- 1 What controls the recent changes in African mineral dust aerosol across the Atlantic?
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9 Abstract

Dust from Africa strongly perturbs the radiative balance over the Atlantic, with emissions that 10 are highly variable from year to year. We show that the aerosol optical depth (AOD) of dust over 11 the mid-Atlantic observed by the AVHRR satellite has decreased by approximately 10% per 12 decade from 1982 - 2008. This downward trend persists through both winter and summer close 13 14 to source and is also observed in dust surface concentration measurements down-wind in Barbados during summer. The GEOS-Chem model, driven with MERRA re-analysis meteorology 15 and using a new dust source activation scheme, reproduces the observed trend and is used to 16 quantify the factors contributing to this trend and the observed variability from 1982 to 2008. 17 We find that changes in dustiness over the East mid-Atlantic are almost entirely mediated by a 18 19 reduction in surface winds over dust source regions in Africa and are not directly linked with 20 changes in land-use or vegetation cover. The global mean all-sky direct radiative effect (DRE) of African dust is -0.18 Wm⁻² at top of atmosphere, accounting for 46% of the global dust total, with 21 a regional DRE of -7.4 \pm 1.5 Wm⁻² at the surface of the mid-Atlantic, varying by over 6.0 Wm⁻² 22 from year to year, with a trend of +1.3Wm⁻² per decade. These large inter-annual changes and 23

the downward trend highlight the importance of climate feedbacks on natural aerosol abundance. Our analysis of the CMIP5 models suggests that the decreases in the indirect anthropogenic aerosol forcing over the North Atlantic in recent decades may be responsible for the observed climate-response in African dust, indicating a potential amplification of anthropogenic aerosol radiative impacts in the Atlantic via natural mineral dust aerosol.

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30 1. Introduction

Mineral dust aerosol is ubiquitous in the atmosphere and arguably the greatest source of 31 particulate matter. Africa is responsible for approximately half of the global emissions (Huneeus 32 et al., 2011) resulting in transport of several hundred teragrams (Tg) of dust across the Atlantic 33 34 towards the Americas throughout the year (Ginoux et al., 2004; Kaufman et al., 2005; Ridley et 35 al., 2012). This has consequences for air quality downwind (Prospero, 1999; Viana et al., 2002) as well as the radiative balance over the Atlantic, via scattering and absorption of solar radiation 36 37 (and to a lesser extent terrestrial radiation), affecting cloud formation (Kaufman et al., 2005; Koren et al., 2010; Twohy et al., 2009) and tropical cyclone formation (Dunion and Velden, 2004; 38 39 Evan et al., 2006). African dust emissions vary greatly from year to year (Ben-Ami et al., 2012; 40 Chiapello, 2005; Ginoux et al., 2004), implying considerable variation in the these impacts on climate and air quality. 41

Global dust emissions vary dramatically on millennial timescales. Sediment core measurements
show that dust deposition over the Atlantic is a factor of 5 higher in the past 2,000 years than
during the African Humid Period (11,700 – 5,000 years ago) and that emissions during glacial
periods are generally 2 - 4 times greater than interglacial periods, likely owing to stronger winds

46 (McGee et al., 2013, 2010). More recently, Mulitza et al. (2010) determined that dust emissions 47 from Africa were negatively correlated with tropical West African precipitation from 1000 B.C. until the end of the 17th century but a sharp increase in dust deposition is observed with the 48 advent of commercial agriculture in the 1800s, indicating the potential for anthropogenic 49 50 changes to influence dust emission. Considerable population growth in Africa over recent decades, by a factor of 3 since 1950 (Prospero and Lamb, 2003), has led to an increase in 51 agricultural activity and urbanization, with speculation that these human-induced land use and 52 53 vegetation changes may also contribute to recent trends in West African dust (Chiapello, 2005; 54 Evan et al., 2011). Since the 1950s, dust emissions from Africa have increased (Evan and Mukhopadhyay, 2010; Mbourou et al., 1997; Prospero et al., 2002), peaking in the 1980s at the 55 56 same time as the extreme droughts experienced in the Sahel region. During this period a robust correlation was observed between dust transported to Barbados in the summer and Soudano – 57 58 Sahel Precipitation Index of the previous year (Prospero and Lamb, 2003). In the past three 59 decades, observations from satellite and surface measurements indicate a breakdown in this relationship (Mahowald et al., 2009), and a decrease in dustiness (Chin et al., 2013; Evan and 60 Mukhopadhyay, 2010; Hsu et al., 2012; Shao et al., 2013; Zhao et al., 2008) coinciding with a 61 greening of the Sahel region (e.g. Olsson et al., 2005). A vegetation-related increase in surface 62 roughness may also have contributed to the stilling of winds (and corresponding reduction in 63 64 dust) over much of the Northern Hemisphere (Vautard et al., 2010; Bichet et al., 2012), including the Sahel region (Cowie et al., 2013). 65

66 Modeling studies generally agree that changes in precipitation over the Sahel, leading to the 67 drought and subsequent greening, can be explained by changes in the inter-hemispheric

temperature gradient across the Atlantic (Chiang and Friedman, 2012; Hwang et al., 2013; 68 69 Rotstayn and Lohmann, 2002; Zhang and Delworth, 2006) which influences the location of the 70 Inter-Tropical Convergence Zone (ITCZ). Changes in dust emissions and transport over the 71 Atlantic are associated with the location of the ITCZ (Doherty et al., 2012; Fontaine et al., 2011) 72 which may provide a feedback by modulating the radiative balance over the Atlantic (Evan et al., 2011). However, whether the observed changes in dust are a direct consequence of the large-73 scale changes associated with the ITCZ location or a consequence of the greening of the Sahel, 74 75 either via a reduction in available dust sources or a stilling of the surface winds, is still unclear.

Dust outflow from Africa is somewhat correlated with the North Atlantic Oscillation (NAO) based 76 on comparison with observations (Moulin et al., 1997; Chiapello, 2005; Nakamae and Shiotani, 77 78 2013) with the strongest relationship north of 15°N (Chiapello and Moulin, 2002). The NAO index, defined by the difference in normalized sea level pressure between the Icelandic low and Azores 79 80 high (Hurrell, 1995), is extremely noisy and our understanding of what causes the fluctuations is 81 limited (Stephenson et al., 2000). While the NAO index represents changes in circulation that 82 affect dust transport, primarily via the Azores high rather than Icelandic low (Riemer et al., 2006), the correlation with observations of dust close to source regions is weak (Nakamae and Shiotani, 83 2013). Therefore, we choose to focus primarily on the physical processes the drive dust emission 84 85 and export (e.g. wind strength and precipitation) rather than the climate indices that may 86 represent them.

In this paper we use surface and satellite dust observations along with a 27 year model simulation
to quantify the driving factors behind the variability and trends in dust loading over the Atlantic,
the importance of vegetation changes for dust emission, and whether the underlying causes are

natural or anthropogenic in origin. The magnitude of modeled and observed variability and
trends in Atlantic dust AOD and direct radiative effect (DRE) are assessed and the causes
quantified using the model. Finally, we discuss a potential driver of changes in surface winds using
three reanalysis datasets and fifteen CMIP5 model simulations.

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95 2. Model description and evaluation

96 2.1 Baseline model description

The GEOS-Chem model (version v9-01-01; http://www.geos-chem.org/) incorporates a global 97 three-dimensional simulation of coupled oxidant-aerosol chemistry, run at a resolution of 2° x 98 2.5° latitude and longitude, and 47 vertical levels in this study. The model is driven by assimilated 99 100 MERRA meteorology for 1982-2008 from the Goddard Earth Observing System of the NASA 101 Global Modeling and Assimilation Office (GMAO), which includes assimilated meteorological 102 fields at 1-hourly and 3-hourly temporal resolution. The aerosol types simulated include mineral dust (Fairlie et al., 2007; Zender et al., 2003), sea salt (Alexander et al., 2005), sulfate-nitrate-103 104 ammonium aerosols (Park et al., 2004), and carbonaceous aerosols (Henze et al., 2008; Liao et 105 al., 2007; Park et al., 2003). Aerosol optical depth (AOD) is calculated online assuming lognormal 106 size distributions of externally mixed aerosols and is a function of the local relative humidity to account for hygroscopic growth (Martin et al., 2003). Aerosol optical properties employed here 107 108 are based on the Global Aerosol Data Set (GADS) (Kopke et al., 1997) with modifications to the size distribution based on field observations (Drury et al., 2010; Jaegle et al., 2010; Ridley et al., 109 110 2012) and to the refractive index of dust (Sinyuk et al., 2003).

111 For all long-term simulations in this study the standard model is modified to include dust aerosol 112 only. Emission, dry deposition and wet scavenging of dust are all simulated as in the standard 113 model (Fairlie et al., 2007). Dust emission in GEOS-Chem is based upon the DEAD dust scheme 114 (Zender et al., 2003), making use of the GOCART source function (Ginoux et al., 2001) as proposed 115 by Fairlie et al. (2007), based on evaluation of dust concentrations over the US. Mineral dust mass is transported in four size bins (0.1 - 1.0, 1.0 - 1.8, 1.8 - 3.0 and 3.0 - 6.0 μ m), the smallest of which 116 is partitioned into four bins (0.10 - 0.18, 0.18 - 0.30, 0.30 - 0.65 and 0.65 - 1.00 μm) when deriving 117 118 optical properties, owing to the strong size-dependence of extinction for sub-micron aerosol 119 (Ridley et al., 2012).

The GEOS-Chem model has been coupled with RRTMG, a rapid radiative transfer model (Mlawer et al., 1997), to quantify the DRE of aerosol species online (Heald et al., 2013). In this version of the model we calculate the shortwave (SW) and longwave (LW) radiative effects for dust based on fluxes at 30 wavelengths under both clear-sky and all-sky conditions.

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125 2.2 Dust emission scheme updates

Two significant changes have been made to the dust scheme relative to the standard GEOS-Chem implementation described thus far. First, we account for sub-grid variability in surface winds and, second, we alter the dust source function to depend on geomorphology and include dynamic vegetation cover. We represent the sub-grid winds as a Weibull probability density function based on statistics derived from the native resolution of the assimilated meteorology wind fields (0.5° x 0.67°). We previously showed that accounting for the sub-grid wind distribution reduces 132 the resolution-dependence of dust emissions in the model by better representing situations 133 when the mean wind speed is approaching the threshold for dust activation (Ridley et al., 2013). 134 To investigate the role of vegetation changes in modulating dust emissions we must simulate dust emission from semi-arid regions. Compared to more recent dust source region studies 135 136 (Ginoux et al., 2012; Koven and Fung, 2008; Schepanski et al., 2007) the original GOCART dust 137 source derived from TOMS aerosol index is likely to underestimate emissions from regions that are not permanent deserts. Satellite-derived dust source data sets often disagree as a result of 138 cloud cover and temporal sampling, particularly in regions where emissions show distinct diurnal 139 140 patterns, such as West Africa (Schepanski et al., 2012). For this reason we implement the geomorphic dust source map of Koven et al. (2008), derived from surface roughness and 141 142 levelness properties, and let vegetation cover attenuate the source strength. We follow Kim et al. (2013) in deriving the bareness fraction from AVHRR Normalized Difference Vegetation Index 143 144 (NDVI) for each year (values <0.15 are considered bare and a potential dust source) to modulate 145 dust emissions from 1982-2008. The updated dust scheme performs at least as well as the original dust scheme when assessed relative to MODIS and AERONET observations over Africa 146 147 and the mid-Atlantic (see supplementary material). The relatively small change in agreement 148 even with substantial changes in source regions suggests that the wind fields dominate the 149 agreement with observations, both in terms of the surface wind strength leading to emissions 150 and the large-scale transport across the continent. Achieving a higher fidelity dust simulation 151 therefore appears to rely more on an improvement of the wind fields than the characterization 152 of the surface properties.

154 **2.3 Regional direct radiative effect (DRE)**

155 Using the updated GEOS-Chem model coupled with RRTMG we quantify the change in radiative flux at the surface resulting from the changes in dust aerosol over the 27 year period (for all-sky 156 157 conditions). Figure 1 shows the average seasonal DRE of dust over Africa and the Atlantic for the 158 whole 27 year period, both at surface and TOA. We define the radiative effect as an increase in down-welling flux and therefore a negative value constitutes a cooling of the Earth. Globally, the 159 160 all-sky radiative effect from African dust is -0.18 Wm⁻² at TOA, accounting for 46% of the total 161 global dust DRE. Across the mid-Atlantic (5 - 20°N, 10 - 50°W) the mean annual radiative effect of dust is -3.2 ± 0.7 Wm⁻² at TOA and -7.4 ± 1.5 Wm⁻² at the surface, including LW contributions 162 of +0.4 ± 0.1 Wm⁻² and +3.1 ± 0.7 Wm⁻², respectively. This region covers less than 5% of the Earth's 163 164 surface but accounts for almost 20% of the global dust radiative effect at TOA and at the surface. The difference between the TOA and surface radiative effects indicates the heating of the 165 166 atmosphere owing to dust. When the dust outflow is over the Sahara (primarily during summer) the airborne dust can be darker than the surface beneath, decreasing the amount of outgoing 167 radiation and producing a warming effect at TOA. 168

While direct comparison with previous estimates is difficult owing to different time periods, conditions and model assumptions, we find the spatial distribution and magnitude of the DRE is broadly consistent with previous modeling studies (Evan and Mukhopadhyay, 2010; Miller et al., 2004; Yoshioka et al., 2007) and observations (Haywood et al., 2003; Highwood, 2003; Hsu et al., 2000). Aerosol size, shape refractive index, altitude, and surface albedo all contribute to the uncertainty in the radiative effect and lead to considerable diversity between models and observations. Several studies have attempted to quantify the key factors leading to uncertainty in radiative effect and radiative forcing (e.g. Balkanski et al., 2007; Evan et al., 2009; Miller et al.,
2004; Myhre et al., 2013; Stier et al., 2013) but this is certainly an area requiring further research
to better constrain model estimates.

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180 **3. Observations**

The primary long-term observations used in this study is a dust AOD (DAOD) dataset over the 181 mid-Atlantic for the period 1982 - 2008, derived from satellite observations by Evan and 182 183 Mukhopadhyay, (2010). Satellite retrievals of AOD from the AVHRR PATMOS-x dataset, an 184 extended and recalibrated retrieval using the moderate resolution imaging spectro-radiometer (MODIS) observations (Zhao et al., 2008), are converted to DAOD using MODIS AOD and fine 185 186 mode fraction products and with NCEP-NCAR reanalysis surface winds, following Kaufman et al. 187 (2005). The 1° x 1° DAOD product is only available over the ocean, between 0 - 30°N and 65 -10°W, therefore we select three 7.5° by 10° regions representing outflow towards North America, 188 189 the Caribbean, and South America in which to compare model and observations (see Fig. 2). We also consider a 10° by 17.5° region off the coast of Africa to assess outflow close to source; initially 190 191 this region was divided into two, north and south of Cape Verde; however, the results are largely 192 the same when considering the region as a whole.

Trade-wind aerosol has been measured almost continually at the Ragged Point site on the east coast of Barbados since 1965 (Prospero and Lamb, 2003). During conditions when the on shore wind exceeds 1 ms⁻¹ (~95% of the time, Savoie et al., 1989) air is drawn through a filter upon a 20m tower, the soluble material is removed and the remaining mineral residue (representing the dust aerosol) weighed after ashing. We use monthly average dust concentrations for the period
1982 – 2008 to evaluate long range dust transport from Africa.

Rather than the standard Boreal seasonal classification (DJF and JJA) we follow the work of Ben-Ami et al. (2012) who show that African dust seasonality can be broken down into three seasons: December to March, April to mid-October, and October through November. Throughout this study we compare the modeled and observed DAOD and surface concentrations either annually or for two seasons, referred to as winter (DJFM) and summer (AMJJAS). The October-November period is characterized by low dust emission and is not shown separately, but included in the annual averages.

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207 4. Dust Transport and Trends

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4.1 Trends and variability in dustiness and the radiative effect

210 To assess how the dust loading over the mid-Atlantic has changed since the 1980s and whether the model captures the observed variability we consider the DAOD derived from satellite data in 211 212 regions close to source and further downwind. Figure 3 shows the seasonally-averaged DAOD for 213 the model and derived from satellite observations in each of the outflow regions indicated in Fig. 214 2 from 1982 to 2008. The DAOD is displayed as an anomaly from the climatological average for 215 the winter and summer seasons. Close to source, the model captures 50 - 80% of the variance in the observations in the winter 216 217 months (monthly correlations between 0.69 and 0.90). The seasonal correlation with the

observations is fair during summer (r = 0.64) with the model struggling to represent the variability

219 and magnitude of emissions between June and August; likely a consequence of underestimated 220 AOD at the Bodélé Depression and across the Sahel during summer (see Supplementary Fig. S2). Downwind, correlations between model and observations range between 0.46 and 0.65 221 222 (excluding the North America region during winter when very little dust is present). The poorer 223 agreement downwind indicates that model transport and removal contribute to the discrepancy 224 with observations. The variability in the model DAOD is generally less than observed, owing to lower model DAOD; however, the normalized variability (normalizing using the climatological 225 mean DAOD) is comparable between model and observations. Previous studies have shown that 226 227 the model removes dust aerosol too rapidly relative to the observations via both wet and dry deposition (Generoso et al., 2008; Ridley et al., 2012) and this is likely to be contributing to the 228 229 reduced variability in DAOD at these downwind regions. Overall we see best agreement for 230 seasons with the highest dust loading, i.e. winter at the South America region and summer in the 231 Caribbean and North Atlantic regions.

Significant decreasing trends in observed DAOD (>95% confidence) are apparent for all seasons 232 and locations, except winter in the North America and Caribbean regions when northerly 233 transport of dust is limited and highly variable, respectively. There are striking similarities 234 between the trends in observed and modeled DAOD, with the model showing significant trends 235 236 at the same locations and during the same seasons. Annually, observed and modeled DAOD 237 decreases between -0.035 (12%) and -0.032 (17%) per decade close to source and -0.021 (12%) to -0.016 (28%) per decade downwind, respectively. Previous studies show similar decreases of 238 239 total AOD per decade in the mid-Atlantic based on the AVHRR PATMOS-x dataset (Mishchenko 240 and Geogdzhayev, 2007; Zhao et al., 2008) and SeaWIFs observations from 1997 – 2010 (Hsu et al., 2012). This suggests that the trends in total AOD over the mid-Atlantic are driven almostentirely by the changes in dust aerosol.

There is significant interannual variability in dust concentration during the spring months (March 243 244 - May), a period responsible for transport of dust to South America based on both satellite and 245 in-situ measurements (Kaufman et al., 2005; Prospero et al., 1981, submitted). Isolating the spring season we find significant trends in DAOD of -0.02 per decade for both observations and 246 model in the South America region (r=0.70) indicating that the decreasing dust trends are present 247 248 and captured by the model in this season as well as the broader seasons considered in this study. 249 Figure 4 shows the anomaly in monthly dust concentration measured at Barbados alongside the modeled surface concentration anomaly. We find that seasonal correlation between the dust 250 251 surface concentration measured at Barbados and simulated concentrations show similar agreement as the DAOD comparisons, with r = 0.69 and r = 0.44 during winter and summer, 252 253 respectively. Surface concentration of dust measured at Barbados has been decreasing during the summer (-3.5 \pm 1.3 μ gm⁻³ per decade); this trend is reproduced by the model (-5.2 \pm 1.1 μ gm⁻ 254 ³ per decade). No trend is present in either observations or model during the winter, consistent 255 with the Caribbean regional DAOD (Fig. 3). 256

The consistency and geographical extent of the downward trends in both the modeled and observed dust suggest that the model simulates the process driving these trends in dust throughout this multi-decadal period.

The inset charts within Fig. 1 show the modeled time series of seasonal DRE for the region 0°N -30°N, 50°W - 15°E, encompassing the mid-Atlantic and West Africa. We note considerable variability in the radiative effect from year to year, primarily in winter when emissions are more

sporadic in the model, confirmed by observations (Ben-Ami et al., 2012). The regional surface 263 cooling varies by up to 8 Wm⁻² in winter and 6 Wm⁻² in summer, and a more modest 4 Wm⁻² and 264 1.5 Wm⁻² at TOA in winter and summer, respectively. Annually, warming trends of +1.27 Wm⁻² 265 and +0.37 Wm⁻² per decade are observed at the surface and TOA, respectively, over the region 266 267 including both the ocean and land from 1982 to 2008. The seasonal trends are similar in magnitude, with significant trends in both winter and summer (95% confidence). Similar trends 268 in DRE exist over both ocean and land at the surface (+1.23 Wm⁻² and +1.32 Wm⁻² per decade, 269 270 respectively) but at TOA there are marked differences between the trends over ocean and land 271 (+0.60 Wm⁻² and +0.00 Wm⁻² per decade) as a result of surface albedo and the high concentration of large dust particles (increasing the LW warming effect). The regional trend constitutes an 272 273 increase in DRE over the past three decades that is comparable in magnitude to the regional increase in CO₂ forcing since 1750 (IPCC, 2013). This illustrates the strong radiative perturbation 274 275 potential of dust: climatic changes that are affecting the emission of African dust are likely to 276 have significant impacts upon the radiative balance over the Atlantic that are not accounted for 277 in the traditional radiative forcing metric.

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4.2 Attribution of variability and trends in dustiness

To attribute the driving forces behind the interannual variability in African dust near source and downwind, four 27 year simulations are performed with interannual variability removed from surface winds, transport, precipitation, or vegetation. This method also removes any inter-annual trend in that variable. The interannual variability of 10-m surface winds are removed by using the 1988 10-m winds for every year, 1988 being an 'average' year in terms of dust emissions (N.B. this only affects the 10-m winds used for calculation of dust emission flux, not for other processes
in the model). This process is repeated by holding vegetation constant at 1988 values or by fixing
precipitation and 3-D winds (other than 10-m surface winds) to 1988 values for the 27 year
period. Here we are investigating only the direct impact of vegetation cover reducing available
surface for dust emission and do not take into account secondary effects such as stilling of winds
from surface roughness changes, discussed further below.

The variability caused by each of the three factors is inferred by assessing the reduction in the 291 292 variance of the DAOD in each region relative to the original model. This method does not account 293 for confounding factors resulting from the variables being dependent (i.e. the large scale winds and 10-m wind strength will be well correlated) but results are found to be robust (within ± 5%) 294 295 when removing the interannual variability using data from 2004 instead of 1988. Figure 5 shows 296 the apportionment of variability in DAOD to the three factors tested for summer and winter. In 297 the coastal Africa and South America regions the 10-m surface wind accounts for at least two thirds of the variability in both seasons. In North America and the Caribbean surface winds 298 299 account for one third to half of the variability in the DAOD, interannual variability in large-scale transport being more important during the winter. Precipitation accounts for only a small fraction 300 of the variability in DAOD in all regions during the winter, but becomes more important 301 302 downwind during the summer. Within the model, precipitation primarily affects the variability in 303 dust loading over the Atlantic via wet scavenging rather than by increasing soil wetness and suppressing emission. Removing the interannual variability of vegetation has a negligible impact 304 on the variability in DAOD suggesting that the changes in dust source region resulting from 305 306 vegetation cover changes are unimportant for the observed variability in dust since the 1980s.

307 The same four simulations described above are used to assess the cause of trends in DAOD. 308 Figure 6 shows the resulting annual DAOD anomaly in each region for each simulation. In the 309 Coastal Africa region it is clear that removing interannual variability in 10-m winds almost entirely removes the trend in DAOD. Further downwind the interannual variability in other meteorology 310 311 contributes between 30 – 50% of the trend; however, the surface wind at source remains the 312 dominant driver. This indicates that, in the model, the trend in dustiness results from a stilling of surface winds over source regions and combines with changes in transport and/or an increase in 313 314 removal downwind, more so in the more northerly outflow regions. We find that the direct effect 315 of vegetation changes on dust emission in the model has a negligible impact on the trend in 316 dustiness over the Atlantic. In all locations except North America the trend with no interannual 317 variability in 10-m surface winds is significantly different to the baseline run (>95% confidence), whereas the trend with no interannual variability in vegetation is indistinguishable from the 318 319 baseline. Although surface winds have been inferred as the likely cause of the observed reduction 320 in Atlantic dust loading (Chin et al., 2013) this is the first time that the link has been quantified. 321 While we do find a positive correlation between the NAO index and the DAOD in the outflow 322 regions during winter, we find no significant trend in the NAO index between 1982 and 2008, 323 therefore the question remains as to what is driving the stilling of winds over Africa. We have 324 shown that changes in vegetation are unlikely to *directly* influence dust emission via changes in 325 source regions but they may still indirectly affect the emissions via stilling of the winds. If 326 vegetation changes are driving the decrease in winds responsible for the change in dustiness over 327 the Atlantic then any increase in vegetation cover is expected to be coincident with the decrease 328 in winds (Bichet et al., 2012).

Figure 7a shows the change in the bareness fraction (a reduction in bareness fraction indicating 329 330 a 'greening') derived from AVHRR between 1982-1986 and 2002-2006 for summer and winter. Figure 7b shows the change in surface winds apparent in the MERRA reanalysis and the regions 331 in which dust emissions decrease by more than 0.5 Tg per grid box. For both seasons we see that 332 333 there is a limited amount of overlap between the location in which vegetation increases and the 334 surface winds (and therefore emissions) decrease. This is not necessarily in disagreement with the conclusions of Cowie et al. (2013); there the focus is on local Sahelian emissions only. Events 335 when dust is transported from elsewhere are excluded and account for between 50% and 90% 336 of all dust events at the Sahel weather stations (personal comm. Sophie Cowie). The reanalysis 337 winds are unlikely to capture the full extent of wind stilling from surface roughness changes and 338 339 therefore may be missing trends in winds and dust emission in the Sahel. However, the model still captures the decreasing trends in dust over the Atlantic, suggesting that emissions from 340 341 regions other than the Sahel are controlling the trends in Atlantic DAOD. The lack of spatial 342 correspondence between the greening and the wind stilling indicate that vegetation is not driving the change in model surface winds though perhaps both are the result of a larger-scale climatic 343 change. Indeed, shifts in dustiness over the past 20,000 years also suggest that large changes in 344 345 dust emission in the past are primarily driven by changes in large-scale winds, rather than vegetation and precipitation changes (McGee et al., 2010). 346

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348 **4.3 Reliability of surface wind trends in reanalyses**

To assess the reliability of the MERRA surface winds in this relatively observation-poor region we compare annual MERRA reanalysis wind trends between 1982 and 2008 with those from the 351 NCEP and ERA-Interim reanalysis products. Figure 8 shows the annual trend in winds for the three 352 reanalyses over the region of interest with trends that are significant at the 95% confidence level indicated. The broad trends across the Atlantic are consistent between the reanalyses, primarily 353 a significant stilling between 10 – 20°N and a strengthening of the wind in the Gulf of Guinea and 354 355 south of the equator. Across North Africa, all three reanalyses show significant stilling in regions 356 associated with dust production. The trends in MERRA are generally stronger than observed in the two other reanalysis products; however, it has been shown that NCEP and ERA-Interim 357 358 reanalysis wind trends are weaker than trends in surface observations (Cowie et al., 2013; 359 Vautard et al., 2010) and therefore the stronger trend in MERRA is expected to agree better with the surface observations. There is some disagreement in the latitude and strength of the stilling 360 361 over the dust source regions but the consistency in the significant stilling trends between reanalysis products bolsters confidence in the MERRA surface wind trends. 362

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5. Potential mechanism to explain the trends in African dust

Thus far, we have shown that the trends in North African dust are driven by stilling of the surface wind, more likely from large-scale changes in circulation than the result of land-use and vegetation changes. Here we consider a potential mechanism that may explain the recent trend. Booth et al. (2012) have shown that changes in aerosol indirect effects over the North Atlantic, primarily driven by anthropogenic aerosol, may play a key role in modulating the North Atlantic sea surface temperature (SST) and that aerosol changes may be the source of the Atlantic Multidecadal Oscillation (AMO) that is often shown to correlate with African dust concentrations 373 (Chin et al., 2013; Shao et al., 2013; Wang et al., 2012). Several studies have shown a southward 374 displacement of the inter-tropical convergence zone (ITCZ) in response to a decrease in the North 375 Atlantic SST (Broccoli et al., 2006; Rotstayn and Lohmann, 2002; Williams et al., 2001). While 376 these studies generally consider the implications in terms of the change in precipitation and 377 drought in the Sahel, the location of the ITCZ is associated with changes in the wind strength and 378 direction over North Africa as well (Doherty et al., 2012; Fontaine et al., 2011). A warming of the North Atlantic is expected to produce a northward shift (or broadening) in the ITCZ, bringing more 379 380 precipitation to the Sahel and also associated with decreasing wind strength, and therefore both 381 a vegetative greening and a reduction in dust emissions, as observed. This presents a potentially 382 important connection between anthropogenic aerosol loading in the Northern mid-latitudes and 383 changes in African dust emissions.

While it is beyond the scope of this paper to investigate the cause of large-scale wind changes 384 385 (GEOS-Chem is not a coupled climate model), we present a brief analysis of surface winds from 386 the CMIP5 models and reanalysis products. Using monthly mean surface wind output from fifteen CMIP5 'historical' simulations we derive surface wind trends for 1982 - 2008 over Africa and the 387 Mid-Atlantic and compare these with the average trend from the three meteorological 388 reanalysis. The surface wind trends in the CMIP5 models over Africa and the Atlantic do not 389 390 match the reanalysis products, with spatial correlations varying between -0.3 and 0.3. However, 391 each of the models considered contain atmospheric modules of differing complexity and may capture different processes. To investigate the potential link between the aerosol indirect effect 392 393 (AIE) and surface wind trends we group the models based upon whether or not they include 394 aerosol feedback upon cloud properties, i.e. the aerosol indirect effect, required to model 395 observed SST changes in Booth et al. (2012). Eight are found to include a parameterization for 396 the feedback and seven do not. Using bootstrapping, we randomly sample three models 500 397 times, obtain the trend in surface winds from the ensemble, and calculate the spatial correlation with the reanalysis products over the North Africa / Atlantic region. Figure 9 shows the 398 399 distribution of correlation coefficients for all samples and for subsets with only those models 400 containing the AIE and those without AIE. We find that when the sample contains only those models known to represent the aerosol indirect effect the correlation with reanalysis surface 401 402 wind trends is better than 91% of the other random samples, with a mean correlation of 0.16 (-0.05 to 0.29). Furthermore, when the sample contains only those models known to not contain 403 404 the aerosol indirect effect the correlation is worse than 90% of the random samples, correlation 405 of -0.08 (-0.22 to 0.13). While these correlations are extremely weak, the results with and without aerosol indirect effect are significantly different to one another (greater than 99% confidence) 406 407 and to the mean of all the randomized samples (0.05), indicating there may be a link between 408 simulating the indirect effect of aerosols and better representation of trends in surface winds, as 409 represented by the assimilated meteorology.

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411 **6. Summary and Conclusions**

This research and previous studies have found that satellite observations across the Atlantic show a significant downward trend in DAOD since the 1980s, persisting through both summer and winter seasons. We also observe decreasing trends in surface dust concentrations at Barbados during summer. The GEOS-Chem model captures the broad trends in dustiness over the Atlantic and we estimate that they lead to an annually averaged warming of +1.23 Wm⁻² per decade over the surface of the mid-Atlantic since 1982. We find that the trends are driven primarily by a reduction in surface winds in regions that are unlikely to be associated with vegetation cover changes. This suggests that the change in African dust emissions since the 1980s cannot be directly attributed to vegetation changes (including anthropogenic land use changes in the Sahel region); therefore, a vegetation feedback on dust emission via surface roughness may be valid on a local scale but appears less important for dust sources responsible for the trends in dustiness over the Atlantic.

424 Along with isolating the cause of the trends, the drivers of interannual variability in dustiness 425 over the Atlantic have been investigated. We find that the interannual variability is primarily controlled by changes in surface winds over Africa, accounting for 60 - 80% of the interannual 426 427 variability in dust AOD off the coast of Africa. Further downwind, transport and (to a lesser 428 extent) precipitation contribute 30 - 60% and 0 - 15% to the variability in dust AOD, respectively, 429 depending upon season. Using the model we find that the variability in dust leads to substantial interannual changes in surface insolation of over 6.0 Wm⁻², averaged over the mid-Atlantic. These 430 431 are likely to have a significant impact on heating of the ocean mixed layer therefore and tropical storm genesis (Evan et al., 2009), highlighting the importance of well-characterized variability in 432 dust emissions in climate models. 433

Finally, we propose a potential connection between anthropogenic aerosol loading over the North Atlantic and the trends in dustiness in the mid-Atlantic. The link between the aerosol direct and indirect effects over the North Atlantic and changes in the SST and ITCZ are well established. We take this one step further and suggest that the wind stilling over Africa, reducing dustiness over the Atlantic in recent decades, may be a further consequence of these interactions. The 439 CMIP5 models do not capture the wind trend over Africa, preventing conclusive evidence of this 440 mechanism. However, the CMIP5 models that include aerosol indirect effects show significantly 441 better agreement with surface wind trends in reanalysis meteorology than those without indirect aerosol effects, offering evidence that the aerosol indirect effect may be critical to the prediction 442 443 of surface winds trends over Africa. This is a potentially important anthropogenic aerosol driver upon 'natural' dust aerosol via climate, capable of amplifying the climate sensitivity to 444 anthropogenic aerosol in the Atlantic, which is not captured by the aerosol radiative forcing 445 446 metric.

447

448 Acknowledgements

449 The authors would like to thank Charlie Koven for the dust source function, Amato Evan for the

450 satellite-derived dust AOD product, John Marsham and Sophie Cowie for providing data on the

451 dust source classification, Kerstin Schepanski for supplying dust activation data derived from

452 SEVIRI, and Owen Doherty for discussions and data relating to the ITCZ. This work was funded

453 by the MIT Charles E. Reed Faculty Initiative Fund and the National Science Foundation (AGS-

454 1238109). The Barbados research is funded with grants to J. M. Prospero from the National

455 Science Foundation AGS-0962256 and NASA NNX12AP45G.

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- 696
- 697 Figure captions



Figure 1 – Spatial maps of the average direct radiative effect of dust (DRE) at the surface and TOA
 are shown for both winter (left) and summer (right) seasons. The average is based on model
 output over the period 1982 – 2008. The inset shows the seasonal average DRE over the region
 0°N - 30°N, 50°W - 15°E. The thin black line indicates the trend over the simulation period.



Figure 2 – The areas of interest for this study. The large black rectangle encloses the region covered by the satellite-derived monthly dust AOD dataset and the blue rectangles (labelled with roman numerals) are the regions within which model and satellite DAOD are compared. These are referred to as (I) Coastal Africa, (II) Caribbean, (III) South America and (IV) North Atlantic. Numbered circles indicate the location of AERONET sites used and the star shows the location of the surface concentration measurements in Barbados.



Figure 3 – Seasonal DAOD anomalies derived from AVHRR PATMOS-x (black) and from the model (red) are displayed for four regions (roman numerals relate to the locations shown in Fig. 2). Trend lines are plotted as solid lines and the trend and one standard deviation uncertainty shown in each panel for the observations (OBS) and the model (MOD). The correlation (R) between model and observations is shown in brackets.



Figure 4 – Seasonally-averaged surface concentrations anomalies from Barbados observations
(black) and from the model (red) are shown for winter and summer. Trend lines are plotted as
solid lines and the slope with one standard deviation indicated for each season. The correlation
(R) between the observations and model data is shown on each panel.



- Figure 5 Attribution of the interannual variability in dust is shown for the four regions in Fig. 2.
- The approximate fraction of the variability owing to 10-m surface winds (blue), transport (orange)
- and precipitation (grey) is displayed for winter (left) and summer (right).





Figure 6 - Annual dust AOD (DAOD) anomalies are shown for the updated model (red, same as the model in Fig. 2), with interannual variability in 10-m winds removed (light blue), with interannual variability in all meteorology, except surface winds, removed (dark blue), and with interannual variability in vegetation cover removed (green). Thin solid lines show the trends, and the trend and one standard deviation uncertainty shown in each panel for the observations and the model. The correlation between observed dust AOD and each version of the model is shown in brackets. N.B. the red line is almost always obscured by the green line.



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Figure 7 – The difference between 2002-2006 and 1982-1986 over North Africa is shown for (a)
fractional vegetation cover, represented by the decrease in bareness fraction, and (b) MERRA 10m wind speed. Black contours outline regions where emissions decrease by more than 0.5 Tg per
grid box.



Figure 8 – Spatial maps of the annual trend in surface winds between 1982 and 2008 based upon
the averaged winds from MERRA, ERA-Interim and NCEP/NCAR reanalysis. Regions with trends
that are significant at the 95% level are indicated with hashing.



Figure 9 – Histogram of the correlation between the spatial distribution of CMIP5 ensemble surface wind trend and the reanalysis surface wind trend over 1982-2008. The histogram represents 500 correlations, each based on the average surface winds of 3 randomly sampled CMIP5 models (black). The blue histogram shows ensembles that only contain models with no aerosol indirect effect (AIE) representation and the magenta distribution only those that do contain a parameterization for the aerosol indirect effect.