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New processes to counteract sedimentation of coarse dust particles are required for climate models to agree with aircraft observations

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Abstract. Recent observations reveal coarser (d > 2.5 µm) mineral dust particles transported unexpectedly great distances from their Saharan source. Transport models represent this coarse dust transport poorly, which has important feedbacks on the Earth's radiative budget, carbon and hydrological cycles, and human health. In this study, we carry out sensitivity tests on the trans-Atlantic transport and deposition processes governing the long-range evolution of mineral dust size distribution in a climate model (HadGEM3-GA7.1). We test the sensitivity of coarser particle transport to sedimentation, convective and turbulent mixing, impaction scavenging, and dust shortwave absorption by removing or scaling the magnitude of these processes. Reductions in sedimentation of ~80% are shown to have the greatest impact on bringing coarse particle transport in the model into better agreement with in-situ aircraft observations. Other tested processes do not result in the multiple orders of magnitude changes in transported coarser dust mass required for agreement with observations. Since sedimentation is a well-understood physical process, we infer that some additional process/es which are misrepresented or not represented at all in the model must be acting to counteract sedimentation. We find that convective and turbulent mixing in the model have minimal impact on coarse particle long-range transport, but are key in controlling the vertical distribution in the Saharan air layer and marine boundary layer, respectively. This study adds to the growing body of evidence that points to processes involved in coarser mineral dust transport and deposition which are not represented accurately or at all in models, which counteract the sedimentation of coarse particles in the real-world. The work in this paper brings the community one step closer to better understanding and modelling of the full dust size distribution and its impacts on weather and climate.

1 Introduction

As the most abundant natural aerosol in the atmosphere by mass, mineral dust has wide-ranging impacts on the Earth system and human activities. Impacting climate, weather, ecosystems (e.g. Kok et al., 2023; Shao et al., 2011), and anthropogenic activities, through altering Earth's radiative budget, carbon and hydrological cycles, human health, aviation, air quality and solar energy. Dust particle size influences the magnitude of these impacts. Fine (d < 2.5 µm), coarse (2.5 < d < 10 µm), super-

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coarse (10 < d < 62.5 µm), and giant (d > 62.5 µm) (size ranges as defined by Adebiyi et al. (2023)) particles vary in terms of mass, mineralogical composition, solubility, and morphology, affecting their interactions with radiation (Adebiyi et al., 2023; Tegen and Lacis, 1996; Adebiyi and Kok, 2020), their impact on biogeochemistry (Marcotte et al., 2020), their efficacy as cloud condensation nuclei (Karydis et al., 2011) and ice nucleating particles (Adebiyi et al., 2023), and their interactions with the human body (Kotsyfakis et al., 2019). Until relatively recently, super-coarse and giant particles were assumed to have short atmospheric lifetimes due to their larger mass and greater settling velocity, and were therefore not frequently measured in observational campaigns. In theory, a spherical dust particle of d = 30 µm should be deposited from 5 km altitude in the Saharan atmospheric boundary layer (SABL) to the surface in ~20 hours (Ryder et al., 2013a). However, recent measurement campaigns have found greater quantities of coarser mineral dust transported for days over hundreds or thousands of kilometres further than expected (Ryder et al., 2019; Weinzierl et al., 2017; Varga et al., 2021; van der Does et al., 2018).

The discovery of longer-range transport of coarse, super-coarse, and giant particles has triggered research exploring how these particles are sustained in the Earth's atmosphere and the consequent impacts of greater mass loading and atmospheric transport. With a greater coarser particle presence, the top-of-atmosphere (TOA) shortwave (SW) effect of dust is less cooling and the TOA longwave (LW) effect is more warming than previously estimated (Adebiyi and Kok, 2020). This results in a total direct radiative effect (DRE) of all dust being less cooling than previously thought at approximately -0.15 $\pm$ 0.35 W m^{-2} (Kok et al., 2023). This value remains uncertain partly due to poor understanding of the quantity and lifetime of coarser dust particles in the atmosphere.

Despite recent advances in our understanding of the coarser size distribution of mineral dust in our atmosphere over long-range transport, climate and forecast models continue to underestimate coarser particle loading and transport (Adebiyi and Kok, 2020). Coarser particles are often disregarded in global climate models (GCMs) due to their assumed short lifetime and additional computational expense, resulting in most models not representing particles greater than 10 or 20 µm in diameter (Zhao et al., 2022; Glиß et al., 2021). Many models also tune the dust so that the magnitude and location of total dust aerosol optical depth (AOD) agrees with satellite retrievals of AOD, however, this is often achieved by predominantly tuning the fine dust fraction. Due to this tuning and lack of or under-representation of coarser particles, a fine bias is created which is evident in many models (Evan et al., 2014; Kok, 2011; Kok et al., 2017), resulting in significant dust mass underestimations. Ryder et al. (2019) suggested dust mass at the Sahara was underestimated by 40% compared to observations through omission of >20 µm particles; this additionally results in underestimations of deposited dust mass downstream (Checa-Garcia et al., 2021). The AeroCom (Aerosol Comparisons between Observations and Models) Phase III initiative found that since AeroCom Phase I, recent iterations of climate models are more likely to represent coarser dust particles, but that the models generally still show a fine size bias (Gliß et al., 2021). The Met Office Unified Model (from which the model configuration used in this paper originates) represents dust particles up to 63.2 µm diameter. However, Ratcliffe et al. (2024) showed that while these coarser particles are emitted in the model, they are deposited too swiftly relative to observations, resulting in an increasing fine bias with distance from the source. Specifically, they found that at the Sahara, there was a mass contribution underestimation by a

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factor of 11, which increased to 240 at Cape Verde, and upon transport to the Caribbean, there were negligible super-coarse particles remaining in the model.

We will now briefly present the mechanisms and processes which are postulated to be involved in coarser particle long-range transport.

Asphericity: Dust particles are typically modelled as spheres, though in reality they tend to be much more aspherical (Otto et al., 2011; Huang et al., 2020). Aspherical particles can experience enhanced drag, increasing particle transport altitude (Colarco et al., 2014; Yang et al., 2013) and lifetime (Mallios et al., 2020; Saxby et al., 2018), Huang et al. (2020) estimate an increase in coarser particle lifetime by ~13%. If the particles were oriented horizontally in the atmosphere, the increase in drag and lifetime would be even greater (Mallios et al., 2020).

Electric charging: Dust particles can undergo charging through collisions with other dust particles, triboelectrification, leading to the whole dust plume carrying observable charge (Savvakis et al., 2024; Harrison et al., 2018; Nicoll et al., 2020). Charged particles may be able to travel higher in the atmosphere and for longer Méndez Harper et al. (2022). Charged particles are shown to be transported higher than non-charged particles in a laboratory setting (Toth III et al., 2020), and charging has been speculated to be a cause of long-range transport in real-world observations (Renard et al., 2018). This process is not considered or represented in GCMs.

Convective mixing: Repeated convective cycling during the day could mix dust particles throughout the depth of the mixed layer after they have settled lower during the nighttime (Gasteiger et al., 2017; Xu et al., 2018). This has been hypothesised to have an impact and tested in modelling of the Saharan air layer (SAL) (Gasteiger et al., 2017) and Tibetan Plateau (Xu et al., 2018). Most GCMs have parameterised convection, though the current move to higher-resolution, storm-resolving modelling (e.g., Slingo et al., 2022; Hohenegger et al., 2020) could result in better representation of dust emissions (Marsham et al., 2011; Roberts et al., 2018), and transport.

Turbulent mixing: Could mix particles up through, and above, the boundary layer, lofting dust away from the surface. Field campaigns have observed updrafts an order of magnitude greater than the settling velocity of coarse 8 µm particles (Denjean et al., 2016). Large-eddy simulations (LES) with resolved turbulence (as opposed to parameterised turbulence in GCMs) have shown improved simulated coarse dust transport representation relative to coarser resolution models (Rodakoviski et al., 2023; Cornwell et al., 2021).

Self-lofting: The SW absorption of dust heats the air around it, creating small turbulent motions, potentially raising the dust higher in the atmosphere (Colarco et al., 2014). Dust plumes could be more susceptible to this process than previously thought due to a) increased presence of more-absorbing coarser particles (Adebiyi and Kok, 2020), b) particle mineralogy being more absorbing than previously thought (Di Biagio et al., 2020; Li et al., 2021), and c) aspherical particles altering the optical properties. The process of self-lofting has been demonstrated for other types of absorbing aerosols such as volcanic ash and biomass burning (Johnson and Haywood, 2023; Khaykin et al., 2022; Boers et al., 2010). While

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self-lofting does appear in GCMs, representations of dust self-lofting are dominantly limited by uncertainty in particle absorption, controlled by the shape, size and mineralogy.

Topography-enhanced lofting: Gentle topography near source can enhance the initial vertical transport of the particles, raising them more quickly towards regions of the atmosphere with larger-scale convective motions. This has been postulated to be the cause of enhanced dust concentrations during a field campaign (Rosenberg et al., 2014), and in satellite observations and reanalysis data (Xu et al., 2018). GCMs, with their grid resolution of O(100 km), may be missing this process by not representing subgrid-scale topography effectively or at all, whereas LES experiments with gentle topography (50-100 m tall x 1 km long) show improved vertical transport of coarser dust particles (Heisel et al., 2021).

A lack in our understanding of these processes means we are still not able to fully explain long-range coarser particle transport. Recent research has looked into reducing the sedimentation or density of dust in models in order to bring the model into agreement with the observations. Drakaki et al. (2022) found that sedimentation of coarse particles had to be reduced by 40-80% in order to be bring an numerical weather prediction (NWP) model into agreement with observations at the Sahara and east Atlantic. Meng et al. (2022) reduced particle density from 2500 kg m^{-3} to as low as 125 kg m^{-3} to improve model coarse size distributions, effectively reducing the sedimentation rate. However, no studies have performed a comprehensive examination of all dust transport processes represented in a climate model.

In this study, we test the sensitivity of different processes governing the transport and deposition of coarser dust particles (sedimentation, impaction scavenging, turbulent and convective mixing, and altered dust SW absorption) in a climate model with a resolution of 1.875° x 1.25°. By doing this, we aim to better understand which processes are most important to the transport and deposition of coarser dust within the model with a view to providing future direction for changes which could be implemented to improve coarser dust transport. In Sect. 2, the methodology of these model experiments will be explained. Sect. 3 will include an overview of the results, which will be discussed together in-depth in Sect. 4. Finally, in Sect. 5, we conclude that sedimentation is the overarching process by which coarser dust transport is governed in the model, with other processes demonstrating much smaller impacts.

2 Methodology

2.1 Model configuration

An atmosphere-only version of the Hadley Centre Global Environment Model 3 (HadGEM3-GA7.1; Walters et al., 2019) is used in this study. The model uses fixed sea surface temperatures (SSTs) but is otherwise free-running. The model configuration has a nominal horizontal resolution of 1.875° × 1.25° (~135 km at mid-latitudes), 85 height levels, of which 29 are below 6 km altitude, and 20 minute time steps. Land surface fields are calculated by the Joint UK Land Environment Simulator land

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surface model (JULES; Walters et al., 2019).

2.1.1 Dust emission scheme

Mineral dust is represented using the Coupled Large-scale Aerosol Simulator for Studies in Climate (CLASSIC) scheme (Woodward, 2001; Woodward et al., 2022). Dust in the model is called at every time step using driving fields from the atmospheric and land surface components of the model (Woodward et al., 2022). Dust emissions are calculated in nine logarithmically-spaced size bins between 0.0632 and 2000 µm diameter. The horizontal (G) and vertical (F) dust fluxes are derived using the method of Marticorena and Bergametti (1995). G is calculated in each size bin (i) using the following,

$$G_{(i)} = \rho B U^{*3}(1+\frac{U^*_{t(i)}}{U^*})(1-(\frac{U^*_{t(i)}}{U^*})^2)\frac{M_{(i)}CD}{g} \tag{1}$$

where ρ is the air density (in kg m^{-3}), B is the bare soil fraction, U^* is the surface layer friction velocity (in m s^{-1}), U^*_t is the threshold friction velocity (in m s^{-1}) from Bagnold (1941) with corrections for soil moisture based on Fecan et al. (1998), M_i is the ratio of dust mass in the size division i to the total mass, C is a constant of proportionality, D is a dimensionless tunable parameter, and g is the acceleration due to gravity(in m s^{-2}). Some of the particles lofted are redeposited to the surface within the same time step and never make it into the model atmosphere. Dust is only transported vertically in the six smallest size bins up to 63.2 µm diameter (Table 1).

Table 1. Size range, representative diameter, and wet scavenging coefficient of the modelled mineral dust size bins in the CLASSIC dust scheme described in Woodward (2001).

Size bin	Diameter range (µm)	Representative diameter (µm)	Scavenging coefficient (Λ)
1	0.0632-0.2	0.112	2×10^{-5}
2	0.2-0.632	0.356	2×10^{-5}
3	0.632-2	1.12	3×10^{-5}
4	2-6.32	3.56	6×10^{-5}
5	6.32-20	11.2	4×10^{-4}
6	20-63.2	35.6	4×10^{-4}

The vertical flux (F) is calculated based on the size distribution of G.

$$F_{(i)} = 10^{(13.4F_c-6.0)}G_{(i)}\frac{\Sigma_{i=1,9}(G_{(i)})}{\Sigma_{i=1,6}(G_{(i)})} \tag{2}$$

where F_c is the soil clay fraction.

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2.1.2 Dust radiative interactions

While in the model atmosphere, dust can interact with radiation in the SW and LW spectral regions. Each dust size bin is treated independently by the radiation scheme, with prescribed values for absorption, scattering, and asymmetry parameter of each size bin in six and nine different spectral bands for the SW and LW, respectively. The refractive indices of dust are based on Balkanski et al. (2007) and are given in the appendix of Bellouin et al. (2011). The particles are assumed to be spherical in the model. By interacting with radiation, the dust can impact atmospheric radiative heating rates, and therefore model meteorology, such as wind and temperature profiles, and subsequently dust emissions themselves. The interaction with radiation can be switched off in the model, allowing for simulations where dust is not able to impact the meteorology.

2.1.3 Atmospheric mixing of dust

Dust is mixed through the atmosphere by (dry and moist) convective mixing (CM) and turbulent mixing (TM). Parameterised convection in HadGEM3-GA7.1 is responsible for both upwards and downwards movements of dust throughout the atmosphere (Lock et al., 2000), and is based on the mass flux scheme of Gregory and Rowntree (1990). Downdrafts and convective momentum transport are also included (Gregory and Allen, 1991). The interaction of CM with dust is not size-dependent in the model. TM is parameterised as a first-order closure (Lock et al., 2000), with additions from Brown et al. (2008) and Lock (2001). TM acts throughout the boundary layer, triggered at the surface by heating and wind shear, as well as at cloud-tops caused by radiative and evaporative cooling. Additionally, TM at the boundary layer top is via an entrainment parameterisation. The effect of TM on dust is size-dependent in the model due to the inclusion of the effects of Brownian motion and inertial effects in the calculation.

2.1.4 Dust deposition processes

Dust is deposited to the surface through impaction scavenging (IS), TM, and gravitational sedimentation. While sedimentation of dust occurs throughout the depth of the atmosphere by Stokes deposition velocity (V_S), dust can experience dry deposition in the two lowest model levels in each time step. Dry deposition velocity (V_D; in m s^{-1}) is represented using an inverse resistance analogue method (Seinfeld and Pandis, 2016)

$$V_D = (R_A + R_B + R_A R_B V_S)^{-1} + V_S \tag{3}$$

where R_A is the aerodynamic resistance (in s m^{-1}) and R_B is the resistance of the quasi-laminar surface layer (in s m^{-1}). R_A is a function of the wind speed, atmospheric stability and surface roughness, and is thus not dependent on particle size. R_B, also known as the surface layer resistance, is the resistance experienced by the particles in the laminar sublayer adjacent to the surface and so is based on molecular scale transport incorporating particle motion by both Brownian diffusion and inertial

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effects. The inclusion of inertial effects results in R_B having a size dependence; this is done by the use of the Stokes number, which is calculated using V_S. V_S is calculated based on a method by Pruppacher and Klett (2010)

$$V_{S(i)} = \frac{{D_{rep(i)}}^2 g \rho_p C_c}{18.0 \mu_{air}} \tag{4}$$

where D_{rep} is the representative diameter (size bin values in Table 1; in µm), ρ_p is the particle density (in g m^{-3}), C_c is the Cunningham correction factor, and μ_{air} is the dynamic viscosity of air (in Pa s^{-1}). C_c has a dependency on D_{rep}.

Wet deposition of dust can occur by IS below-clouds. In the model, wet deposition is composed of two diagnostics: wet deposition by convective precipitation and by large-scale precipitation. Removal by both mechanisms is collated and treated as one in this study, calculated in kg m^{-2} s^{-1}. The rate of impaction scavenging is controlled by a dimensionless scavenging coefficient (Λ), the precipitation rate (R), and dust concentration (C) using the equation

$$\frac{\delta C_{(i)}}{\delta t} = -\Lambda_{(i)} R C_{(i)} \tag{5}$$

The values of Λ for each size bin, i, are shown in Table 1 and generally increase with the particle diameters represented in HadGEM3-GA7.1. These values are based on experimental measurements by Volken and Schumann (1993) and have more recently been corroborated by Laakso et al. (2003). More information on the impaction scavenging scheme is given in Jones et al. (2022).

2.1.5 Tuning

The model has been tuned so that dust emissions from the Sahara are more closely matched with observations from the Fennec airborne field campaign compared to the CMIP6 configuration of HadGEM3-GA7.1. The Fennec campaign measured dust up to 300 µm diameter in the atmosphere over the northwestern Sahara during June 2011 (Ryder et al., 2013b). The tuning of the model is to account for processes not included in the model, such as wind gustiness at the source, and the relationship of soil moisture in the model's top soil level and at the soil surface, as well as for the effect of spatial and temporal averaging onto grid box areas and model time steps (Woodward et al., 2022). In this model setup, there is an increase in emitted coarser volume size distribution which brings the 500-1000 m altitude volume size distribution (VSD) into better agreement with the Fennec observations (Figure S1 in Supplementary Material). There are three tuning parameters which are varied to achieve this: a global multiplier of horizontal dust flux (D, used in equation 1), a friction velocity multiplier (k_1), and a top level soil moisture multiplier (k_2). k_1 and k_2 are used in the calculation of U^* (Woodward et al., 2022). The tuning carried out on this simulation is based purely on the Sahara and trans-Atlantic dust transport, considering no other sources across the globe and is thus only suitable for the purposes of this study. The tuning has increased the VSD of coarser particles and reduced the VSD of fine particles to create an overall shape of the VSD more akin to the Fennec observations. While not a perfect match to the

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observations, the improved VSD at the source will mean our investigation into the loss of coarser material over transport is not affected by a fine bias at emission.

2.2 Model experiments

205 In order to test the sensitivity of coarser dust transport in the model, we have removed or scaled the impact of various relevant modelled processes on dust. Figure 1 shows the relevant processes in the model atmosphere. Two control experiments are used, one with radiative effects (RE) turned off, Control, where comparable experiments have identical meteorology and therefore identical dust emissions, and one with RE turned on (i.e. emissions and atmospheric circulation change between simulations), REControl. Eight simulations have been performed in an RE off setup with changes to transport and deposition mechanisms
210 (experiments listed in Table 2). Only one additional simulation was run with RE on, in which the SW absorption of dust has been tripled. In this study, we analyse monthly averaged simulation data for June of each year, spanning a total of 20 years (1995-2014) for each experiment. June was selected as it is one of the dustiest months in the Sahara and coincides with two of the three aircraft campaigns (Fennec and SALTRACE) used in this study for comparison with the model.

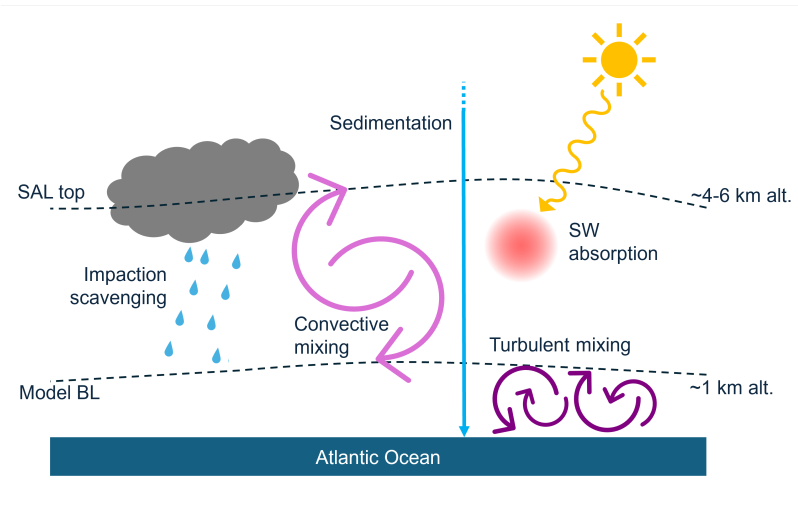

Figure 1. Schematic of the processes affecting dust transport and deposition in the model which have been altered in this study. The two horizontal dashed lines represent the model defined boundary layer (BL) top at ~1 km altitude and the SAL top at ~4-6 km.

215 Firstly, we aim to test reductions in the sedimentation of dust in a HadGEM3-GA7.1 climate model configuration. To alter the sedimentation, Stokes velocity, V_S (in Equation 3), is altered in the UM and JULES code. Unlike other experiments in this study, changes need to be implemented in the JULES code as V_S is used in the calculation of the Stokes number, which is used to calculate surface layer resistance. Four simulations with reductions to the sedimentation of dust are run. One experiment

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Table 2. Sensitivity experiment names, name abbreviations used in the text and whether the experiment has radiative effects turned on or off.

<table>
<tr><th>Simulation name</th><th>Abbreviation</th><th>RE on/off</th></tr>
<tr><td>Radiative effects Off Control</td><td>Control</td><td rowspan="9">Off</td></tr>
<tr><td>No sedimentation</td><td>NoS</td></tr>
<tr><td>95% reduction in sedimentation</td><td>0.05S</td></tr>
<tr><td>80% reduction in sedimentation</td><td>0.2S</td></tr>
<tr><td>50% reduction in sedimentation</td><td>0.5S</td></tr>
<tr><td>No impaction scavenging</td><td>NoIS</td></tr>
<tr><td>No convective mixing</td><td>NoCM</td></tr>
<tr><td>Doubled convective mixing</td><td>2xCM</td></tr>
<tr><td>No turbulent mixing (over the ocean)</td><td>NoTM</td></tr>
<tr><td>Radiative effects On Control</td><td>REControl</td><td rowspan="2">On</td></tr>
<tr><td>Tripled SW absorption</td><td>3xSW</td></tr>
</table>

tests the effect of 'turning off' sedimentation completely (NoS), in this case, V_S is overwritten to equal 0 at all times and locations in the model. In other experiments, the sedimentation is reduced by 50% (0.5S), 80% (0.2S), and 95% (0.05S) (Table 2); in these cases, V_S is simply multiplied by 0.5, 0.2, and 0.05, respectively. These changes are on the order of magnitude of changes made by Drakaki et al. (2022) and Meng et al. (2022) in their experiments testing reduced sedimentation and particle density.

We test the impact of removing CM on dust (NoCM) as well as the impact of doubling the effect of CM (2xCM). We also test the impact of removing TM of dust from the model (NoTM). As TM contributes to the uplift and emission of dust over land in HadGEM3-GA7.1, we employ a grid box land cover mask to keep TM turned on over land, but turned off over the ocean. TM appears in the calculation of the dust deposition velocity (Equation 3) in the form of R_A and R_B, the aerodynamic and quasi-laminar sublayer resistances, and thus we reduce R_A and R_B to 0 to represent a NoTM environment. It is important to note that in neither of these experiments (NoCM and NoTM) are we removing these mixing mechanisms from the model, only their interactions with dust transport and deposition.

One simulation is performed to test the sensitivity of coarser dust to wet deposition in the model. Values of the prescribed scavenging coefficient, Λ (in Equation 5 and Table 1), are set to 0 for all size bins in a 'no impaction scavenging' experiment (NoIS).

In the final sensitivity experiment, we have tripled the SW absorption of dust as a large change to understand if there is any sensitivity to size resolved transport through absorption and SW heating in the model (3xSW). The nature of this change, relating to the RE means this experiment is carried out in a simulation with RE turned on. The original values for the SW mass

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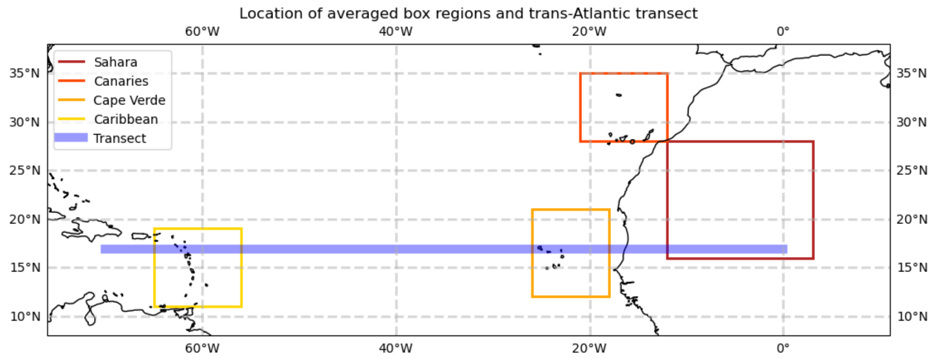

Figure 2. Map showing the location of box regions used for averaging model data in this study, as well as a transect covering the trans-Atlantic dust plume.

absorption coefficient of dust have been tripled at all wavelengths (original and tripled values at 550 nm wavelength shown in Table 3). Scattering mass coefficients remain unchanged. The LW RE and scattering properties remain unchanged. Due to the unchanged scattering properties, the single scattering albedo (SSA; the ratio of scattering to extinction) is lower in the 3xSW experiment compared to REControl, especially at coarser size ranges.

Table 3. Mass absorption efficiency range (in $m^2\ kg^{-1}$) and single scattering albedo (SSA) at 550 nm wavelength including the original values and those used in the 3xSW experiment.

Size bin	Mass absorption efficiency ($m^2\ kg^{-1}$)		SSA	
	Original	3xSW	Original	3xSW
1	16.94	50.83	0.98	0.95
2	28.19	84.56	0.99	0.98
3	29.99	89.97	0.97	0.91
4	21.70	65.09	0.92	0.78
5	15.32	45.96	0.80	0.57
6	8.65	25.96	0.64	0.37

Model data is averaged over box regions related to the location of in-situ airborne campaign data used in this study for evaluation. Additionally, we use a trans-Atlantic transect slicing through the centre of the mean summer dust plume to analyse changes along a consistent latitude. These regions are shown in Figure 2. The box regions correspond to those used in Ratcliffe et al. (2024).

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2.3 Observations

Measurements from three airborne field campaigns between the western Sahara and Caribbean are used to evaluate the models vertically-resolved trans-Atlantic transport and deposition of mineral dust, including the coarse, super-coarse, and giant size ranges. The Fennec campaign provides measurements at the west Sahara and Canary Islands, taken during June 2011 (Ryder et al., 2013b, a). The AERosol properties – Dust (AER-D) campaign measured dust at Cape Verde during August 2015 (Ryder et al., 2018). Finally, the Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE) campaign measured dust at Cape Verde and the Caribbean in June-July 2013 (Weinzierl et al., 2017). Descriptions of the processing carried out on the data are the same as in Ratcliffe et al. (2024) where additional details can be found.

Comparisons between observations and models are complex for many reasons, including differences in both temporal and spatial scales. Previously, Ratcliffe et al. (2024) assessed how representative of ’normal conditions’ these campaigns are and whether any bias in data collection is large enough to skew comparisons, and found that these campaigns are suitable for climatological comparisons. We use 20 June averages from the model in order to create a model average representative of a climatology.

3 Results

3.1 Comparison of modelled size distributions against aircraft observations

Figure 3 shows the mean VSD between 2-4 km altitude in each of the experiments and from the three field campaigns at four locations. First, we compare the Control with the observations; the results indicate a tendency for the model to underestimate coarser mass compared to the observations, particularly in size bin 6 (20-63.2 µm). The extent of the underestimation is worse at locations downwind of dust sources than over the Sahara and is especially depleted (7 orders of magnitude less than observations) at the Caribbean (Figure 3d). This shows that regardless of emissions tuning in the model to improve agreement at the Sahara, a significant problem remains in the model resulting in the overly swift loss of coarser particles over long range transport that is not present in the observations. The same issue was found in Ratcliffe et al. (2024), showing that regardless of the emitted size distribution, the issue of overly swift deposition remains. Figure S2 in the Supplementary Material shows all of these experiments together as their normalised vertical profiles.

At the Sahara, there is little difference between the Control, 2xCM, NoCM and NoTM experiments at any size range (Figure 3a). Compared to the Fennec observations, these experiments have too few size bin 1, 2, 5 and 6 particles (0.0632-0.632 and 6.32-63.2 µm).

The NoIS experiment is the only experiment to have a dominant and large impact on the finer particles, with an increase in fine VSD of more than one order of magnitude at the Sahara. At coarser diameters, there is minimal impact from the removal

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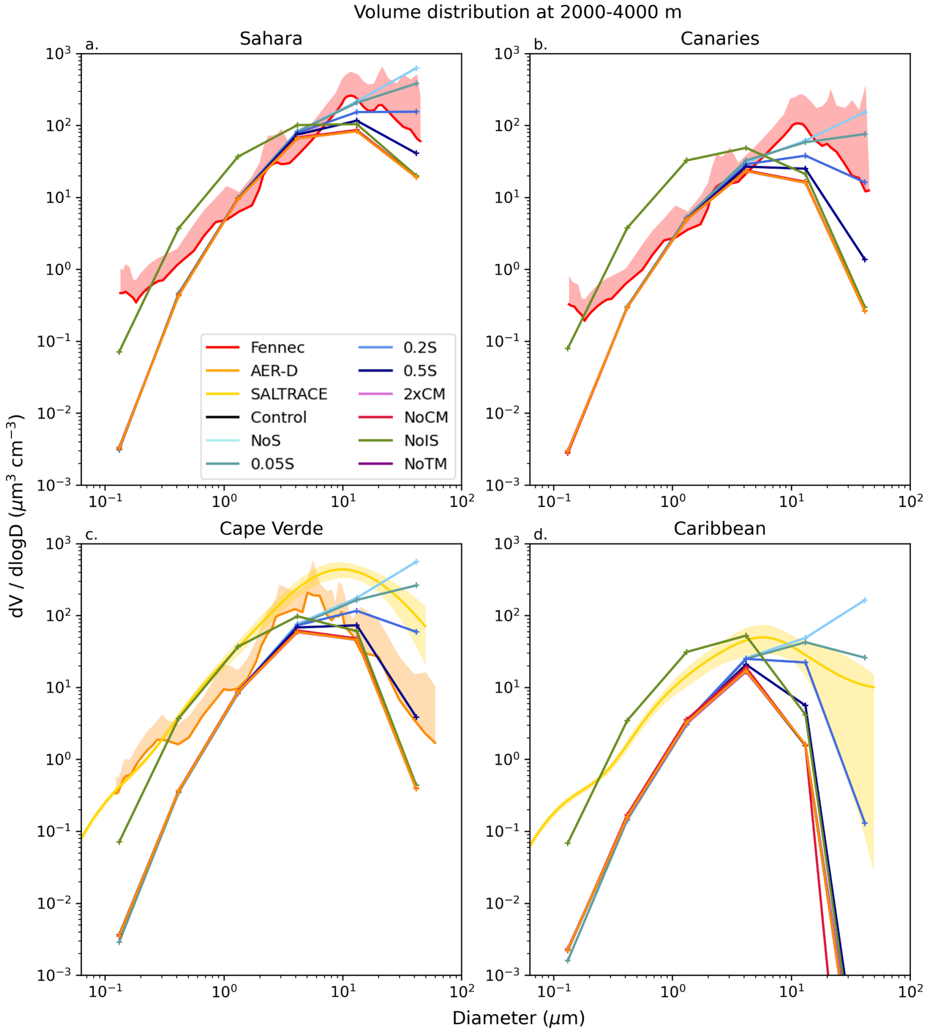

Figure 3. VSD (in μm^3 cm^{-3}) from each model experiment and field campaign averaged between 2-4 km altitude shown at the Sahara (a), Canaries (b), Cape Verde (c) and Caribbean (d). The shading above the Fennec and AER-D campaign VSDs represents one standard deviation. For clarity, the standard deviation shading is only shown above the line due to the logarithmic scale. Shading above and below the SALTRACE VSDs represents the 10th and 90th percentiles of the measurements. The Control simulation terminated at 3.57×10^{-6} μm^3 cm^{-3} in size bin 6 at the Caribbean.

of impaction scavenging. Given that we are analysing a volume distribution, changes at the fine range represent much greater changes to the number distribution due to their smaller size and thus, volume. This increase of one order of magnitude of VSD represents a much greater change to the number concentration of fine particles in the NoIS experiment; the size bin 1 number concentration in the SABL is 2000% greater compared to the Control (not shown) and significantly changed the shape of the total mass profile (Figure S2 in the Supplementary Material). At the Caribbean, the reduction in IS results in improved fine

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particle VSD with respect to the observations compared to the other experiments.

Contrastingly, the sedimentation experiments have the greatest impact on the VSD at coarser size ranges, predominantly impacting size bins 5 and 6. The larger the change to sedimentation, the greater the increase in coarser particle VSD. At the Sahara, the NoS experiment VSD is nearly 2 orders of magnitude greater in size bin 6 than the Control simulation. In terms of the VSD shape at the Sahara, the 0.5S and 0.2S experiments produce VSDs with a shape most similar to the observations, suggesting a 50-80% reduction in sedimentation is required in order to bring the model into better agreement with the observations.

By looking at the sedimentation experiments together, we are given an idea of the coarser dust sensitivity to sedimentation. At the Sahara, all four of the sedimentation experiments and the Control have a size bin 6 VSD within 2 orders of magnitude of each other, however, upon transport to the Caribbean, they are 8 orders of magnitude apart. Without sedimentation, the NoS experiment raises the size bin 6 VSD value by too much at all locations so that the VSD peaks at too coarse diameters. Contrastingly, with 50% sedimentation, the effect of gravitational settling is not large enough to cause a noticeable difference to the size bin 6 VSD from the Control at the Caribbean. The reduction in sedimentation required to bring the shape of the coarser size distribution into agreement with the observations increases with distance from the Sahara; a reduction of slightly more than 50% at the Sahara, 50-80% at the Canaries and Cape Verde, and 80-95% at the Caribbean is required.

As the VSDs of the NoTM, NoCM and 2xCM experiments closely follow the Control experiment, the disparity between these experiments and the observations grow at a similar rate with distance from the Sahara (Figure 3). Except for at the Caribbean, where the NoCM VSD is 2 orders of magnitude lower than the Control experiment in size bin 6. The normalised total mass concentration profile of all REoff experiments shows changes in the vertical dust profile of the experiments compared to the Control (Figure S2 in the Supplementary Material). Figure S2 shows that while some experiments are similar in VSD to the Control, they may have more important changes in the vertical distribution of dust.

The model coarser VSD was only brought into agreement with observations through large change to the sedimentation. Changes to CM and TM did not have strong enough effects on the coarser particles in comparison. Finally, the NoIS experiment showed a dominant impact on the finer size bins. We will now look in more detail at the individual processes and the experiments used to analyse them.

3.2 Sensitivity to Sedimentation Rate

As shown in Figure 3, the sedimentation experiments had the greatest impact on coarser size distribution. With reductions in sedimentation, coarser dust is retained in the model atmosphere for longer, as is shown by the increased VSD at the Caribbean. Figure 4 shows the normalised size bin 6 mass profiles in all sedimentation experiments and the Control with mass centroid altitude (MCA) over the depth of the model atmosphere; the altitude at which 50% of the mass is below and 50% is above (Lu

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et al., 2023).

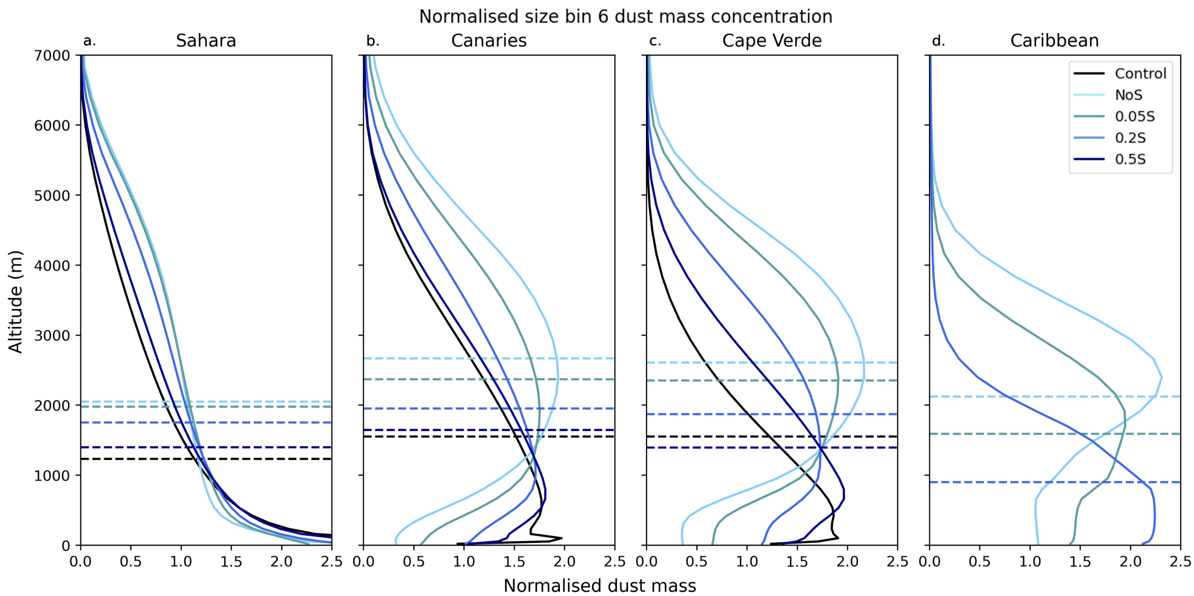

Figure 4. Normalised size bin 6 mass concentration profiles from the sedimentation experiments and Control. Dashed horizontal lines show the size bin 6 MCA over the whole profile. The profiles have been normalised using the size bin 6 mean mass across the whole depth of the profile. Only mass concentrations greater than 0.001 µg m^{-3} are shown.

By first looking at the MCA values, we can see that by decreasing sedimentation, the size bin 6 mass is transported higher within the model atmosphere. At the Sahara, the NoS MCA is more than 0.8 km higher than the Control MCA. At the Canaries and Cape Verde, the NoS MCA is between 1-1.5 km higher. At the Caribbean, a negligible mass of size bin 6 mass remains in the Control and 0.5S experiments.

At the Sahara, the general shape of the size bin 6 profile is not largely different between the five simulations compared here (Figure 4a), though the simulations with a greater reduction in sedimentation have relatively more mass between 1-6 km than the experiments with no or smaller reductions in sedimentation. At the Canaries and Cape Verde, there is a more noticeable difference in the profile shapes (Figure 4bc). In the Control, the mass is more concentrated closer to the surface, with no discernible evidence of the MBL/SAL structure in the dust profile. However, as sedimentation decreases, the MCA rises, creating a profile with more realistic structuring of decreased concentration between 0-1 km associated with the MBL, and a peak in concentration in the SAL between 2-4 km altitude. Ratcliffe et al. (2024) found that the model configuration used in this study has fairly good representation of total dust mass vertical distribution, but transported coarser dust particles too low within the model atmosphere. Here, we show that the greater the reduction in sedimentation, the higher the MCA of the size bin 6 dust mass, suggesting reduced sedimentation can improve the vertical distribution of coarser dust mass over long-range transport in the model.

The sedimentation experiments show that increasing reductions of sedimentation with distance are required in order to bring the model into better agreement with the observations. These results show that a sedimentation velocity reduction of at least 80% is needed to enable the vertical structure and horizontal transport of coarser particles to better match that of observations.

3.3 Sensitivity to Convective Mixing

Figure 5 shows size-resolved transects – at 16.9°N from 70°W (at the Caribbean) to 0°E (at the Sahara) as in Figure 2 – of the absolute difference in dust mass concentration between the NoCM and Control experiments. In size bins 2-4, there is an increase in mass concentration within the SABL and SAL, especially between 1-2 km in the mid- and west-Atlantic. This increase within the SAL is accompanied by a decrease in concentration in the MBL and free troposphere. Without CM the dust is not mixed throughout the depth of or out of the SAL, nor is it mixed downwards from the SAL base towards the MBL. This allows dust to build-up in the lower SAL in the NoCM experiment, depleting the concentration of dust in the MBL at the same time. As the SAL top lowers with westwards transport, the increased concentration of dust transported within the SAL becomes more concentrated, resulting in an enhanced difference to the Control simulation.

The mass concentration distribution in size bin 1 has a more unique reaction to the removal of CM, with an increase in concentration in the MBL and a decrease between 1-2 km altitude between 30-70°W (Figure 5a). This suggests that the size bin 1 particles interact in a different way with the CM processes to the other size bins, the reasons for which are not clear.

In size bins 5 and 6 over land, there is an increase in mass concentration in NoCM up to 4-5 km altitude near the top of the SABL, with a decrease above this. This suggests that similarly to size bins 2-4 in NoCM, the particles are not as efficiently mixed out of the SABL, increasing concentrations within the SABL. These results are somewhat counter-intuitive as we might expect that lack of CM would result in weaker transport of coarse dust mass upwards within the SABL, though the model results in Figure 5 show increased size bin 5 and 6 dust mass over the SABL in the absence of CM. In size bin 6, the mass of particles transported across the Atlantic is so small in both the Control and NoCM experiments that the absolute difference plot shows a negligible difference, matching the negligible change in VSD shown in Figure 3. In size bin 5, the increased concentration of particles within the SABL results in increased dust transported westwards, with slight increases within the SAL. There is an increased mass concentration within the lower SAL as the dust settles above the SAL-MBL inversion.

Removing CM has limited the vertical transport of dust in all size bins to within the SAL, reducing transport into the free atmosphere. Horizontal transport to the north, south, and east of the transect is also reduced (not shown), suggesting decreased mixing of dust out of the dust plume in all directions, increasing the concentration of dust within the SABL/SAL, and thus the mass of dust transported to the the West Atlantic in some size bins. Size bin 6 appears to only have noticeable changes to mass

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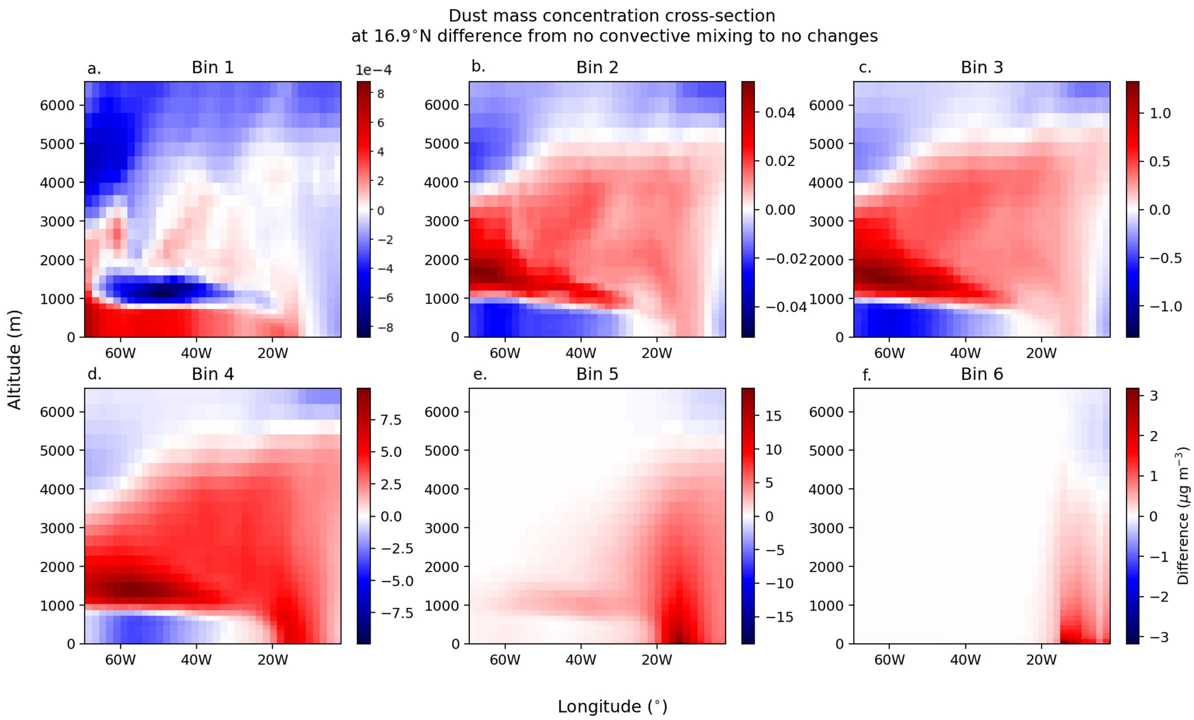

Figure 5. Impact of NoCM experiment: Cross section of difference of the size-resolved mass concentration in the NoCM and Control experiments in µg m^{-3}. At 16.9°N, the cross sections span from 70°W to 0°E, and extend up to 6.5 km. The colour bar scale varies between each plot.

concentration over land; a relative, percentage change shows up to a 10% increase in size bin 6 mass as far as 45°W, though these changes are only to very small mass concentrations of dust (less than 0.001 µg m^{-3}; Figure S3 in the Supplementary Material). The particles in size bins 5 and 6 have too great a mass to fully experience the same effect as occurs to particles in size bins 2-4 whereby they are suspended in the lower SAL above the MBL-SAL inversion. The coarser particles are not dependent on CM to be mixed into the MBL from the lower SAL, instead, their sedimentation is strong enough to overcome transport across the MBL-SAL inversion. This is why we see increased dust concentrations in the western Atlantic MBL for size bin 5. The size bin 5 particles are somewhat retained within the lower SAL, so experience prolonged easterly transport, though eventually settle through the MBL, resulting in an increase in mass concentration relative to the Control in the MBL. In terms of spatial dependence on CM, in all size bins, the greatest increase in concentration occurs at distance from the Sahara, suggesting that the removed CM has a cumulative impact, growing with distance.

Showing that CM plays an important role in the vertical distribution of dust throughout the SAL, we also carried out an experiment with doubled CM. We postulate that if we can increase the amount of mixing in the SAL, then the coarsest particles may be subject to multiple convective events during transport, which could extend particle lifetime. Figure 6 shows cross

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sections in the same way as Figure 5, except this time showing the difference between the 2xCM and Control simulations.

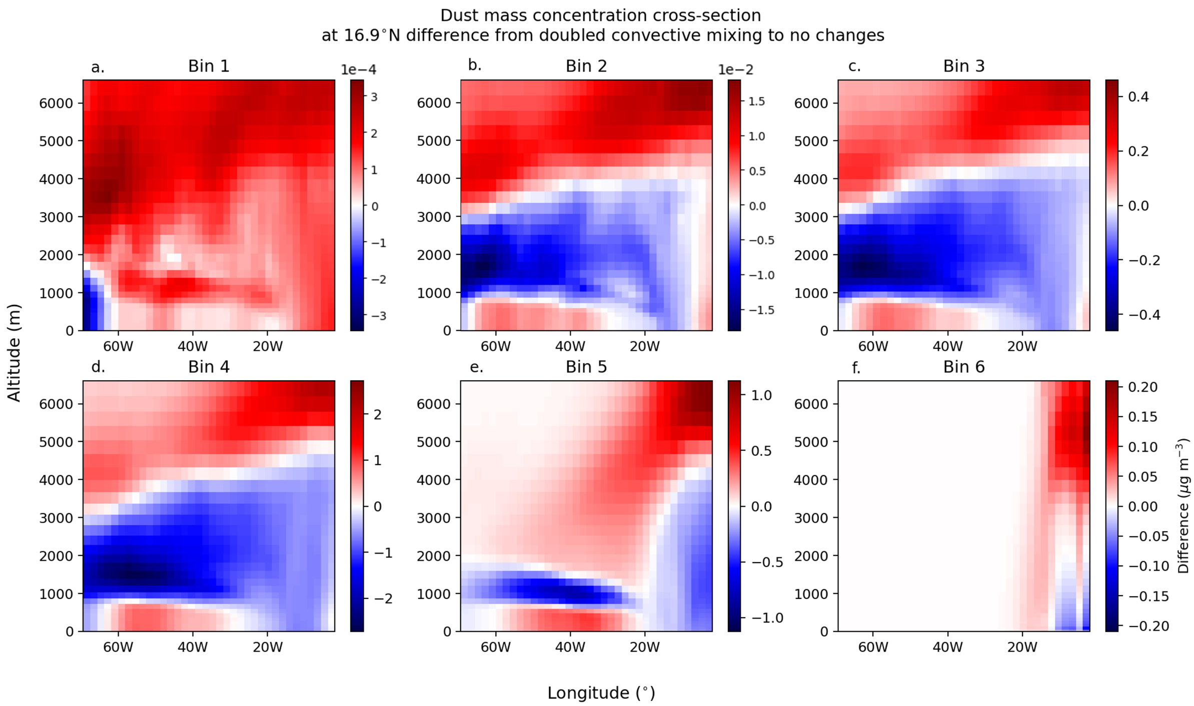

Figure 6. Impact of 2xCM experiment: Cross section of absolute difference of the size-resolved mass concentration in the 2xCM and Control experiments in µg m^{-3}. At 16.9°N, the cross sections span from 70°W to 0°E, and extend up to 6.5 km. The colour bar scale varies between each plot.

For the most part, the 2xCM experiment has the opposite effect on vertical dust distribution to the NoCM experiment; mass concentration is increased in the free troposphere and MBL, and concentration is lower within the SAL in size bins 2-4 (Figure 6bcd). Generally at the Sahara (east of ~17°W on Figure 6), most size bins show decreased concentrations below 4-5 km altitude, with increased dust mixed above this and out of the SABL. As with Figure 5, the greatest changes to size bin 6 only occur over land. We also see the opposite behaviour to NoCM for size bins 5 and 6 in 2xCM over the Sahara, whereby doubled CM results in reduced dust mass within the main dust plume altitudes (beneath 5 km), indicating that these size bins are transported too high vertically in this experiment (also shown in Figure S4 in the Supplementary Material).

All size bins show an increase in mass concentration above the SAL, and size bins 2-5 show an increase in the MBL to 65°W (Figure 6). This shows that with increased CM, dust is more thoroughly mixed throughout and out of the SAL, reducing mass within the SAL, having an opposite effect to the NoCM experiment. The increase of size bin 5 dust in the upper SAL (above 2 km over the Atlantic) is caused by the particles which were mixed above the SABL over the Sahara settling down through

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the inversion at the SAL top due to their greater mass and gravitational sedimentation in comparison to the particles in size bins 2-4 which remain above the SAL.

While the 2xCM experiment does result in additional upwards transport of dust, the extension of mixing is too great, mixing the dust beyond the SAL top, making it an unrealistic simulation, as in reality dust transport is capped by a strong inversion at the top of the SAL. In the NoCM experiment, the lack of CM resulted in dust transported too low within the SAL, resulting in another unrealistic vertical distribution. Although the CM changes investigated here are unrealistic, they provide an insight into the strength of these processes in governing coarser dust transport and have shown that CM is vital for determining the vertical distribution of dust in the model's atmosphere, especially the importance of CM for mixing dust across the temperature inversion separating the SAL and the MBL. This has some effect on the horizontal distribution of the dust too, but the main impact is seen in the vertical changes within the SAL and MBL.

3.4 Sensitivity to Turbulent Mixing

The next mixing mechanism which plays a role in both the transportation and deposition of mineral dust in the model is turbulent mixing (TM). TM was 'turned off' to test its impact, this experiment is called NoTM. However, because TM is heavily responsible for lofting emitted dust into the atmosphere, TM will remain active ('turned on') over land (see Section 2.2 for a more detailed explanation). The NoTM experiment is therefore expected to have little to no change in comparison to the Control at the Sahara.

Figure 7 shows a series of size-resolved cross sections of mass concentration difference between the NoTM and Control simulations. The cross sections are at approximately 16.9°N between 70°W and 0°E.

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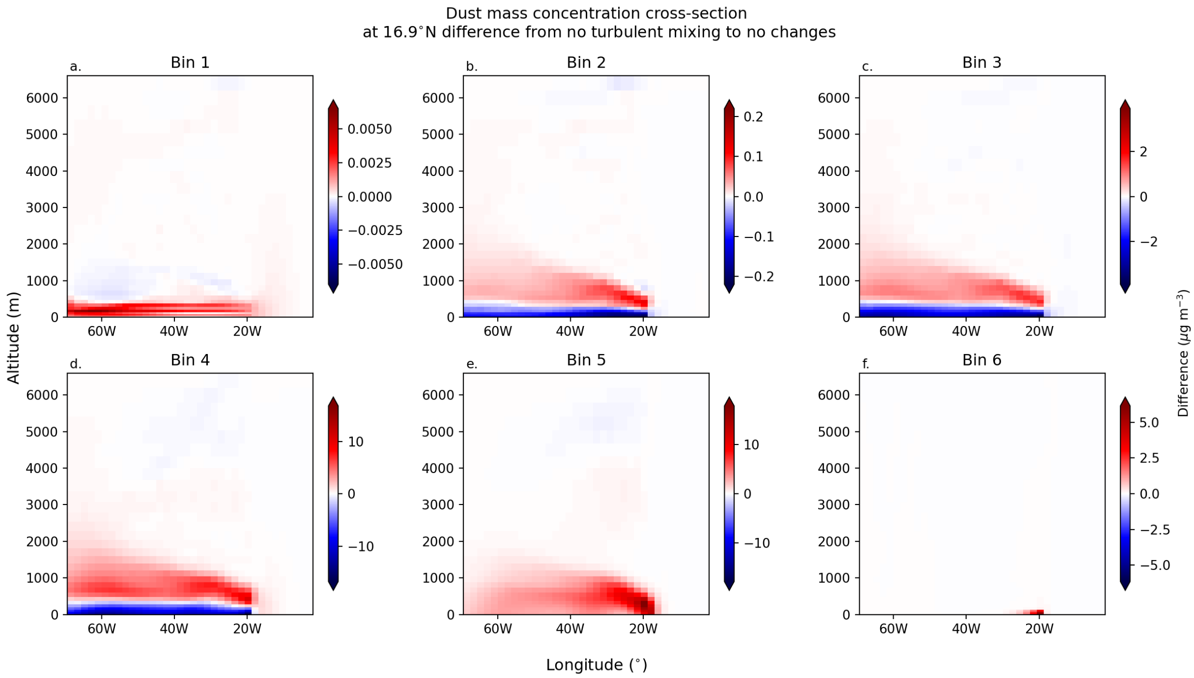

Figure 7. Impact of NoTM: Cross section of absolute difference of the size-resolved mass concentration in the NoTM and Control experiments. At 16.9°N, the cross sections span from 70°W to 0°E, and extend up to 6.5 km. The colour bar scale varies between each plot.

As expected, there is little to no change in mass concentration in any size bin over land (east of ~17°W). Where there are slight changes, for example in size bins 1 and 4, these are likely to due to remixing of dust from over the ocean back over land. Switching off turbulent mixing over the ocean tends to aid the retention of dust between 500 m to 2 km where TM would otherwise have been active in mixing dust down to the surface and very lowest layers of the atmosphere.

Three patterns in the response to removed TM over the ocean are visible in the six size bins. In size bin 1, there is an increase in mass concentration below 500 m altitude of up to 0.006 μg m^{-3}, with a small decrease above this up to ~1300 m. In size bins 2-4, the opposite pattern occurs. Below 500 m, there is a decrease in mass concentration, topped by an increase. Finally, in size bins 5 and 6, there is not the same dipole in mass concentration change. Instead, there is simply an increase in mass concentration which is strongest just off the coast of Africa.

Despite appearing to have different reactions to removed TM, size bins 2-6 all show responses indicative of reduced mixing within the MBL. In size bins 2-4, the dust appears to collect in the upper MBL, and is not mixed down to lower levels as it would be normally. Instead, without the TM, the size bin 2-4 dust remains suspended within the upper MBL. However, the coarser size bins 5 and 6 succumb to their greater gravitational settling velocity, depositing through the lower MBL to the surface. This change in mass vertical distribution in the MBL removes the sharp change in dust mass concentration usually present at the MBL-SAL interface, replacing it with a smooth gradient in concentration from the surface to the MBL-SAL interface (Figure S2 in the Supplementary Material).

Size bin 1 particles are particularly dependent on TM for their deposition due to their small size and subsequently low mass resulting in little dependence on sedimentation. Thus without their main removal mechanism active, the size bin 1 particles concentrate in the lower atmosphere (Figure 7a) with few pathways to atmospheric removal available. This results in widespread, increased concentrations of size bin 1 particles over the North Atlantic (not shown), predominantly relying on wet deposition over land, and TM over land for removal. In removing TM, we reduced R_A and R_B to 0 in equation 3. R_B accounts in part for Brownian diffusion which is most dominant for particles with d < 0.1 μm. This may be why the impact on size bin 1 (0.0632-0.2 μm) is relatively unique to the other size bins.

All size bins show little to no change to mass concentration within the SAL suggesting that TM dominates mixing within the boundary layer towards the surface. With distance from the Sahara, the vertical depth over which increases to mass concentration in size bins 2-5 occur grows upwards into the lower SAL. This suggests that there is some mixing of dust up into the SAL by convective mixing or diffusion from the upper MBL in this NoTM experiment.

The NoTM experiment shows improved coarser particle (mainly size bin 5) transport to the west Atlantic, however, the changes to the vertical profile of dust result in a divergence of the model from the observed vertical dust profile (not shown). Thus, similar to the CM experiments, this experiment provides useful insight into the interaction of TM with dust in this climate model and its impact on vertical mixing within the MBL.

3.5 Sensitivity to Impaction scavenging

Removing IS from the model had the greatest impact at finer size ranges. Without IS, the concentration of fine dust particles increased in the atmosphere, not only in the SAL, but into the free troposphere too. The NoIS experiment changed the vertical profile of total dust mass to the greatest degree out of all the experiments, with large quantities of fine dust transported above the SAL, into the free troposphere (Figure S2 in the Supplementary Material). Figure 3 shows the increased volume size distribution of the four finest size bins, with more than one order of magnitude increase in size bin 1. At the coarser size range, the lack of IS also resulted in an increase of size bin 5 and 6 volume size distribution. At Cape Verde (and the Caribbean), the size bin 5 VSD was 20-100% (100-4000%) greater and the size bin 6 VSD was 8-40% (40-100%) greater than in the Control. While this change is by no means insignificant, it lacks the magnitude required to bring this simulation into agreement with the observations. For example, the 0.2S simulation increased size bin 6 VSD by $1.6\times10^{7}\%$ to bring the model up to the 10th percentile of observations seen during the SALTRACE campaign.

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Despite the IS coefficient increasing roughly with size (as shown in Table 1), the impact of IS decreases with size. This is due to the impact of sedimentation increasing much more rapidly with size than the impact of IS, and thus, the sedimentation is so dominant for the coarsest size bins that changes to IS are negligible in comparison.

3.6 Sensitivity to Tripled SW Absorption

A large degree of uncertainty still exists in the size distribution and mineralogical composition of emitted mineral dust, which impacts the dust radiative effects. Li et al. (2021) and Di Biagio et al. (2020) suggest that the SW absorption of dust is often assumed to be too small and that increasing it can improve model meteorology (Balkanski et al., 2021). In order to test the impact of dust absorption on dust transport, we have tripled the SW absorption of dust in the model to encourage a significant reaction. By doing this, we have to turn on the RE of dust in the model, meaning that this experiment (3xSW) and a new control simulation (REControl) are not parallel to each other or the previous experiments due to variations in model meteorology evolution. Thus, 3xSW and REControl are therefore not as easily comparable to the previous simulations and to each other. Analysis of the emissions from these two simulations over the 20 year modelled period shows that the two simulations have comparable dust emissions (Figure S5 in the Supplementary Material). Thus, while the comparison is not as robust as those carried out previously, the two simulations are assumed to be comparable within this study.

Figure 8 shows the vertically-resolved percentage difference of VSD from Cape Verde to the Caribbean in REControl (a) and the 3xSW (b) experiments, where a negative value indicates loss of particle VSD between Cape Verde and the Caribbean. Figure 8c shows the difference between these differences; i.e. whether the 3xSW experiment retains more particles than the REControl due to the increased SW absorption. Where values are positive, this indicates that the 3xSW experiment retained more particles over the course of long-range transport than in the REControl. Further analysis (not shown here) has shown that the trans-Atlantic dust plume did not change location in terms of latitude significantly between the two experiments, so changes seen in Figure 8 are not a consequence of a shifted plume mass.

In the REControl, Figure 8a shows that all size bins experience a decrease of at least 40% in VSD at all heights between Cape Verde and the Caribbean (except for size bin 1 and 2 particles in the 0-500 m layer). The highest percentage reductions in VSD are observed between 5-6 km altitude in all size bins, likely due to the expected gradual lowering of the SAL top with westwards transport (Carlson, 2016). This model has been shown previously to suffer from overly swift coarser particle deposition, resulting in negligible concentrations of size bin 6 reaching the Caribbean. In Figure 8a, there is nearly 100% loss of size bin 6 particles between Cape Verde and the Caribbean in REControl. In size bin 5, 90-100% of the VSD is lost upon transport to the Caribbean.

In the 3xSW experiment, there is reduced loss of VSD between Cape Verde and the Caribbean relative to REControl. Figure 8c shows the difference between the difference plots in Figure 8 a and b. We can confirm that there is less VSD loss in the

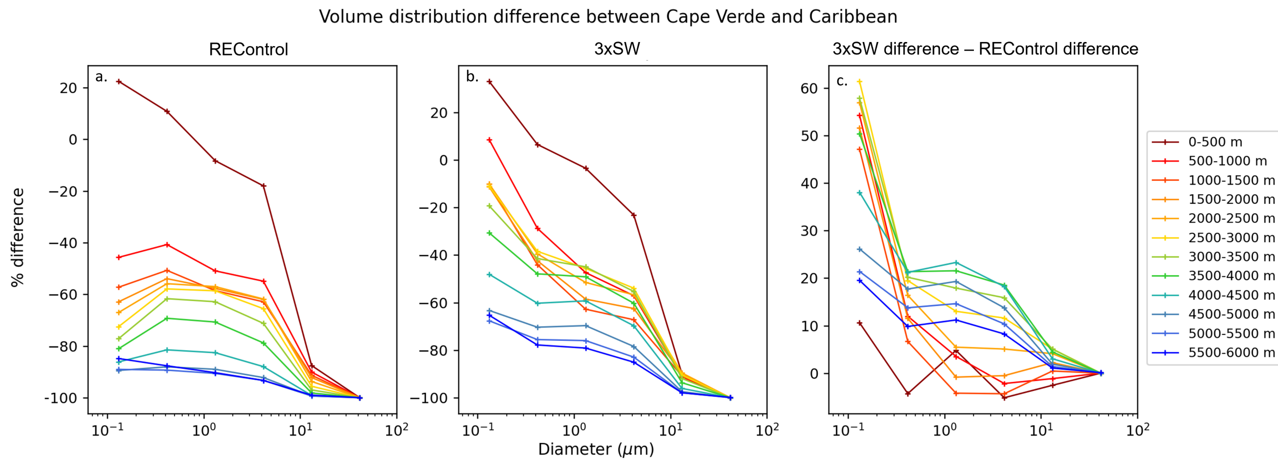

Figure 8. Change of VSD between Cape Verde and the Caribbean in the REControl (a) and the 3xSW (b) experiments. The difference in change (i.e., (a) minus (b)) over transport between the two experiments is also shown (c). In (a) and (b), values of -100% indicate complete trans-Atlantic loss at the indicated diameter.

3xSW experiment compared to the REControl due to the mostly positive values. While the increased SW absorption dominantly impacts the VSD of fine particles, there is a small 1% increase in the VSD of size bin 6 particles reaching the Caribbean at every height level in the 3xSW experiment. There is an increase of size bin 5 particles reaching the Caribbean of up to 5% above 1 km altitude. This increase is not on the order of magnitude required to bring the model into agreement with the observations.

In REControl, VSD loss between Cape Verde and the Caribbean increases with altitude, however, in 3xSW, this is only true in size bin 1 or above 2.5 km altitude in other size bins. This suggests that the dust plume is transported higher in 3xSW compared to REControl. The total dust MCA in 3xSW was found to be higher at the Canaries, Cape Verde, and Caribbean by up to 500 m, suggesting that the tripling of dust SW absorption has had a lofting effect on the dust plume (Figure S6 in the Supplementary Material). Additional analysis showed an increase of the air temperature of up to 1 K at the Sahara, a raising of the MBL-SAL temperature inversion and a heating in the central SAL (3-4 km altitude) of 0.6-0.9 K in the mid-east Atlantic (Figure S7 in the Supplementary Material). These changes in air temperature are likely a direct result of the increased dust SW absorption heating the surrounding air. The strengthening of the temperature inversion at the SAL top reduces erosion of the SAL by the free troposphere, reducing the loss of dust VSD at altitude.

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4 Discussion

In the model, there are various processes which affect and determine the transport and deposition of mineral dust particles (represented in Figure 1). These processes are often size-dependent, impacting the finest and coarsest particles in different ways. In order to dissect which processes most dominantly impact the coarser particles in the model, we have run a series of experiments removing or scaling these processes.

We have shown that sedimentation has dominant control on the long-range transport of coarser particles in this model. Testing of the CM, TM, IS, and dust SW absorption has negligible impact on the long-range transport of coarser particles in the model. Sedimentation is the only process in the model which has both sufficient strength and size dependence that it can be adjusted to bring the model's coarser particle transport into agreement with observations. Reductions in sedimentation by ~80% are required in order to bring the model into better agreement with the observations in terms of the VSD. The improvement of model results following a significant reduction in sedimentation is not unique to HadGEM3-GA7.1. Drakaki et al. (2022) suggested a reduction of 40-80% was required at the Sahara and East Atlantic. Also, Meng et al. (2022) showed that decreasing dust particle density by an order of magnitude, resulting in a reduction of sedimentation velocity by 90-95%, brought their model into agreement with observations in Saharan dust outflow regions. These studies cover varying grid resolution and model configurations; this study used a 135 km resolution global climate model, Drakaki et al. (2022) used a 15 km resolution NWP setup with a higher order advection scheme in the horizontal and vertical, and Meng et al. (2022) used a 190 km resolution global nudged climate simulation. Despite this, the results of this study and those of Drakaki et al. (2022) and Meng et al. (2022) suggesting order of magnitude reductions in settling velocity are remarkably similar and suggest that the model resolution is not the issue (at least in super-km-scale modelling). As sedimentation is a well-understood process, we theorise that the reductions suggested here and elsewhere instead highlight an overarching issue in our understanding of the processes acting on coarser mineral dust particles in the atmosphere that are counteracting their sedimentation by ~80%.

The overarching impact of CM, TM and gravitational settling on vertical movements of dust in the model are shown in a schematic alongside the resulting impact of the NoCM and NoTM experiments (Figure 9). The first panel (Figure 9a) shows an approximation of the ranges in which TM, CM, and sedimentation act within the atmosphere in the Control simulation. TM is mostly dominant in the MBL and CM in the SAL, while sedimentation is present throughout the depth of the atmosphere. In the NoTM experiment (Figure 9b), reduced mixing within the MBL resulted in excessive finer dust mass (size bins 2-4) collecting in the upper MBL, unable to be mixed down to the surface. The continued presence of CM in the SAL in the NoTM experiment, resulted in normal concentrations and vertical distribution of dust within the SAL and free troposphere. Coarser particles (size bins 5 and 6) with their greater mass, were still most influenced by sedimentation, and continued to deposit through the lower MBL to the surface. However, in the NoCM experiment (Figure 9c), the removal of CM had a dominant impact on the vertical distribution within the SAL of the finer dust, resulting in higher concentrations of finer dust collecting in the lower SAL, unable to be mixed down into the MBL. Sedimentation dominates the vertical transport of the coarser particles,

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continuing to mix them across the SAL-MBL inversion downwards towards the surface.

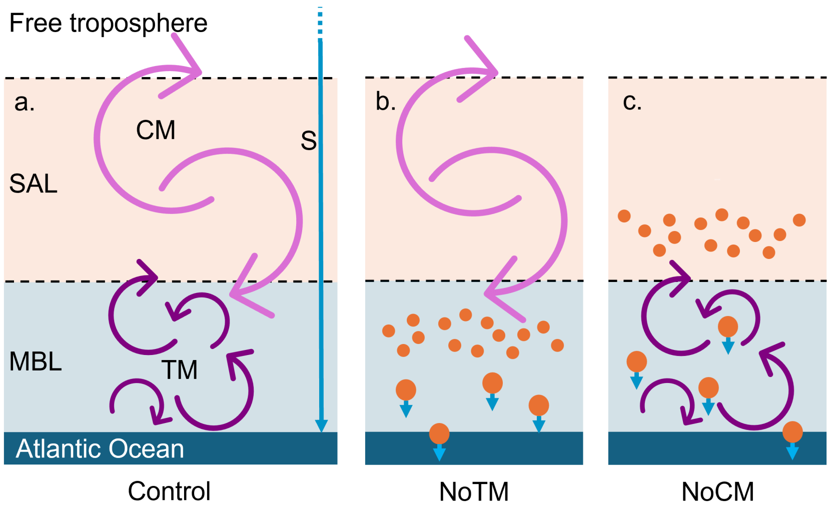

Figure 9. Schematic showing the simplified vertical extent of convective mixing (CM; pink), turbulent mixing (TM; purple), and gravitational sedimentation (S; blue) in the model over the Atlantic Ocean (a). Additionally, the resultant clustering of finer dust (size bins 2-4) in the NoTM (b) and NoCM (c) experiments is shown. The depth of the MBL is approximately 1 km, and the height of the SAL top is between 4-6 km.

While neither the NoTM or NoCM experiments had a great enough impact on the long-range transport of coarser dust mass to reach agreement with observations, we have shown the importance of the mixing mechanisms for maintaining and governing the modelled vertical distribution of dust. The experiments carried out in this paper show that the finer, lighter particles are more subject to CM, TM and IS that the coarser particles, which are dominantly governed by gravitational sedimentation. Rodakoviski et al. (2023) found that a LES of a turbulent shear layer was capable of improving the lifetime of coarse, aspherical dust particles to that which could result in long-range transport. This finding suggests that the representation of TM within a model may be key to better representations of coarser particle long-range transport. Rodakoviski et al. (2023) represent turbulent motions within the SAL, whereas TM in HadGEM3-GA7.1 is represented mainly in the MBL, below the SAL, meaning that the majority of dust in the modelled SAL is not under the influence of turbulent motions. Despite not appearing to have a great impact on the transported coarser particle concentration, CM could have a greater impact in simulations with resolved CM as opposed to the parameterised CM used in HadGEM3-GA7.1. Marsham et al. (2011) showed that parameterised CM greatly underestimated dust uplift compared to storm-resolving simulations. The current move towards higher resolution climate models with improved CM and TM could provide further understanding into coarser particle lifetime.

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5 Conclusions

In this study, we conducted a series of climate model experiments to increase understanding of processes that impact the transport and deposition of coarser mineral dust particles, to provide direction for future improvements in dust transport and climate impacts. A series of 20-year sensitivity tests are run using the HadGEM3-GA7.1 climate configuration from the Met Office Unified Model. These experiments, testing sedimentation, convective mixing (CM), turbulent mixing (TM), impaction scavenging, and dust shortwave (SW) absorption, reveal important information about the role these processes have in determining the transport and distribution of mineral dust particles in the model.

We have shown that sedimentation is the only process capable of making the multi-order of magnitude changes required to bring the model into agreement with observations in terms of coarser particle long-range transport. All other tested processes were found to have minimal impact on coarser particle transport. Reducing sedimentation results in coarser particles being transported further and at higher altitudes. In order to improve the coarser particle size distribution over long-range transport, a reduction of up to 80% reconciles the modelled dust size distribution with observations at the Sahara, but in order to match observations at the Caribbean, a reduction of up to 80-95% was required. Sedimentation is a well-understood process, hence, we theorise an overarching issue in our understanding of the processes acting on coarser mineral dust particles in the atmosphere which is counteracting their sedimentation by ~80%. Processes not considered in the model and/or not well understood in practice may be the root cause of recurring underestimations of coarser particle long-range transport. Processes such as asphericity, electrical charging of particles, and convective and turbulent mixing are suggested to be responsible. Given evidence from previous studies, it may be that all of these processes are responsible to some degree.

Impaction scavenging was found to be more important for the removal of fine particles due to the overarching dominance of sedimentation in controlling the deposition of coarser particles. We found that tripling SW absorption, despite being a large and unrealistic change, did not greatly impact the concentration of coarser particles retained in long-range transport, decreasing the percentage loss of size bin 6 particles between Cape Verde and the Caribbean by only 1%. Instead, we found that by tripling SW absorption of dust, the air temperature was raised by up to 1 K over the Sahara and within the SAL, and that the total dust mass was carried up to 500 m higher in the atmosphere.

TM and CM have been shown here to be important in controlling the vertical distribution of dust in the model marine boundary layer (MBL) and Saharan air layer (SAL). CM controls the vertical distribution of dust within the SAL and the mixing of dust out of it (both above, below and horizontally); without CM, the dust concentration in the lower SAL increases. TM controls the vertical distribution of dust primarily within the MBL, mixing dust downward through the depth of the MBL towards the surface, and governing the deposition of the finer particles to the surface. Without TM, dust collects in the upper MBL, less readily mixed down through the lowest 500 m of the atmosphere. The coarser particles with their larger mass still have great enough sedimentation to be relatively insensitive to these processes being altered and settle through the SAL and MBL regardless.

In this study, we use the HadGEM3-GA7.1 model configuration with a tuned emitted dust size distribution that emits more coarser particles than the standard tuning used in the HadGEM3-GA7.1 CMIP6 setup. We have shown that despite improving the emissions, the divergence of the modelled transported size distribution is exacerbated with transport. Thus, while emissions are an important factor in dust modelling, the transport is clearly creating a bias which cannot be improved with tuning and improved emissions alone.

Through completing a series of sensitivity studies in a climate model, we have shown that counteracting sedimentation is the most efficient way to bring the long-range transport of coarser particles in the model into agreement with in-situ aircraft observations. This work confirms the presence of one or more processes counteracting the sedimentation of coarser particles by a compelling 80-95%. Though we have a better understanding of the magnitude of the counteraction, the nature of these processes remains a mystery. In order to diagnose these processes, we suggest future research into model representations of mechanisms altering coarser particle lifetime, such as asphericity, electrical charging, TM, and tests to better understand how higher grid resolution can change model processes such as TM and CM, which consequently impact dust transport.

Code and data availability. Due to intellectual property rights restrictions, we cannot provide either the source code or documentation papers for the UM. The Met Office Unified Model is available for use under licence. A number of research organisations and national meteorological services use the UM in collaboration with the Met Office to undertake basic atmospheric process research, produce forecasts, develop the UM code, and build and evaluate Earth system models. For further information on how to apply for a licence, see http://www.metoffice.gov.uk/research/modelling-systems/unified-model (last access: 11 September 2025; MetOffice, 2025).

Aircraft data for the Fennec and AER-D campaigns are available at the Center for Environmental Data Archive at https://dx.doi.org/10.5285/1f4555d258 and http://catalogue.ceda.ac.uk/uuid/d7e02c75191a4515a28a208c8a069e70, respectively. Aircraft data for the SALTRACE campaign is currently in the process having a doi created. Data from the model simulations are in the process of having a doi created.

Author contributions. NGR, CLR, NB, SW, AJ, and BJ designed the research, discussed the methodology and results. NGR carried out the analysis and wrote the paper. BW, LMW and JG provided the SALTRACE data. All authors read and commented on the paper.

Competing interests. The authors declare that they have no conflict of interest.

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