hf isotopic values for fine-grained dust samples and lower $^{176}\text{Hf}/^{177}\text{Hf}$ for coarser-grained samples is observed for all the sediment-trap samples (Fig. 7.7C), in combination with the fine-grained Barbados samples that have high ϵHf values. These samples all plot close to the zircon-depleted sediment array (Fig. 7.4B). Here it seems that the zircon effect for these samples is superimposed on the isotopic signature resulting from different dust provenances. The low ϵHf values for the samples collected during M65 can be similarly explained as a relative enrichment of zircons, as these are generally deposited and collected close to the source. The more distal samples collected during the M89 and 64PE395 sampling campaigns have more radiogenic Hf isotopic values close to the zircon-free sediment array, similar to the Barbados and M4 samples.

7.4.4 Comparison to Dust Collected at Barbados

To examine the influence of long-range transport, the presented data are compared to atmospheric dust collected at Barbados between 2003 and 2011, in 2007 and in 2013, with Sr isotopic values averaging 0.715804 ± 0.0026 (Pourmand et al., 2014), 0.720120 ± 0.0034 (Kumar et al., 2018) and 0.711754 ± 0.0015 (Bozlaker et al., 2018), respectively. These are similar to the shipboard-collected aerosols, but less radiogenic than the deposited dust as found in the sediment-trap samples (Fig. 7.3B). The dust mean particle size for mineral dust collected at Barbados is much finer-grained (smaller than $5 \mu\text{m}$; Li-Jones and Prospero, 1998) than the samples analysed for this study. As a result, one would expect the highest $^{87}\text{Sr}/^{86}\text{Sr}$ values, in line with the expected relation between particle size and Sr isotopic composition. Nevertheless, these values appear to be among the least radiogenic, and the Sr isotopic composition are more likely the result of differences in dust provenance (that is related to the age of the source rocks and the mineralogical composition). Dust collected at Barbados in 2013 by Bozlaker et al. (2018) is expected to be similar to the dust collected in the same period by the sediment traps. However, these dust samples collected at Barbados have systematically lower Sr isotopic values, and therefore are not necessarily related to dust provenance only, and thus possibly related to transport distance, which affects dust particle size and mineralogy. The reason why this is not clearly present for the shipboard-collected aerosol samples is due to the different years and season of dust collection, and therefore overprinted by the differences in dust provenance. The lack of a clear relationship between Sr isotopic composition and particle size of the shipboard-collected aerosol samples could also result from a strong Sr signal from one particular size class within the sample, thereby dominating the Sr signal from over the other size classes. This also illustrates the significance of source-related signals in contrast to size-controlled signatures of dust particles.

Seasonal changes in isotopic signature related to differences in provenance were also found by Zhao et al. (2018), who compared the isotopic signature of clay-sized sediments from source areas to long-range transported dust to Barbados during 2013 and 2014 dust seasons, and by Kumar et al. (2018), who compared dust collected at the proximal Cape Verde Islands to dust collected at Barbados. The latter group found more variable Sr and Nd isotopic compositions during winter at the proximal Cape Verde Islands, reflecting changing dust provenance (Kumar et al., 2018). They also found isotopic compositions to be more stable during the summer, similar to the dust collected at distal Barbados, reflecting a more stable dust source or consistent mixing of different sources during this time. Therefore, the measured Sr isotopic signal in the shipboard-collected aerosol samples is likely the result of the provenance of the dust, which can vary between the seasons, and also annually.

7.4.5 Examining Possible Contributions from Amazon River Sediments

The westernmost mooring M4 (49°W) is located in the vicinity of Amazon-River outflow, and therefore a possible sediment contribution may come from Amazon riverine lithogenic particles. Saharan dust is virtually the only source of lithogenic sediments to the deep sea in the eastern Atlantic Ocean, but west of the mid Atlantic ridge there is a large influence of riverine sediments from the Amazon and Orinoco rivers and nepheloid layers (Biscaye and Eittrheim, 1977; Grousset and Biscaye, 2005). In two of the twenty-four sampling intervals, #12 (13-04-2013 – 29-04-2013) and #24 (22-10-2013 – 07-11-2013), very high deposition fluxes were observed (Van der Does et al., 2016; Korte et al., 2017), leading to question the origin of the lithogenic particles as Saharan dust only. Nutrients carried with the Amazon River outflow reached the sediment-trap sampling site and influenced phytoplankton productivity, resulting in increased coccolithophore fluxes (Guerreiro et al., 2017). The question remains if part of the lithogenic fraction found in the sediment-trap samples may originate from Amazon River sediments. Geochemical analysis of these samples and synchronous samples at the easternmost mooring M1 (23°W) could shed light on the provenance of the lithogenic fraction found in the sediment traps. To test the hypothesis of a possible influence of Amazon sediments to the sediment trap at M4, the data are compared to sediments collected from the Amazon Basin and the Bodélé Depression (Abouchami et al., 2013), and to suspended sediment load from two tributaries of the Amazon River; the Madeira and Solimoes Rivers (Viers et al., 2008).

The combination of Sr and Nd isotopes can give a better insight into the potential source areas of the deposited dust (Grousset and Biscaye, 2005; Scheuven et al., 2013; Pourmand et al., 2014; Sharifi et al., 2018; Zhao et al., 2018). Sediments collected in the Amazon Basin (Abouchami et al., 2013) and from the Solimoes and Madeira Rivers (Viers et al., 2008) show different $\epsilon_{\text{Nd}} - ^{87}\text{Sr}/^{86}\text{Sr}$ values than the Saharan dust samples (Fig. 7.3B) as collected by the sediment traps, via shipboard aerosol collection and the atmospheric dust sampled at Barbados (Pourmand et al., 2014; Bozlaker et al., 2018). The Solimoes River sediments have lower Sr isotopic values than the sediment-trap samples but similar values to the

shipboard-collected aerosol samples. However, the Solimoes River sediments yield more radiogenic Nd isotopic values. The Madeira River sediments show more similar Nd isotopic values but more radiogenic Sr isotopic values compared to the various dust samples. The combination of both radiogenic isotopes clearly illustrates the difference between the Amazon-derived sediments and Saharan-dust samples. In contrast to the geochemical signatures found in South-American sediments, the samples collected from the Bodélé Depression (Abouchami et al., 2013) are more similar to the Saharan dust samples, although over a wide range for both Sr and Nd composition, and strengthens the hypothesis of a north African provenance dominating the Atlantic dust particles.

The REE concentration patterns can also be used to distinguish between different sources of lithogenic material. The REE patterns of the dust samples collected at Barbados (Pourmand et al., 2014) are nearly identical to the shipboard-collected aerosol samples (Fig. 7.5B). The sediment-trap samples also show similarities with the Barbados samples, and have an apparent enrichment of HREE (Fig. 7.5A). Between 1 and 3% of the REEs can be dissolved into seawater, with especially the middle REEs (Eu–Er) showing greater solubility than LREE and HREE (Greaves et al., 1994). This small amount of solubility may have slightly affected the middle REEs of sample M4-12 (Fig. 7.5A). The samples are also closely related to the samples collected from the Bodélé area (Abouchami et al., 2013), with a similar Eu-enrichment. The sediment-trap samples are also very similar to the aerosol samples collected over the Atlantic, as well as to the aerosol samples from Barbados, demonstrating that the dust fraction was effectively isolated from the marine samples. The similarity of REE patterns from sediment-trap samples from M1 and M4 already indicates a minimal influence of Amazon-derived sediments to the M4 samples, as these will never reach as far east as M1 without being sedimented to the ocean floor. At the same time, the REE profiles from the sediment traps differ greatly from those originating from the Amazon Basin, demonstrating the different origin of the sediments in the sediment-trap samples.

The difference with the sediments collected from the Amazon Basin once again highlights the unlikely contribution from Amazon River sediments to the sediment-trap samples. The Amazon Basin sediments show depleted Middle REEs and enriched HREEs, making the samples distinctly different from the Saharan dust samples. However, the sediments collected from the two tributaries of the Amazon River, the Solimoes and Madeira River (Viers et al., 2008), show more similar REE concentration patterns to the Saharan dust samples, with an enrichment in Eu and no enrichment in HREEs, and even a HREE depletion for the Madeira River samples (Fig. 7.5A). The REE signature from the Amazon sediments is the result of LREE depletion due to salt-induced coagulation or river borne colloidal REEs in low salinity regimes, making them more fractionated (Sholkovitz, 1993). The Madeira and Solimoes rivers show less fractionation since these are more representative of upstream waters that did not experience mixing with the oceanic end member. Riverine sediments that

would have travelled towards the sediment traps would be expected to show this fractionated REE signature, which is not present in the sediment-trap samples. Instead, the sediment traps show a small enrichment in HREEs compared to the aerosol samples, which is likely due to the influence of seawater REE signature; enriched in HREEs and depleted in LREEs (Elderfield, 1988; Sholkovitz, 1993).

7.4.6 REE Anomalies

Ce anomalies of the aerosol samples are all positive (>1), meaning similar to the continental crust (Sholkovitz, 1993). Positive Ce anomalies can be indicative of more zircon particles in the sample. Therefore, a positive Ce anomaly would be expected for Saharan dust samples, in contrast to the dissolved fraction that shows a negative Ce anomaly (Sholkovitz, 1993). The sediment-trap samples indeed all show negative Ce anomalies. However, while others have reported both positive and negative Eu anomalies for refractory fractions in sediment-trap samples (Tachikawa et al., 1997), all the samples from this study (aerosols, sediment-trap samples and Barbados samples) show positive Eu anomalies. The individual shipboard-collected aerosol samples seem to mostly group together for each sampling campaign, indicating this dust, sampled in close proximity not more than a day apart, likely has a stable source area.

The shipboard-collected aerosol samples show Sm-Nd model ages and La_N/Lu_N values similar to the aerosol samples collected at Barbados, and to sediments from the Bodélé depression. Contrastingly, the data differ greatly from sediments from the Madeira River and Amazon Basin, but are similar to sediments collected from the Solimoes River (Fig. 7.6D).

7.5 Conclusions

Our results do not show a systematic shift in Sr isotope composition with dust particle size as expected with changes in mineralogy of transported particles of progressively smaller size. We therefore hypothesize that a combination of particle size, related to transport distance and seasonal changes in dust transport, and changes in dust provenance should be considered when interpreting Sr isotope data in aerosols. Also the Nd isotopic compositions do not show clear relations to dust particle size, in line with previous results (Feng et al., 2009; Meyer et al., 2011). The relation to particle size as shown by the Hf isotopic composition is due to the zircon effect (Patchett et al., 1984; Aarons et al., 2013), as coarse-grained zircon particles are deposited close to the source, thereby progressively shifting the Hf isotopic composition of aerosols to more radiogenic values with distance from the source. Therefore, the Hf isotopic composition would only be suited for source identification when the dust is sampled in a single location, as the Hf isotopic composition is influenced by dust transport distance and particle size. When sampling in a single location, the Hf isotopic composition would only change as a function of changing sources, and not as a result of changing sampling locations.

The similarity of REE patterns and isotopic composition of the samples from the distal sampling station M4 to the most proximal station M1 shows that Saharan dust dominates the terrigenous fraction all along the trans-Atlantic transect, and that the contribution of Amazon-derived sediments to the sediment-traps is minimal. The REE patterns of the sediment-trap samples and shipboard-collected aerosols are also similar to dust collected at Barbados and samples from the Bodélé Depression in Chad, and very different from Amazon basin sediments. This makes an Amazon origin of the lithogenic particles again unlikely. The REE data from two tributaries of the Amazon River, however, are not conclusive, albeit expected to become more fractionated downstream. These are excluded as potential sediment sources to the sediment-trap samples due to their different Sr-Nd isotopic signatures to the Saharan dust samples. Rather, the combination of Sr-Nd isotopes of the dust samples point to a source somewhere in the western region of northern Africa.

Sediment-trap samples that sampled at the same time interval (M1-12 and M4-12; M1-24 and M4-24) are expected to have the same dust source, as they were sampled at the same time and at the same water depth. This is because the source of the dust within one dust cloud can be assumed to remain the same downwind, as no additional dust is added to the dust cloud while crossing the Atlantic Ocean. However, the composition of the dust can slightly change downwind due to the sedimentation of coarser and heavier particles earlier during transit, although the source remains the same. Differences in isotopic signature are therefore related to transport distance as a result of size and mineralogy fractionation, and not to changes in dust provenance for that particular time interval. The same can be seen when comparing the sediment-trap samples to dust collected even further downwind at Barbados in the same sampling year. However, the shipboard-collected aerosol samples from individual sampling campaigns are collected in close proximity to each other with little time between them, and also show a similar compositional variability as the sediment-trap samples, which is apparently a normal range for the average isotopic composition of a dust cloud. Sampling on an even higher resolution in both space and time should give more insights on the reproducibility of such shipboard-collected aerosols, and the short-term evolution of the (average) isotopic composition of a dust cloud. In interpreting the geochemical composition of aerosols, the influence of source provenance based on soil composition, sampling season and year, transport mechanisms, distance from the source and thereby particle size and mineralogy segregation should be considered.

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Supplement

Table S7.2. Sr-Nd-Hf isotope data of samples used in this study (from sediment traps, shipboard-collected aerosols and Mauritanian soil sediments). This data is compared to literature, comprising samples collected at Barbados (Pourmand et al., 2014; Bozlaker et al., 2018), from the Amazon Basin and Bodélé Depression, Chad (Abouchami et al., 2013), and two tributaries from the Amazon River, the Solimoes and Madeira Rivers (Viers et al., 2008), available in the online version of this paper. M4-Bulk samples represent analyses on bulk sediment-trap samples, including the marine fraction. Sr isotopic values closely resemble the global seawater value of 0.709202 ± 0.000003 (Kuznetsov et al., 2012).

Sample ID	⁸⁷ Sr/ ⁸⁶ Sr Adjusted	95% CI	¹⁴³ Nd/ ¹⁴⁴ Nd	95% CI	εNd	95% CI	¹⁷⁶ Hf/ ¹⁷⁷ Hf	95% CI	εHf	95% CI
M1-U9	0.722220	0.000010	0.511944	0.000010	-13.4	0.20	0.282351	0.00001	-15.3	0.30
M1-U12	0.722219	0.000008	0.511931	0.000006	-13.6	0.11	0.282347	0.00002	-15.5	0.55
M1-U24	0.723324	0.000013	0.511912	0.000006	-14.0	0.11	0.282298	0.00001	-17.2	0.41
M4-U12	0.722835	0.000011	0.511971	0.000017	-12.9	0.33	0.282523	0.00002	-9.3	0.68
M4-U24	0.718942	0.000011	0.511976	0.000002	-12.8	0.05	0.282422	0.00003	-12.8	0.98
M4-U12-Bulk	0.709284	0.000005	0.512004	0.000006	-12.2	0.12	0.282478	0.00004	-10.9	1.44
M4-U24-Bulk	0.709410	0.000006	0.511994	0.000008	-12.4	0.16	0.282524	0.00003	-9.2	1.18
M65-D3	0.710224	0.000021	0.512025	0.000047	-11.8	0.92	0.282737	0.00014	-1.7	5.00
M65-D4	0.713739	0.000019	0.511982	0.000033	-12.6	0.64	0.282289	0.00001	-17.5	0.38
M65-D5	0.711212	0.000015	0.511962	0.000018	-13.0	0.35	0.282284	0.00002	-17.7	0.86
M89-F09	0.713991	0.000013	0.511986	0.000018	-12.6	0.34	0.282519	0.00002	-9.4	0.73
M89-F10	0.714388	0.000007	0.512000	0.000022	-12.3	0.42	0.282525	0.00002	-9.2	0.61
M89-F11	0.714739	0.000013	0.511977	0.000022	-12.7	0.43	0.282505	0.00001	-9.9	0.31
64PE395-F04	0.714097	0.000008	0.512058	0.000013	-11.2	0.26	0.282572	0.00003	-7.5	1.02
64PE395-F06	0.713781	0.000011	0.512003	0.000014	-12.2	0.27	0.282552	0.00002	-8.2	0.64
64PE395-F08	0.714866	0.000012	0.512025	0.000032	-11.8	0.63	0.282534	0.00001	-8.9	0.43
PNBA	0.723066	0.000028	0.511882	0.000007	-14.6	0.13	0.282292	0.00001	-17.4	0.32
Akjoujt	0.745105	0.000011	0.511605	0.000009	-20.0	0.17	0.281698	0.00001	-38.4	0.47
Jraif	0.727067	0.000013	0.511977	0.000011	-12.7	0.21	0.282240	0.00002	-19.3	0.81

Table S7.3. Rare Earth Element (REE) concentrations (top), concentrations normalised to Post Archean Australian Shale (PAAS), and REE anomalies of samples used in this study (from sediment traps, shipboard-collected aerosols and Mauritanian soil sediments). This data is compared to samples collected at Barbados (Pourmand et al., 2014; Bozlaker et al., 2018), from the Amazon Basin and Bodélé Depression, Chad (Abouchami et al., 2013), and two tributaries from the Amazon River, the Solimoes and Madeira Rivers (Viers et al., 2008), available in the online version of this paper.

REE Concentrations (ng/g)														
Sample ID	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
M1-U9	38406.4	64796.9	7817.7	28339.9	5093.0	1076.6	4805.4	739.2	4543.2	961.8	2888.8	475.6	3177.8	519.9
M1-U12	37093.8	64123.3	7732.3	27788.6	4916.0	1026.2	4617.1	701.4	4328.2	915.7	2786.8	452.7	3087.6	494.2
M1-U24	36265.6	67051.3	7741.7	28038.8	5028.9	1086.5	4872.7	751.0	4710.7	993.7	3052.0	486.9	3362.8	528.1
M4-U12	40770.8	72175.6	8516.1	30476.8	5125.9	1025.4	4511.4	633.3	3699.2	784.3	2337.5	397.8	2675.1	437.9
M4-U24	41023.6	74152.5	8653.0	31188.7	5437.5	1122.5	4986.2	724.7	4347.5	901.5	2690.3	432.9	2932.2	462.4
M4-U12-Bulk	9651.7	21059.2	2250.5	8576.5	1703.6	372.4	1575.2	237.5	1303.7	275.1	766.9	143.2	842.5	167.1
M4-U24-Bulk	12597.0	25124.4	2919.2	11111.5	2188.0	477.3	2096.3	311.0	1779.9	368.9	1063.4	177.9	1156.3	197.1
M65-D3	8351.5	17814.5	1900.3	7334.8	1353.0	289.7	1260.8	172.2	967.6	184.1	510.8	72.9	469.2	68.8
M65-D4	10146.1	21364.5	2406.6	9345.6	1798.2	393.2	1697.5	243.9	1423.8	277.7	790.2	112.9	738.6	107.7
M65-D5	35756.1	78555.2	8665.6	34110.1	6600.4	1431.4	6150.8	894.1	5224.7	1013.7	2856.9	401.7	2618.1	374.9
M89-F09	36328.8	76332.6	8494.9	32586.6	6191.3	1311.8	5692.1	792.7	4524.5	869.8	2454.2	347.5	2355.1	327.9
M89-F10	11922.7	24787.8	2756.8	10517.4	1987.3	418.2	1783.4	251.0	1433.9	275.1	775.9	109.7	747.9	103.6
M89-F11	10847.1	22501.9	2494.0	9496.9	1792.5	378.0	1608.9	227.2	1300.3	249.8	705.1	99.9	682.9	94.5
64PE395-F04	19563.7	42275.6	4540.4	17911.6	3372.0	701.7	3163.8	436.6	2474.8	471.9	1312.9	182.2	1211.9	167.0
64PE395-F06	48775.4	106086.5	11139.7	44097.1	8266.4	1736.4	7707.7	1072.2	6113.2	1167.8	3267.8	455.8	3057.2	419.9
64PE395-F08	16181.8	36052.1	3726.0	14461.6	2699.6	571.5	2387.9	337.5	1920.2	363.0	1011.3	141.9	961.3	130.7
PNBA	7283.0	13039.2	1737.8	6637.2	1327.8	316.5	1110.1	167.9	911.0	199.9	570.5	116.2	756.9	156.7
Akjoujt	8995.4	15936.9	1875.1	6970.1	1377.2	401.2	1332.5	216.6	1223.1	269.8	747.7	146.6	889.3	181.8
Jraif	13004.0	24569.7	3155.4	12188.2	2435.2	518.3	2224.8	316.9	1774.0	349.9	986.8	158.1	1086.2	175.4

REE PAAS-normalized														
Sample ID	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
M1-U9	0.862	0.734	0.770	0.759	0.740	0.886	0.795	0.829	0.853	0.913	0.939	1.054	1.055	1.185
M1-U12	0.832	0.727	0.762	0.745	0.714	0.845	0.764	0.787	0.813	0.870	0.906	1.004	1.025	1.127
M1-U24	0.814	0.760	0.763	0.751	0.731	0.894	0.806	0.843	0.885	0.944	0.993	1.080	1.116	1.204
M4-U12	0.915	0.818	0.839	0.817	0.745	0.844	0.747	0.711	0.695	0.745	0.760	0.882	0.888	0.999
M4-U24	0.921	0.840	0.853	0.836	0.790	0.924	0.825	0.813	0.816	0.856	0.875	0.960	0.974	1.054

Sample ID	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
M4-U12-Bulk	0.217	0.239	0.222	0.230	0.247	0.307	0.261	0.266	0.245	0.261	0.249	0.318	0.280	0.381
M4-U24-Bulk	0.283	0.285	0.288	0.298	0.318	0.393	0.347	0.349	0.334	0.350	0.346	0.394	0.384	0.449
M65-D3	0.187	0.202	0.187	0.197	0.197	0.238	0.209	0.193	0.182	0.175	0.166	0.162	0.156	0.157
M65-D4	0.228	0.242	0.237	0.250	0.261	0.324	0.281	0.274	0.267	0.264	0.257	0.250	0.245	0.246
M65-D5	0.802	0.890	0.854	0.914	0.959	1.178	1.018	1.003	0.981	0.963	0.929	0.891	0.869	0.855
M89-09	0.815	0.865	0.837	0.873	0.899	1.080	0.942	0.889	0.850	0.826	0.798	0.771	0.782	0.748
M89-10	0.268	0.281	0.272	0.282	0.289	0.344	0.295	0.282	0.269	0.261	0.252	0.243	0.248	0.236
M89-11	0.243	0.255	0.246	0.254	0.260	0.311	0.266	0.255	0.244	0.237	0.229	0.222	0.227	0.215
64PE395-F04	0.439	0.479	0.447	0.480	0.490	0.578	0.524	0.490	0.465	0.448	0.427	0.404	0.402	0.381
64PE395-F06	1.095	1.202	1.098	1.182	1.201	1.429	1.275	1.203	1.148	1.109	1.063	1.011	1.015	0.957
64PE395-F08	0.363	0.409	0.367	0.388	0.392	0.470	0.395	0.379	0.361	0.345	0.329	0.315	0.319	0.298
PNBA	0.163	0.148	0.171	0.178	0.193	0.260	0.184	0.188	0.171	0.190	0.186	0.258	0.251	0.357
Akjoujt	0.202	0.181	0.185	0.187	0.200	0.330	0.221	0.243	0.230	0.256	0.243	0.325	0.295	0.415
Jraif	0.292	0.278	0.311	0.327	0.354	0.427	0.368	0.356	0.333	0.332	0.321	0.351	0.361	0.400

Sample ID	Ce anomaly	%	Eu anomaly	%	ΣLREE	ΣHREE	LREE/HREE	La _N /Lu _N	Sm/Nd	¹⁴⁷ Sm/ ¹⁴⁴ Nd	Sm-Nd Model Age (Ga)
M1-U9	0.9032	-9.7	1.1511	15.1	3.866	7.625	0.51	0.727	0.180	0.10866	1.8
M1-U12	0.9141	-8.6	1.1395	14.0	3.780	7.295	0.52	0.739	0.177	0.10697	1.8
M1-U24	0.9656	-3.4	1.1594	15.9	3.818	7.870	0.49	0.676	0.179	0.10845	1.8
M4-U12	0.9351	-6.5	1.1317	13.2	4.133	6.425	0.64	0.916	0.168	0.10170	1.6
M4-U24	0.9499	-5.0	1.1419	14.2	4.239	7.173	0.59	0.873	0.174	0.10542	1.7
M4-U12-Bulk	1.0884	8.8	1.2038	20.4	1.154	2.261	0.51	0.569	0.199	0.12010	1.9
M4-U24-Bulk	0.9981	-0.2	1.1780	17.8	1.471	2.954	0.50	0.629	0.197	0.11907	1.9
M65-D3	1.0777	7.8	1.1741	17.4	0.970	1.399	0.69	1.195	0.184	0.11154	1.7
M65-D4	1.0411	4.1	1.1905	19.0	1.219	2.084	0.58	0.927	0.192	0.11635	1.8
M65-D5	1.0741	7.4	1.1890	18.9	4.419	7.508	0.59	0.939	0.194	0.11700	1.9
M89-09	1.0466	4.7	1.1703	17.0	4.290	6.605	0.65	1.090	0.190	0.11488	1.8
M89-10	1.0416	4.2	1.1781	17.8	1.391	2.087	0.67	1.133	0.189	0.11425	1.8
M89-11	1.0424	4.2	1.1803	18.0	1.259	1.896	0.66	1.130	0.189	0.11413	1.8
64PE395-F04	1.0805	8.1	1.1367	13.7	2.335	3.540	0.66	1.153	0.188	0.11383	1.7
64PE395-F06	1.0967	9.7	1.1513	15.1	5.777	8.781	0.66	1.143	0.187	0.11335	1.8
64PE395-F08	1.1187	11.9	1.1944	19.4	1.918	2.740	0.70	1.219	0.187	0.11287	1.7

Sample ID	Ce anomaly	%	Eu anomaly	%	ΣLREE	ΣHREE	LREE/HREE	La _N /Lu _N	Sm/Nd	¹⁴⁷ Sm/ ¹⁴⁴ Nd	Sm-Nd Model Age (Ga)
PNBA	0.8824	-11.8	1.3871	38.7	0.853	1.785	0.48	0.457	0.200	0.12097	2.1
Akjoujt	0.9368	-6.3	1.5645	56.4	0.954	2.227	0.43	0.487	0.198	0.11948	2.5
Jraif	0.9232	-7.7	1.1796	18.0	1.561	2.821	0.55	0.730	0.200	0.12081	1.9

Chapter 8:

Concluding Remarks

Within the project “*TRAFFIC: Trans-Atlantic fluxes of Saharan dust and ocean-climate impacts*” I tried to quantify and characterize the transport and deposition of Saharan dust along a trans-Atlantic transect. This project was a collaboration with the ERC-funded project DustTraffic, which focused on the marine-environmental effects of dust deposition in the Atlantic Ocean, including the bio-availability of carried nutrients, the influence on plankton in the ocean, and the effects on carbon sequestration.

While many questions were answered, many more remain unanswered, or new questions arose. The studied transect is a unique set-up due to its geographic location, extent (both spatial and temporal), and the combination of the many in-situ sampling techniques, namely by sediment traps, moored surface buoys, seafloor sediments, and shipboard collected aerosol samples.

8.1 Downwind Signatures

As was shown in Chapter 3, particle size shows a sharp decrease downwind, and the seasonal amplitude is largest at the easternmost station M1. However, the amount of downwind decrease in modal particle size is not the same for every season, and the summer season shows the sharpest downwind decrease (Fig. 8.1A) compared to the other seasons. The dust deposition flux shows the same trend of sharpest downwind decrease in summer (Fig. 8.1B), although this relation is a bit more skewed as station M2 is located more to the north (13°N) than the rest of the transect (12°N). This results in an overall lower deposition flux at M2 (Chapter 4), but not significantly altering the size of the deposited dust (Fig. 8.1A). This sharper downwind decrease in summer for both particle size and dust deposition flux could be related to the large amount of wet deposition in summer, that washes the dust out of the atmosphere (Chapter 4). At M1, this results in not only deposition of the coarsest dust, but also in most dust deposition, creating a sharper downwind decrease in deposition flux in summer (Fig. 8.1B). During such a wet deposition event, I expect that particles of all sizes are approximately equally removed from the atmosphere. However, as there are generally more particles in the smaller size classes, these have a bigger chance of escaping wet deposition and are thus transported further. This would result in a sharper downwind decrease

during the wet season, and a more modest relation from autumn to spring. The effect of wet deposition could be tested using a sampler that only catches wet deposited dust. It could be mounted to the dust-collecting buoys, and use the meteorological sensor in an opposite manner: only open for sampling during rain events. The sampler could sample synchronously with the dry-dust collection of the buoys, or be programmed to only sample single wet-dust deposition events.

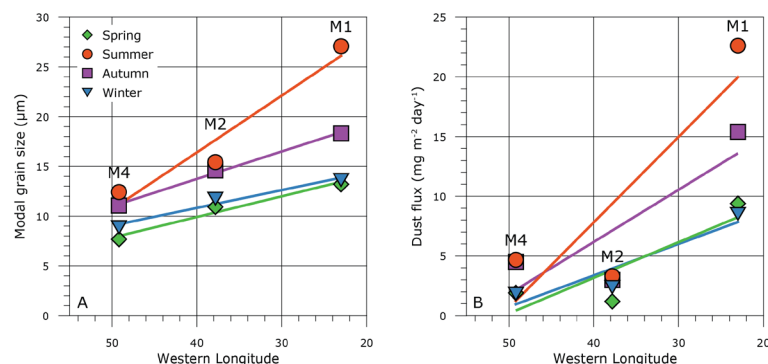


Figure 8.1. A: Average modal particle size of the upper traps (1,200 m) at station 13M1 (23°W, 12°N), 13M2 (38°W, 13°N), and 13M4 (49°W, 12°N). B: Average dust deposition flux for the same stations.

8.2 Particle Size of Dust Aerosols Collected at Barbados

Downwind the current transect at Barbados in the Caribbean, dust has been collected for over 50 years (e.g. Delany et al., 1967; Prospero et al., 1970; Prospero et al., 2014), of which mostly the atmospheric dust concentrations have been determined. The particle size of this dust reaching Barbados received far less attention, although few results indicate that median particle sizes are around 2.5 μm (Arimoto et al., 1997; Li-Jones and Prospero, 1998). Some selected samples of dust collected at Barbados (13°N, 59°W) were analysed on the *Coulter LS13 320* laser diffraction particle sizer (see Chapter 2) at NIOZ to determine the dust's particle size. Indeed, the modal grain size of these samples, collected in February, April, May, and June of 2013, ranges between 2.5 and 4.0 μm (Fig. 8.2A). This is very similar to the dust collected by the buoys in 2014, which had modal grain sizes ranging between 2.8 and 5.4 μm (Chapter 6). There is also a secondary mode on the coarse end of the grain-size distributions of the Barbados dust, which ranges between 10.8 and 34.6 μm, with individual particles in the samples detected up to 66 μm. Due to the detection limit of the laser particle sizer, giant dust particles (>75 μm) could also be present in the samples not represented in the particle-size distributions in low amounts, which was the case for the sediment-trap samples (Fig. 8.2B).

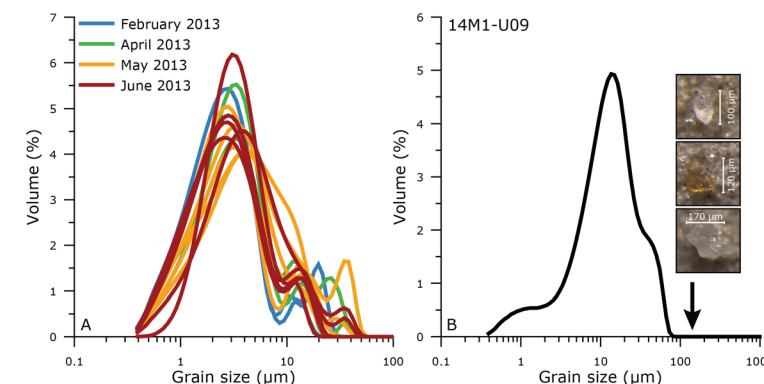


Figure 8.2. A: Grain-size distributions of dust collected at Barbados in February, April, May, and June of 2013. B: Example of giant (>100 μm) individual dust particles from one of the sediment-trap samples (14M1-U09) that remain undetected by the laser particle sizer.

The general assumption states that dust particles larger than 20 μm are expected to be deposited within the first 12 hours (Ryder et al., 2013a). Across the Atlantic Ocean in Puerto Rico, the size of dust particle generally does not exceed ~12 μm (Maring et al., 2003), although occasionally up to 30 μm (Reid et al., 2003). It is also assumed that essentially all the atmospheric dust mass arriving at Barbados is described by particles <10 μm (Prospero et al., 2014), although particles up to 30 μm have also been observed (Weinzierl et al., 2017). The results shown here are the first indication that particles >20–30 μm could play an important role for the total dust mass, with the possibility of giant (>75 μm) dust particles, reaching Barbados.

At Barbados, the dust is collected directly from the atmosphere, and can therefore be related directly to the dust-collecting buoys at two stations along the transect. By extending the current trans-Atlantic transect with an additional station on land it could provide important insights into the downwind fining of dust particles, and could reveal a possible contribution from giant dust particles. By directly comparing this to the dust deposition before this station and to other atmospheric parameters like precipitation, we could infer in more detail the mechanisms involved with the transport and deposition of Saharan dust over the Atlantic Ocean, and study more precisely the difference between atmospheric dust and dust deposition into the ocean.

8.3 Particle-Shape Analysis with Sympatec QicPic

The shape of dust particles and their downwind and seasonal or annual trends could shed more light on the sorting of particles over long distances. Large particles with a platy shape are generally transported over greater distances, due to their aerodynamical advantages (Stuut et al., 2005; Scheuvers and Kandler, 2014). For small particles (<11 μm), settling velocities are higher for more asymmetrical particles. For larger particles (>30 μm), the opposite is

true, and more symmetrical particles have higher settling velocities (Li and Osada, 2007). By determining the shape of the dust particles transported and deposited along the transect, important characteristics can be determined, such as their influence on radiation budgets and fertilising or ballasting potential. Due to the different surface area, particles with different shapes can play a role in chemical reactions acting on the particles (Scheuvers and Kandler, 2014). In dust archives, particle shape can be used as a provenance tracer (Shang et al., 2017), or an indicator for aeolian transport processes (Tysmans et al., 2009).

The image analyses carried out using the *Sympatec QicPic* (Chapter 2) can not only be used for determining the particles' size, but also their two-dimensional shape. One way to express the (symmetrical) shape of a particle is with its aspect ratio, which is the ratio of the minimum to the maximum Feret diameter (Fig. 8.3C). A high aspect ratio implies a highly symmetrical particle in 2D. Other parameters calculated by the *QicPic* are: (1) the diameter of a circle of equal projection area (EQPC); (2) sphericity, calculated as the ratio of the circumference of the EQPC to the real circumference; and (3) its convexity, defined as the ratio of the projection area itself and the area of the convex hull. These shape parameters rely on the number of pixels that make up the individual particles. However, since some dust particles appear nearly transparent and are not completely captured by the camera, these parameters can potentially give a false indication of the particle's characteristics (Fig. 8.2B of Chapter 2). The aspect ratio does not rely on the number of pixels that make up the particle, and is therefore particularly suited for determining the shape of mineral dust particles.

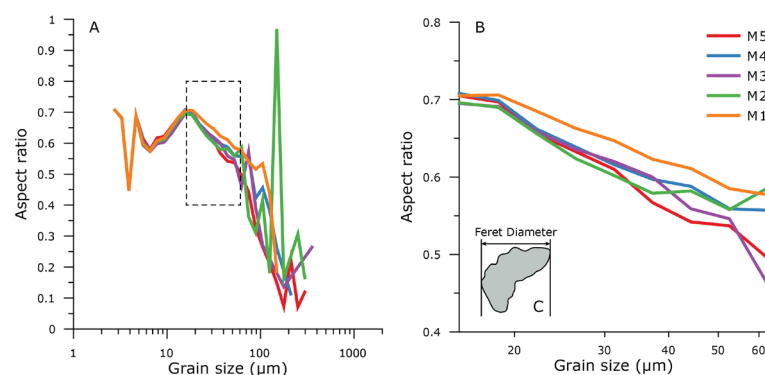


Figure 8.3. A: Aspect ratio of seafloor sediments collected at station M1–M5 for all particle sizes. B: Zoom for particles between 15.6 and 62.5 μm . C: Definition of the Feret diameter used by the *Sympatec QicPic*.

Analysis with the *QicPic* was performed on seafloor core-top sediments from stations M1–M5. The results reveal nearly identical aspect ratios for all samples up to sizes of 15 μm (Fig. 8.3A). However, due to the limited resolution of the camera of $2 \times 2 \mu\text{m}$, these particles are made up of very few pixels, resulting in little variation in the shape of the particles. Contrastingly, particles larger than $\sim 70 \mu\text{m}$ have very variable

and irregular shapes (Fig. 8.3A), partially related to the 2-dimensional nature of the method, and the low amount of particles in these size classes. For particles between 15.6 and 62.5 μm , the aspect ratio clearly decreases with increasing particle size, meaning that larger particles are less symmetrical than smaller particles (Fig. 8.3B). This would imply a non-linear increase in settling velocity with increasing particle size (Li and Osada, 2007). Dust collected at station M1 shows highest symmetry in every size class. At the other stations more downwind the symmetry is lower, but similar for M1–M5. This can also be due to the low amount of particles in the larger ($>40 \mu\text{m}$) size classes at the more distal stations. The results show that asymmetrical particles are favoured in their downwind transport (e.g. platy mica particles), which is most notably from station M1 ($\sim 700 \text{ km}$ from the northwest African coast) to M2 ($\sim 2,300 \text{ km}$), and no additional decrease in aspect ratio is observed further downwind. Rather, fine-grained particles seem to be favoured over particles of a specific shape in their transport downwind. In addition, particle shape shows seasonal variations, with presumably more platy-shaped minerals in spring (Chapter 3), which could rather be related to the mineralogy of the source soil.

Thus far, no record of the downwind particle-shape evolution of Saharan dust exists, and the samples from this transect prove to be very suitable. Future analysis of the sediment-trap samples will not only shed more light on the downwind trends in dust particle shape, but may also reveal seasonal or inter-annual differences, possibly related to dust-transporting wind systems and transport altitude, and the source area of the dust particles. Additionally, it might reveal differences in shape between dust sampled by the buoys that collect airborne dust only, and the sediments traps that collect both wet- and dry-deposited dust (Chapter 6). However, a great disadvantage of particle-shape analysis with the *QicPic* is that the analysis relies on the resolution of the camera. As 90–99% of all particles in our dust samples are $<15 \mu\text{m}$, most of the sample is represented by particles made up of too few pixels to identify the particle's shape. Although particles between 15.6 and 62.5 μm seem to produce promising results, this makes it a very selective process, and the analysed particles may not be a proper representation of the entire sample to give reliable results. It would, however, be a great addition to the characterisation of giant ($>75 \mu\text{m}$) mineral dust particles (Chapter 5).

8.4 Continuation with the Existing Transect

Extending the duration of the trans-Atlantic monitoring project, resulting in the long-term trends of dust transport and deposition, would give many more insights in the processes involved. Especially the most proximal station M1, where unfortunately the second year of sampling was not complete due to a malfunctioning carousel, could reveal many more insights on the seasonal variability of dust transport and deposition. Sampling at a higher resolution, as was done for the next year (2016) of sampling, could shed more light on for instance the difference between the dust collected by the upper (1,200 m) and lower (3,500 m) sediment traps, as discussed in Chapter 6. This could reveal the mentioned size separation

that happens during large dust events, when in the deeper traps the coarse-grained particles are observed first due to their higher settling velocity, but the associated higher deposition flux lags behind that as the finer-grained particles take longer to settle down to the deeper trap (Clemens, 1998). Also, identifying the geochemical and mineralogical characteristics of the dust collected along the transect could inform on the changing dust sources with the seasons and between different years (e.g. Friese et al., 2017), transport pathways, and downwind depositional trends. The different mineral and chemical compositions of dust from different source areas could in turn affect its fertilising and mineral ballast potential.

To reduce the risk of losing failing equipment in the east of the transect, where most dust is deposited, two instead of one mooring could be deployed, for instance at 23°W (currently M1) and 25°W (Fig. 8.4). This will also allow for a very detailed analysis of downwind trends. This additional mooring could replace the mooring at M5, as the origin of the lithogenic particles in this sediment trap is not clear, and possible contributions from particles other than mineral dust are conceivable (Korte et al., 2018a). The east of the transect could then be complemented by one of the dust-collecting buoys, while the other remains in the middle of the transect (Fig. 8.4).

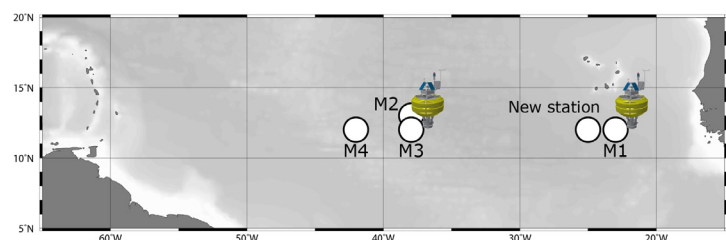


Figure 8.4. Proposed new transect of moorings at the four existing stations M1–M4, and one additional station at 12°N, 25°W. Buoys could be placed at M1 and M3.

8.5 Conclusions

This study has provided many new insights on the transport and deposition of mineral dust from northern Africa over and across the Atlantic Ocean. The collaboration between the two joint projects regarding this dust-monitoring transect (i.e. TRAFFIC and DustTraffic) has led to many more important insights beyond this dissertation, and this multi-disciplinary approach showed the biological impacts of deposited mineral dust in the ocean (e.g. Korte et al., 2018b). All together, these findings can be used for interpreting paleo-dust records, and for calibrating climate models predicting future dust emissions in a changing climate and in turn the response of climate to these changes in the dust cycle.

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Biography

Michèle van der Does was born on September 18th 1990 in Slootdorp, The Netherlands. She graduated from secondary school (VWO) at 'RSG Wiringherlant' in 2008. She then started her Bachelor Earth Sciences at the Vrije Universiteit in Amsterdam, with a specialisation in Geology. Her thesis focused on climatic variations recorded in speleothems from Belgium. She further specialized with a Master in Applied Environmental Geosciences, starting in 2011. Her first Master project focused on Saharan dust deposition off the northwest African coast from submarine sediment traps offshore Cape Blanc. For her thesis she studied trans-Atlantic dust deposition from seafloor sediments and aerosols, for which she joined her first research cruise across the Atlantic. In 2013 she started as a PhD student at the Royal Netherlands Institute for Sea Research on Texel, for which she also spent four weeks in Miami, FL USA. Her work has accumulated in this PhD thesis, focusing on the transport and deposition of Saharan dust across the tropical North Atlantic Ocean.

