The mysterious long-range transport of giant mineral dust particles

van der Does, M., Knippertz, P., Zschenderlein, P., Harrison, G. and Stuut, J.-B. (2018) The mysterious long-range transport of giant mineral dust particles. Science Advances, 4 (12). eaau2768. ISSN 2375-2548 doi: https://doi.org/10.1126/sciadv.aau2768 Available at http://centaur.reading.ac.uk/80426/

To link to this article DOI: http://dx.doi.org/10.1126/sciadv.aau2768

Publisher: American Association for the Advancement of Science

The mysterious long-range transport of giant mineral dust particles

Michèlle van der Does[1*], Peter Knippertz[2], Philipp Zschenderlein[2], R. Giles Harrison[3], Jan-Berend W. Stuut[1]

[1] NIOZ – Royal Netherlands Institute for Sea Research, Department of Ocean Systems, and Utrecht University, Texel, The Netherlands
[2] Institute of Meteorology and Climate Research, Karlsruhe Institute of Technology, Karlsruhe, Germany
[3] Department of Meteorology, University of Reading, Reading, UK
[*] Correspondence to: mdoes@nioz.nl

Short title:
Long-range transport of giant dust particles

One-sentence summary:
Strong winds, turbulence, convective uplift and electric charge may keep giant particles suspended in air much longer

Abstract

Giant mineral dust particles (>75 µm 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 µm 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.

1. Introduction

About 30 years ago, scientists first observed so-called giant (>75 µm diameter) wind-blown mineral dust particles at large (>10,000 km) distances from their source (*1*). These sand-sized mineral aerosols or dust particles, the largest of which (>200 µm) were all individual quartz grains, were transported from Asia to the remote Pacific Ocean. In Europe, giant dust particles were found >4,000 km from their Saharan source (*2*), and dust particles up to 300 µm in diameter were sampled during aircraft campaigns over northwestern Africa (*3*). In marine sediment traps, positioned underneath the main Saharan dust plume in the Atlantic Ocean, giant particles are dominated by large quartz particles >100 µm, found at distances up to 4,400 km from the west African coast (*4*).

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 (*5*). 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 (*5*-*7*). Wind velocities of up to 25 m s^{-1} associated with the Atlantic extension of the African easterly jet can lead to fast westward transport, particularly around 4 km (*6*, *8*).

It is often assumed that the particle size of long-range transported mineral dust does not exceed 20–30 µm (*9*-*12*), and climate model simulations often limit particle diameters to only <10 µm (*13*). 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 (*3*), and enhance the absorption of longwave radiation (*14*), possibly causing a net atmospheric warming as shown by Kok et al. (*15*). This latter study (*15*) also demonstrated a significant effect on the atmospheric radiative balance when larger particles up to 20µm are incorporated into climate models. If giant dust particles (>75µm) would be considered, the effects on atmospheric radiation budgets could be tremendous. In addition, the under- (or non-) representation of particles larger than 10 µm 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 (*16*). In addition, they influence cloud microphysics by acting as giant cloud condensation nuclei, which can accelerate the hydrological cycle through increasing precipitation rates (*17*). This demonstrates why a mechanism explaining the long-range transport of giant dust particles is urgently needed.

2. Results and Discussion

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. (*4*), 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 S1) at approximately three meters above sea level. These samples show giant dust particles (>75 µm; Fig. 1) that were collected at 2,400 and 3,500 km, respectively, from the west African coast at sampling stations M3 and M4 (Fig. 2). These are mostly well-rounded quartz particles up to 450 µm 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.

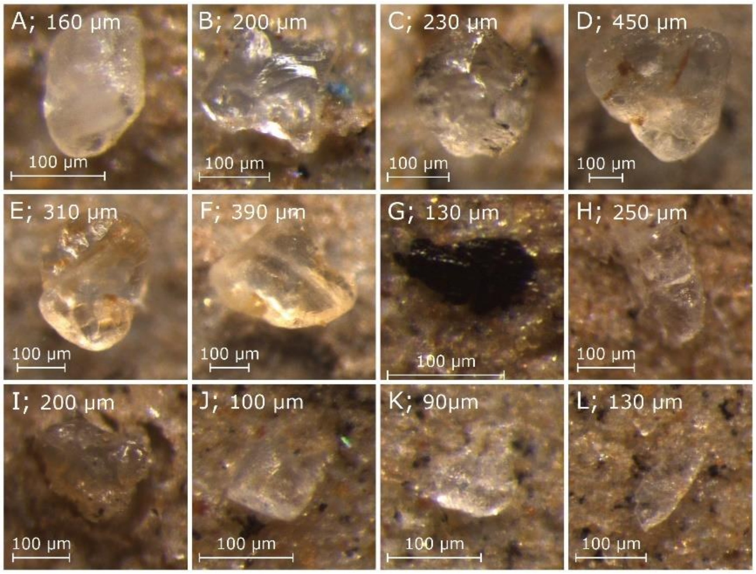

Figure 1. Giant mineral dust particles sampled by the MWAC samplers at M3 (12° N, 38° W) and M4 (12°N, 49°W) in 2014 and 2015, with their approximate diameters. A–C: 2014-M3; D–F: 2014-M4; G–I: 2015-M3; J–L: 2015-M4.

The main reason why climate models do not incorporate dust particles >10 µm, despite the growing evidence for their existence for 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 (*10*, *18*). Settling velocities for particles >20 µm are overestimated by Stokes' Law due to turbulence created by the falling of larger particles and are therefore determined empirically (*10*, *19*). Using the formula from Bagnold (*19*), we compare the settling velocities of giant mineral dust particles of 100, 200, and 300 µm (Table 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 (Fig. 2), even at high wind velocities of 25 m s^{-1}. Clearly some additional mechanisms are needed to keep these dust particles aloft.

Potential Mechanisms

Several studies suggest such mechanisms including atmospheric vertical mixing (*18, 20, 21*) or large dust storms and turbulence (*1, 9, 11*), 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 (*22, 23*). 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. 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 (*24*), 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 (*25, 26*). 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.

Table 1. Settling velocities after Bagnold (19), 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})^{a}	**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	1000	175 km	30 km
300	1500	117 km	20 km

Winter Scenario

In boreal winter, dust transport occurs at lower altitudes and at lower wind speeds (Table 1). There is insufficient convection over the sampling area to aid the long-range transport of dust particles, as the ITCZ is shifted southward (*27*). 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. 2A). Transport at higher levels is unlikely, as winds become increasingly more westerly with height (*28*). Assuming the sedimentation velocities for particles of 100 μm given in Table 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.

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 (*29*), 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.

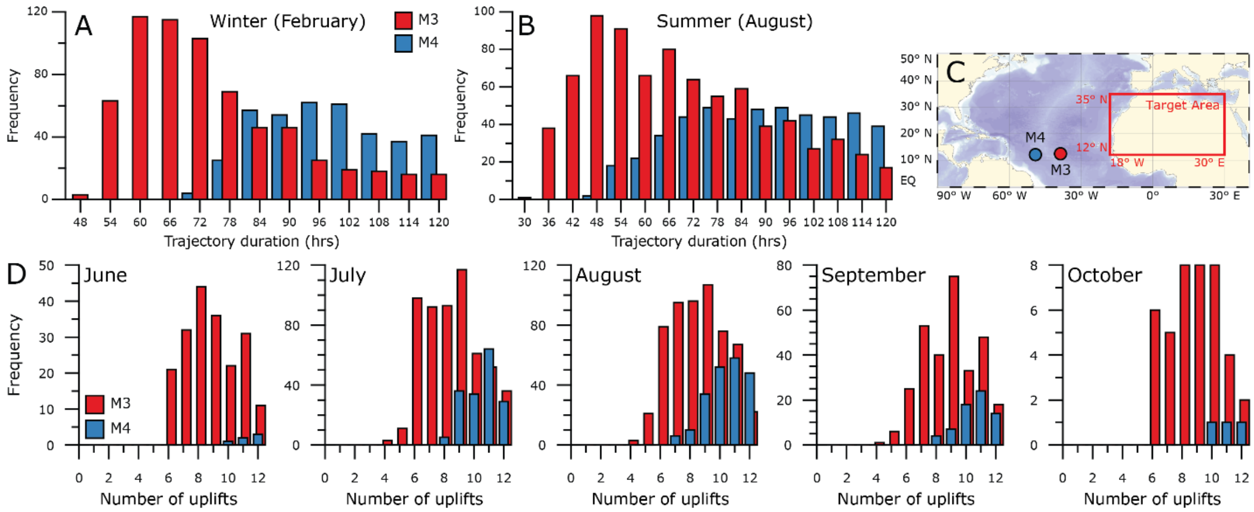

Figure 2. Distribution of travel times for backward trajectories from buoys M3 (red) and M4 (blue) to the target area (panel C), for February (A) and August (B). D: Frequency of the minimum number of deep convective uplift cycles needed for a 100 µm particle to travel from the target area (panel C) to the sampling buoys M3 (red) and M4 (blue), assuming a constant sedimentation velocity of 400 mm s^{-1} *(Table 1), for June–October. All computations are based on ERA-Interim data during the ten-year period 2006–2015.*

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 (*25*, *26*). Renard et al. (*26*) 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 (*30*, *31*). 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 (*32*) 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 (*25*) 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 (*33*), 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 (*32*, *34*). 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 ($E = -10^2$ Vm^{-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 ($E = \pm \sim 10^4$ Vm^{-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 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 (~10^4 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. 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 ~10^6 *e* cm^{-3} (*34*) and re-suspended dust on the order of 10^3 to 10^4 *e* / particle (or 10^{12} *e* cm^{-3}) (*32*). Larger charges are likely for giant particles, scaling with particle area, but at very large charges and fields, charge emission would prevent further electrification (*35*). One highly relevant factor is particle composition (*30, 32*), as it is known that mineralogy affects the charging of particles. The giant dust particles found at the buoys are mostly quartz (Fig. 1) and it is found experimentally that quartz particles may charge more easily than clay minerals (*32, 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.

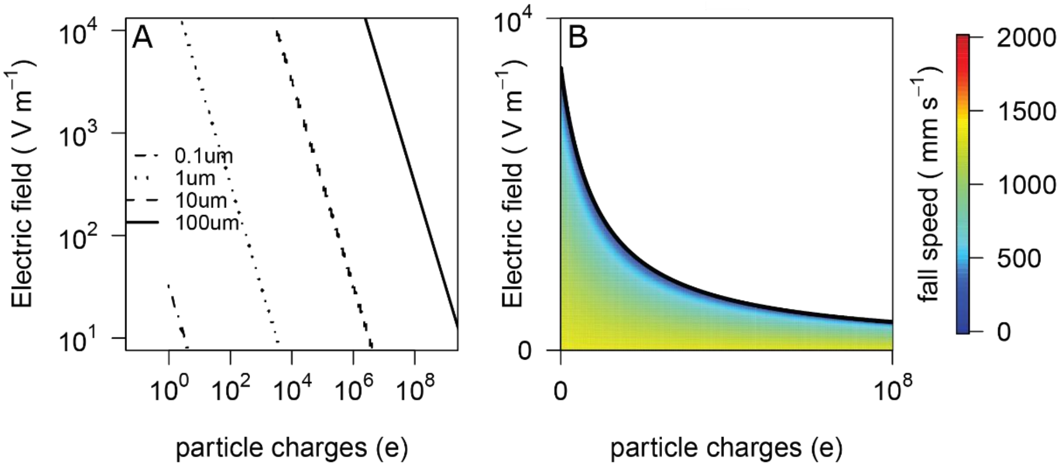

Figure 3. Influence of charge and electric field on the net force on a particle. A: Combination of particle charge and local electric field required for the magnitude of the electric force experienced by a particle to equal the particle's weight, for particles having diameters of 0.1, 1, 10 and 100 µm. 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 $C_D = 1.5$).

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 (*36*); see Methods) at the latitude of the buoys (i.e., ~12°N) occurs within the African easterly jet (*37*), reducing the minimum transport time in August to M3 at 600 hPa to 30 hours, and to M4 to 48 hours (Fig. 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 (*4*). 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 (*25*). 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 (*27*), where convection is frequent. In August the tropical cyclone season also intensifies (e.g. *38*). Convective cells can uplift individual particles to the high tropical tropopause at altitudes of about 15 km (*37, 39, 40*). 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 (*41*). 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 (*37*).

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 1; *19*), 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. 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. 2D).

According to satellite estimates, every uplift cycle reduces the mass of dust by about a factor of 60, as 1.6% (±0.7%) of the Saharan dust layer mass can escape the deep-convective cloud between 8 and 12 km (*41*). 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 (*10, 41*). As initial giant (37.5-300 µm) dust particle concentrations within 12 hours of uplift are estimated by Ryder et al. (*11*) to be up to 12,000 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

(*42*). 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 (*25*), and most dust uplifted by convection is deposited by wet deposition (*10*, *41*). Therefore, convection and charging could also potentially work against each other.

3. Summary and Conclusions

We presented evidence that giant mineral dust particles are transported through the atmosphere across the Atlantic Ocean, thousands of kilometers 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 (*5*-*7*), 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 carrousel 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.

Material and Methods

MWAC Samplers

In conjunction with the sediment traps described in Van der Does et al. (*4*), 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. 2), for two consecutive years (Table S1). These buoys were equipped with one MWAC (Modified Wilson and Cooke; *43*) 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 (*43*) conclude that the MWACs are the least inefficient samplers. Sampling efficiency varies slightly with different wind speeds, but without apparent trends (*43*). 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 (*44*). Maximum wind velocities at the sampling stations approximated 14 m s^{-1} (Table S1). 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.

Table S1. Sampling duration of MWAC samplers on the dust-collecting buoys at M3 and M4 (Fig. 2) together with statistics on the co-located wind measurements.

	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

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. (*4*) (Fig. S1). 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. S1).

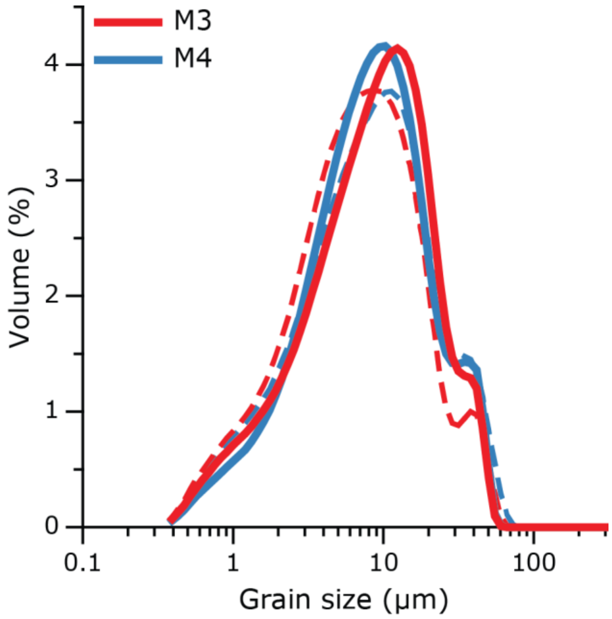

Figure S1. Grain-size distributions of MWAC samples collected in 2014 (solid lines) and 2015 (dashed lines).

We have tried to eliminate any possible contamination of giant particles to our dust samples. Microscope analysis (Fig. 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 carrousel 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 (*45*). 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.

Estimation of travel paths based on Stokes' Law

Typically, dust is lifted to a maximum altitude of about 7 km within the SAL (*5*). 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 (*19*), 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; *4*), 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.

Trajectory calculation for giant mineral dust particles

Five-day backward trajectories were calculated using Lagranto (*46*, *47*) 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:

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

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 (*36*) 3-dimensional wind fields. The trajectories were started every six hours at the locations of buoys M3 and M4 (see Fig. 2), resulting in a total of about 1200 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. 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 (*5*, *23*, *48*). 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 (*28*). 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 (*49*).
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 (*50*).
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 (*19*) was added globally. This settling velocity was converted into pressure coordinates, as ERA-Interim defines vertical velocities in Pa s^{-1} using equation 1:

   $$\omega = -\rho g w \qquad (1),$$

   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 2:

   $$\rho = \frac{p}{RT} \qquad (2),$$

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

   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

---

[1] Reference: https://ntrs.nasa.gov/archive/nasa/casi.ntrs.nasa.gov/19770009539.pdf; last accessed 30 January 2018

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.

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

$$U + F_E + F_D = W \qquad (3).$$

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 ~1:1000, hence $U \ll W$, and *U* can be neglected. The drag force depends on the particle's projected area *A* (= πr^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* and the drag force parameterized as $C_D \frac{1}{2} A \rho_a v^2$, hence the equilibrium description of equation (3) becomes:

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

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.

References

1. P. R. Betzer *et al.*, Long-Range Transport of Giant Mineral Aerosol-Particles. *Nature* **336**, 568-571 (1988).
2. N. J. Middleton, P. R. Betzer, P. A. Bull, Long-Range Transport of 'Giant' Aeolian Quartz Grains: Linkage with Discrete Sedimentary Sources and Implications for Protective Particle Transfer. *Marine Geology* **177**, 411-417 (2001).
3. C. L. Ryder *et al.*, Optical properties of Saharan dust aerosol and contribution from the coarse mode as measured during the Fennec 2011 aircraft campaign. *Atmospheric Chemistry and Physics* **13**, 303-325 (2013).
4. M. Van der Does, L. F. Korte, C. I. Munday, G. J. A. Brummer, J. B. W. Stuut, Particle size traces modern Saharan dust transport and deposition across the equatorial North Atlantic. *Atmos. Chem. Phys.* **16**, 13697-13710 (2016).
5. C. Tsamalis, A. Chedin, J. Pelon, V. Capelle, The seasonal vertical distribution of the Saharan Air Layer and its modulation by the wind. *Atmospheric Chemistry and Physics* **13**, 11235-11257 (2013).
6. F. Chouza, O. Reitebuch, A. Benedetti, B. Weinzierl, Saharan dust long-range transport across the Atlantic studied by an airborne Doppler wind lidar and the MACC model. *Atmos. Chem. Phys.* **16**, 11581-11600 (2016).
7. A. M. Adams, J. M. Prospero, C. Zhang, CALIPSO-Derived Three-Dimensional Structure of Aerosol over the Atlantic Basin and Adjacent Continents. *Journal of Climate* **25**, 6862-6879 (2012).
8. M. Sarnthein, G. Tetzlaff, B. Koopmann, K. Wolter, U. Pflaumann, Glacial and Interglacial Wind Regimes over the Eastern Subtropical Atlantic and North-West Africa. *Nature* **293**, 193-196 (1981).
9. H. Tsoar, K. Pye, Dust Transport and the Question of Desert Loess Formation. *Sedimentology* **34**, 139-153 (1987).
10. C. S. Zender, H. Bian, D. Newman, Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology. *Journal of Geophysical Research: Atmospheres* **108**, (2003).
11. C. L. Ryder, E. J. Highwood, T. M. Lai, H. Sodemann, J. H. Marsham, Impact of atmospheric transport on the evolution of microphysical and optical properties of Saharan dust. *Geophysical Research Letters* **40**, 2433-2438 (2013).
12. J. F. Kok, E. J. R. Parteli, T. Michaels, I., D. Bou Karam, The physics of wind-blown sand and dust. *Reports on Progress in Physics* **75**, 106901 (2012).
13. N. Huneeus *et al.*, Global dust model intercomparison in AeroCom phase I. *Atmospheric Chemistry and Physics* **11**, 7781-7816 (2011).
14. S. Otto *et al.*, Atmospheric Radiative Effects of an In Situ Measured Saharan Dust Plume and the Role of Large Particles. *Atmospheric Chemistry and Physics* **7**, 4887-4903 (2007).
15. J. F. Kok *et al.*, Smaller desert dust cooling effect estimated from analysis of dust size and abundance. *Nature Geoscience* **10**, 274-278 (2017).
16. H. Van der Jagt, C. Friese, J. B. Stuut, G. Fischer, M. Iversen, The ballasting effect of Saharan dust deposition on aggregate dynamics and carbon export: aggregation, settling and scavenging of marine snow. *Limnol. Oceanogr.*, (2018).
17. R. Posselt, U. Lohmann, Influence of Giant CCN on warm rain processes in the ECHAM5 GCM. *Atmos. Chem. Phys.* **8**, 3769-3788 (2008).
18. H. Maring, D. L. Savoie, M. A. Izaguirre, L. Custals, J. S. Reid, Mineral dust aerosol size distribution change during atmospheric transport. *Journal of Geophysical Research: Atmospheres* **108**, (2003).
19. R. A. Bagnold, *The physics of blown sand and desert dunes*. (Methuen, London, 1941).
20. J. Gasteiger *et al.*, Particle settling and vertical mixing in the Saharan Air Layer as seen from an integrated model, lidar, and in situ perspective. *Atmos. Chem. Phys.* **17**, 297-311 (2017).
21. C. Denjean *et al.*, Size distribution and optical properties of mineral dust aerosols transported in the western Mediterranean. *Atmos. Chem. Phys.* **16**, 1081-1104 (2016).
22. W. Yang, A. Marshak, A. B. Kostinski, T. Várnai, Shape-induced gravitational sorting of Saharan dust during transatlantic voyage: Evidence from CALIOP lidar depolarization measurements. *Geophysical Research Letters* **40**, 3281-3286 (2013).
23. J. B. Stuut *et al.*, Provenance of Present-Day Eolian Dust Collected off NW Africa. *Journal of Geophysical Research-Atmospheres* **110**, (2005).
24. L. Garcia-Carreras *et al.*, The Turbulent Structure and Diurnal Growth of the Saharan Atmospheric Boundary Layer. *Journal of the Atmospheric Sciences* **72**, 693-713 (2015).
25. K. A. Nicoll, R. G. Harrison, Z. Ulanowski, Observations of Saharan dust layer electrification. *Environmental Research Letters* **6**, (2011).
26. J. B. Renard *et al.*, In situ measurements of desert dust particles above the western Mediterranean Sea with the balloon-borne Light Optical Aerosol Counter/sizer (LOAC) during the ChArMEx campaign of summer 2013. *Atmospheric Chemistry and Physics* **18**, 3677-3699 (2018).
27. N. Žagar, G. Skok, J. Tribbia, Climatology of the ITCZ derived from ERA Interim reanalyses. *Journal of Geophysical Research: Atmospheres* **116**, (2011).
28. R. A. Tomas, P. J. Webster, Horizontal and Vertical Structure of Cross-Equatorial Wave Propagation. *Journal of the Atmospheric Sciences* **51**, 1417-1430 (1994).
29. P. Knippertz *et al.*, Dust mobilization and aerosol transport from West Africa to Cape Verde—a meteorological overview of SAMUM- 2. *Tellus B* **63**, 430-447 (2011).
30. F. Esposito *et al.*, The role of the atmospheric electric field in the dust-lifting process. *Geophysical Research Letters* **43**, 5501-5508 (2016).
31. J. F. Kok, N. O. Renno, Enhancement of the emission of mineral dust aerosols by electric forces. *Geophysical Research Letters* **33**, (2006).
32. R. G. Harrison *et al.*, Applications of Electrified Dust and Dust Devil Electrodynamics to Martian Atmospheric Electricity. *Space Science Reviews* **203**, 299-345 (2016).
33. R. G. Harrison, K. A. Nicoll, G. J. Marlton, C. L. Ryder, A. J. Bennett, Saharan dust plume charging observed over the UK. *Environmental Research Letters* **13**, 054018 (2018).
34. W. M. Farrell *et al.*, Electric and magnetic signatures of dust devils from the 2000–2001 MATADOR desert tests. *Journal of Geophysical Research: Planets* **109**, (2004).
35. K. T. Whitby, B. Y. H. Liu, The electrical behaviour of aerosols. In *Aerosol science*, C. N. Davies, Ed. (Academic Press, London, New York, 1966).

36. D. P. Dee *et al.*, The ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Quarterly Journal of the Royal Meteorological Society* **137**, 553-597 (2011).
37. A. H. Fink, D. G. Vincent, P. M. Reiner, P. Speth, Mean State and Wave Disturbances during Phases I, II, and III of GATE Based on ERA-40. *Monthly Weather Review* **132**, 1661-1683 (2004).
38. S. K. Kimball, M. S. Mulekar, A 15-Year Climatology of North Atlantic Tropical Cyclones. Part I: Size Parameters. *Journal of Climate* **17**, 3555-3575 (2004).
39. C. Xu, Y. Ma, K. Yang, C. You, Tibetan Plateau impacts global dust transport in the upper troposphere. *Journal of Climate*, (2018).
40. T. Takemi, Explicit Simulations of Convective-Scale Transport of Mineral Dust in Severe Convective Weather. *Journal of the Meteorological Society of Japan. Ser. II* **83A**, 187-203 (2005).
41. K. Sauter, T. S. L'Ecuyer, Observational evidence for the vertical redistribution and scavenging of Saharan dust by tropical cyclones. *Geophysical Research Letters* **44**, 6421-6430 (2017).
42. M. Peters, G. Tetzlaff, The structure of West African Squall Lines and their environmental moisture budget. *Meteorology and Atmospheric Physics* **39**, 74-84 (1988).
43. D. Goossens, Z. Y. Offer, Wind tunnel and field calibration of six aeolian dust samplers. *Atmospheric Environment* **34**, 1043-1057 (2000).
44. D. Goossens, Z. Offer, G. London, Wind tunnel and field calibration of five aeolian sand traps. *Geomorphology* **35**, 233-252 (2000).
45. M. Van der Does, G. J. A. Brummer, L. F. Korte, J. B. W. Stuut, Seasonality in Saharan dust particle characteristics across the Atlantic Ocean: transport versus deposition. *Aeolian Research*, (in preparation).
46. H. Wernli, H. C. Davies, A lagrangian-based analysis of extratropical cyclones. I: The method and some applications. *Quarterly Journal of the Royal Meteorological Society* **123**, 467-489 (1997).
47. M. Sprenger, H. Wernli, The LAGRANTO Lagrangian analysis tool – version 2.0. *Geoscientific Model Development* **8**, 2569-2586 (2015).
48. C. A. Friese *et al.*, Environmental factors controlling the seasonal variability in particle size distribution of modern Saharan dust deposited off Cape Blanc. *Aeolian Research* **22**, 165-179 (2016).
49. M. Klose, Y. Shao, M. K. Karremann, A. H. Fink, Sahel dust zone and synoptic background. *Geophysical Research Letters* **37**, (2010).
50. E. A. Afiesimama, Annual cycle of the mid-tropospheric easterly jet over West Africa. *Theoretical and Applied Climatology* **90**, 103-111 (2007).

Acknowledgements

This project was funded by the Netherlands Organisation for Scientific Research (NWO, project no. 822.01.008, *TRAFFIC*) and the European Research Council (ERC, project no. 311152, *DUSTTRAFFIC*), both awarded to JBS. The captains, crews, and scientists of RV *Pelagia* cruises 64PE378 and 64PE395, RRS *James Cook* cruise JC134, and NIOZ technicians are thanked for deployment and retrieval of the dust-collecting buoys. PZ is funded through subproject C4 "*Coupling of planetary-scale Rossby wave trains to local extremes in heat waves*" of the Transregional Collaborative Research Center SFB/TRR 165 "*Waves to Weather*", funded by the German Research Foundation (DFG).