Atmos. Chem. Phys. Discuss., 14, 3583–3627, 2014 www.atmos-chem-phys-discuss.net/14/3583/2014/ doi:10.5194/acpd-14-3583-2014 © Author(s) 2014. CC Attribution 3.0 License.



This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

What controls the recent changes in African mineral dust aerosol across the Atlantic?

D. A. Ridley¹, C. L. Heald^{1,2}, and J. M. Prospero³

¹Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, MA, USA

²Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA, USA

³Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, FL, USA

Received: 10 January 2014 - Accepted: 22 January 2014 - Published: 10 February 2014

Correspondence to: D. A. Ridley (daridley@mit.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.



Abstract

Dust from Africa strongly perturbs the radiative balance over the Atlantic, with emissions that are highly variable from year to year. We show that the aerosol optical depth (AOD) of dust over the mid-Atlantic observed by the AVHRR satellite has de-⁵ creased by approximately 10% per decade from 1982–2008. This downward trend persists through both winter and summer close 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 and using a new dust source activation scheme, reproduces the observed trend and is used to quantify the factors contributing to this trend and the observed variability from 1982 to 2008. We find that changes in dustiness over the East mid-Atlantic are almost entirely me-

- diated by a reduction in surface winds over dust source regions in Africa and are not directly linked with 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, ac-
- ¹⁵ counting for 46 % of the global dust total, with a regional DRE of -7.4±1.5 Wm⁻² at the surface of the mid-Atlantic, varying by over 6.0 Wm⁻² from year to year, with a trend of +1.3 Wm⁻² per decade. These large inter-annual changes and 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 over past 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.

1 Introduction

Mineral dust aerosol is ubiquitous in the atmosphere and arguably the greatest source of particulate matter. Africa is responsible for approximately half of the global emissions (Huneeus et al., 2011) resulting in transport of several hundred teragrams (Tg)



of dust across the Atlantic towards the Americas throughout the year (Ginoux et al., 2004; Kaufman et al., 2005; Ridley et al., 2012). This has consequences for air quality downwind (Prospero, 1999; Viana et al., 2002; Kaufman et al., 2005b) and also the radiative balance over the Atlantic, via scattering and absorption of solar radiation

(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; Evan et al., 2006; Kaufman et al., 2005a). African dust emissions vary greatly from year to year (Ben-Ami et al., 2012; Chiapello, 2005; Ginoux et al., 2004), implying considerable variation in the aforementioned impacts on climate and air quality.

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 2000 yr than during the African Humid Period (11700–5000 yr ago) and that emissions during glacial periods are generally 2–4 times greater than interglacial peri-

- ods, likely owing to stronger winds (McGee et al., 2013, 2010). More recently, Mulitza et al. (2010) determined that dust emissions from Africa were well 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 advent of commercial agriculture in the 1800s, indicating the potential for anthropogenic changes to influence dust
- ²⁰ emission. Population growth in Africa over recent decades, by a factor of 3 since 1950 (Prospero and Lamb, 2003), has led to an increase in agricultural activity and urbanization, with speculation that human-induced land use and vegetation changes may also contribute to recent trends in West African dust (Chiapello, 2005; Evan et al., 2011). Since the 1950s, dust emissions from Africa have increased (Evan and Mukhopad-
- ²⁵ hyay, 2010; Mbourou et al., 1997; Prospero et al., 2002), peaking in the 1980s at the 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–Sahel Precipitation Index of the previous year (Prospero and Lamb, 2003). In the past three decades, observations from satellite and surface measure-



ments indicate a decrease in dustiness (Chin et al., 2013; Evan and Mukhopadhyay, 2010; Hsu et al., 2012; Shao et al., 2013; Zhao et al., 2008) coinciding with a greening of the Sahel region (e.g. Olsson et al., 2005). A vegetation-related increase in surface roughness may also have contributed to the stilling of winds (and corresponding reduc-

tion in dust) over much of the Northern Hemisphere (Vautard et al., 2010; Bichet et al., 2012), including the Sahel region (Cowie et al., 2013).

Modeling studies generally agree that changes in precipitation over the Sahel, leading to the drought and subsequent greening, can be explained by changes in the interhemispheric temperature gradient across the Atlantic (Chiang and Friedman, 2012;

- ¹⁰ Hwang et al., 2013; Rotstayn and Lohmann, 2002; Zhang and Delworth, 2006) which influences the location of the Inter-Tropical Convergence Zone (ITCZ). The location of the ITCZ is associated with changes in dust emissions and transport over the Atlantic (Doherty et al., 2012; Fontaine et al., 2011) 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-scale changes associated with the ITCZ location or a consequence of the greening of the Sahel, 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 on comparison with observations (Moulin et al., 1997; Chiapello, 2005;

- Nakamae and Shiotani, 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 high (Hurrell, 1995), is extremely noisy and our understanding of what causes the fluctuations is limited (Stephenson et al., 2000). While the NAO index represents changes in circulation that affect dust trans-
- ²⁵ port, 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, 2013). Therefore, we choose to focus primarily on the physical processes the drive dust emission and export (e.g. wind strength and precipitation) rather than the climate indices that may represent them.



In this paper we use surface and satellite dust observations along with a 27 yr 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. First, we evaluate an ⁵ updated dust scheme in the GEOS-Chem model using satellite (MODIS) and ground-based observations (AERONET and surface concentrations). Second, the magnitude of modeled and observed variability and trends in Atlantic dust AOD are assessed and the causes quantified using the model. Third, we use the RRTMG radiative transfer model coupled with GEOS-Chem to quantify the variability and trends of the direct ¹⁰ radiative effect of African dust. Finally, we discuss a potential driver of changes in surface winds using three reanalysis datasets and fifteen CMIP5 model simulations.

2 Model description

The GEOS-Chem model (version v9-01-01: http://www.geos-chem.org/) incorporates a global three-dimensional simulation of coupled oxidant-aerosol chemistry, run at a resolution of $2^{\circ} \times 2.5^{\circ}$ latitude and longitude, and 47 vertical levels in this study. 15 The model is driven by assimilated MERRA meteorology for 1982–2008 from the Goddard Earth Observing System of the NASA Global Modeling and Assimilation Office (GMAO), which includes assimilated meteorological 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-ammonium 20 aerosols (Park et al., 2004), and carbonaceous aerosols (Henze et al., 2008; Liao et al., 2007; Park et al., 2003). Aerosol optical depth (AOD) is calculated online assuming lognormal 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 are based on the Global Aerosol Data Set (GADS) 25 (Kopke et al., 1997) with modifications to the size distribution based on field observa-



tions (Drury et al., 2010; Jaegle et al., 2010; Ridley et al., 2012) and to the refractive index of dust (Sinyuk et al., 2003).

For all long-term simulations in this study the standard model is modified to include dust aerosol only. Emission, dry deposition and wet scavenging of dust are all simu-

- Iated as in the standard model (Fairlie et al., 2007). Dust emission in GEOS-Chem is based upon the DEAD dust scheme (Zender et al., 2003a), making use of the GO-CART source function (Ginoux et al., 2001) as proposed 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 μm), the smallest of which 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 optical properties, owing to the strong size-dependence of extinction for submicron aerosol (Ridley et al., 2012).

The GEOS-Chem model has been coupled with RRTMG, a rapid radiative transfer model (Mlawer et al., 1997), to calculate the direct radiative effect (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.

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 variabil-

- ity 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 ($0.5^{\circ} \times 0.67^{\circ}$). We previously showed that accounting for the sub-grid wind distribution reduces the resolution-dependence
- ²⁵ of dust emissions in the model by better representing situations when the mean wind speed is approaching the threshold for dust activation (Ridley et al., 2013).

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 (Ginoux et al., 2012; Koven and Fung, 2008; Schepanski et al., 2007)



the original GOCART dust 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 cloud cover and temporal sampling, particularly in regions where emissions show distinct diurnal patterns, such as West Africa (Schepapski et al. 2012). For this reason we implement the geomorphic dust source

- ⁵ (Schepanski et al., 2012). For this reason we implement the geomorphic dust source map of Koven et al. (2008), derived from surface roughness and 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 (NDVI) for each year (values < 0.15 are considered bare and a potential dust source)</p>
- to modulate dust emissions from 1982–2008. Figure 1 compares the normalized dust emissions for both the GOCART and geomorphic source maps averaged over the period 2004–2008, both scaled to give the same total emissions for the region. The key source regions of the Bodélé Depression and in Central Sahara are well represented in both, with more weighting given to the former when using the geomorphic source
- ¹⁵ map. Relative to the potential source regions outlined in Formenti et al. (2011), five of the six regions are represented in the current dust scheme; however, emissions from the region at the Mali – Niger border (20° N, 5° E) appear weak in both dust schemes. Another key difference is that the area from which emissions can occur is broader in the geomorphic source map, with potential emissions extending further south if vegetation
- ²⁰ cover permits. The dust emission frequency derived from SEVIRI satellite observations by Schepanski et al. (2007) shows that emissions are possible from a broader region, in agreement with the geomorphic source map. While it has been shown that geomorphic source maps can lead to erroneous dust emissions in the US as a result of the vegetation-dependence (Fairlie et al., 2007; Zender et al., 2003b), comparison
- ²⁵ between the model and observations in Africa suggests that the new source function performs at least as well as the original GOCART source function in this region (see Sect. 4.1).



3 Observations

The primary long-term observations used in this study is a dust AOD (DAOD) dataset over the mid-Atlantic for the period 1982–2008, derived from satellite observations by Evan and Mukhopadhyay (2010). Satellite retrievals of AOD from the AVHRR PATMOS-

- x dataset, an 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 mode fraction products and with NCEP-NCAR reanalysis surface winds, following Kaufman et al. (2005b). The 1° × 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, 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 this region was divided into two, north and south of Cape Verde, however the results are largely the same when considering the region as a whole.
- The MODIS instrument provides near daily coverage of the globe and AOD retrievals at 550 nm that are well validated against AERONET observations (Levy et al., 2010). The MODIS Deep Blue retrieval is used to estimate AOD over bright surfaces (Hsu et al., 2004), providing complementary observations in regions not covered by AERONET in the Sahara. In this study we use daily Level 3 MODIS observed AOD
 from the Aqua satellite (13:30 LT equatorial overpass, Collection 5) with the Deep Blue
 - retrieval where no standard retrieval is available.

The Aerosol Robotic Network (AERONET) is a well-established global network of sun photometers used to derive aerosol optical depth (AOD) at a set of standard wavelengths (440, 675, 870 and 1020 nm) on a daily basis (Holben et al., 1998). Almucantar

²⁵ measurements (a series of measurements taken for multiple azimuth angles at the elevation of the sun) allow size and scattering information to be estimated from the retrieved extinction, producing a measure of the contribution of fine (< 1 um) and coarse aerosol to the AOD (Dubovik et al., 2002). In this study, we use daily AOD retrievals</p>



from eight AERONET sites across West Africa (locations shown on Fig. 2) between 2004 and 2008 to evaluate the model representation of AOD close to source.

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 condi-

- tions 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 20 m 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 (J. Prospero, personal communication, 2013) 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.

4 Dust transport and trends

4.1 Climatological evaluation of GEOS-Chem

Several previous studies have assessed the skill of the DEAD dust scheme in the GEOS-Chem model and show reasonable agreement with satellite and surface observations both close to source and downwind (Fairlie et al., 2007; Generoso et al., 2008; Johnson et al., 2010; Ridley et al., 2012). Given the model alterations described in Sect. 2 (inclusion of a sub-grid wind parameterization, a new source map and dynamic
 vegetation), we re-evaluate the model for North Africa using AERONET observations and MODIS retrievals close to source and surface concentrations downwind in Barba-



dos. We use a five year simulation (2004–2008) with all aerosol components included to allow for direct comparison with AOD observations from MODIS and AERONET.

Figure 3 compares the seasonal AOD (averaged over 2004–2008) observed by MODIS to the GEOS-Chem simulation with the original and updated dust scheme.

- ⁵ The spatial correlation between model and MODIS coarse AOD over the dust source region in North Africa (10–36° N, 22.5° W–32.5° E) is slightly improved in all five years when using the updated dust scheme, from 0.72 to 0.78 in winter and from 0.63 to 0.67 in summer. This improvement is largely the result of the reduction in emissions from the West Coast relative to those from the Bodélé Depression produced by the new
- source function and vegetation modulation. However, the fraction of modeled AOD at the Bodélé Depression is still low relative to the observations; this contributes to the regional model AOD low biases of 21 % and 38 % in winter and summer, respectively. We find that suppression of emissions by soil moisture and vegetation is limited in the Bodélé region, suggesting that MERRA surface winds are too weak, particularly in
- ¹⁵ August and September, to generate the large flux of dust observed. Biomass burning aerosol below approximately 12° N during the winter and sea salt aerosol in coastal regions may both influence the agreement with MODIS and AERONET, but we expect these effects to be small relative to the dust aerosol that accounts for over 70% of the annual AOD between 10° N and 36° N in the model.
- Figure 4 shows comparison of daily measurements at eight AERONET sites with the updated model AOD (including non-dust species) during the period 2004–2008. The temporal correlation averaged across the sites (and weighted by the number of observations) increases from 0.75 to 0.78 during winter, and increases from 0.66 to 0.72 in summer when using the updated dust scheme. The RMS error relative to AERONET
- is reduced during the summer at all eight sites but worsened slightly at Sahelian sites during the winter using the updated dust scheme. We find that including the sub-grid wind PDF reduces the RMS error across almost all sites and seasons but the inclusion of the new source function and vegetation modulation masks this improvement in winter, suggesting that wintertime source regions are now over-represented in the Sahel



(excluding the Bodélé Depression). While the total improvement relative to the observations is small, the new dust emission scheme is considered to be more realistic as it represents both sub-grid winds and the modulation of dust emissions from vegetation changes. General agreement with daily observations from the AERONET sites is

- ⁵ good, with the majority of days falling within a factor of 2 of the observations and seasonal correlation coefficients greater than r = 0.6 for all but one location. In the Sahel, there is a tendency for the model to overestimate the AOD during high aerosol loading (predominantly in winter) and underestimate the AOD in summer, potentially from poor representation of dust emissions driven by local convective storm down drafts,
- i.e. Haboobs (Marsham et al., 2011). Attribution of the wintertime overestimation is confounded by the presence of biomass burning aerosol. In addition, the AERONET sites are clustered in the Sahel biasing the comparison to this particular region and underrepresenting the Sahara. Tamanrasset, located in the Sahara but perhaps not representing the area well (Cuesta et al., 2008), indicates a similar picture to the Sahel
 with the model AOD biased high during winter and low during summer.
- Figure 5 shows the comparison of both the original and updated dust model with a monthly climatology of surface dust concentrations at the Barbados site over the period 1982–2008. The observed climatology shows a peak in the average concentration in June (30 μgm⁻³) and a minimum in December (7.5 μgm⁻³), with greatest inter-annual variability between March and June. The updated source map shifts the peak in modeled concentration from July to June, in better agreement with the observations, and also improves the seasonality by increasing the summertime peak average concentration from 21.0 μgm⁻³ to 26.0 μgm⁻³ and decreasing the wintertime average concentration slightly.
- ²⁵ We also investigated the possibility of using the SEVIRI dust source activation frequency (DSAF) to constrain the emissions in the model. We scaled the wind threshold for emission such that the model produced the best fit with the number of dust event days per month, derived from four years of SEVIRI observations (Schepanski et al., 2007). While the wind threshold scaling boosted emissions from the Bodélé Depres-



sion during summer and reduced them in coastal regions, which would be expected to improve the model based on Fig. 3, the improvement to the model dust AOD was minimal when compared with both MODIS and AERONET. This suggests that the efficacy of DSAF for improving the simulation of dust may be limited by the inability to consider the size of the dust event, in agreement with Tegen et al. (2013). Scaling the wind threshold for emission may be masking errors in MERRA surface winds rather than providing useful information on surface susceptibility to emission; therefore, we have chosen not to include the DSAF scaling in the simulations used in this study.

The model exhibits little change in skill considering that the source regions have changed quite dramatically between the standard and updated dust schemes. This suggests that the wind fields dominate the agreement with observations, both in terms of the surface wind strength leading to emissions and the large-scale transport across the continent. Achieving a higher fidelity dust simulation therefore appears to rely on an improvement of the wind fields, rather than the characterization of the surface properties.

4.2 Trends and variability in dustiness

5

20

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 regions close to source and further downwind. Figure 6 shows the seasonally-averaged DAOD for the model and derived from satellite observations in each of the outflow regions indicated in Fig. 2 from 1982 to 2008. The DAOD is displayed as an anomaly from the climatological average for the winter and summer seasons.

Close to source, the model captures 50–80 % of the variance in the observations in the winter 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 and magnitude of emissions between June and August; likely a consequence of underestimated AOD at the Bodélé Depression and across the Sahel



during summer (see Fig. 3). Downwind, correlations between model and observations range between 0.46 and 0.65 (excluding the North America region during winter when very little dust is present). The poorer agreement downwind indicates that model transport and removal contribute to the discrepancy with observations. The variability in the

- ⁵ model DAOD is generally less than observed, owing to lower model DAOD; however, the normalized variability (normalizing using the climatological mean DAOD) is comparable between model and observations. Previous studies have shown that 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 reduced variability in DAOD at these downwind regions. Overall we see best
- to the reduced variability in DAOD at these downwind regions. Overall we see best agreement for seasons with the highest dust loading, i.e. winter at the South America region and summer in the Caribbean and North Atlantic regions.

Significant decreasing trends in observed DAOD (> 95% confidence) are apparent for all seasons and locations, except winter in the North America and Caribbean
regions when northerly transport of dust is limited and highly variable, respectively. There are striking similarities between the trends in observed and modeled DAOD, with the model showing significant trends at the same locations and during the same seasons. Annually, observed and modeled DAOD 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 total AOD per decade in the mid-Atlantic based on the AVHRR PATMOS-x dataset (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 almost entirely by the changes in dust aerosol.

There is significant interannual variability in dust concentration during the months March–May, a period responsible for significant transport of dust to South America (Prospero et al., 1981). We find that the downward trends in DAOD are present in both the traditional summer season (June–August) as well as spring (March–May), a period responsible for significant transport of dust to South America based on both satellite



and in-situ measurements (Kaufman et al., 2005b; Prospero et al., 2014). Isolating the spring season we find significant trends in DAOD of -0.02 per decade for both observations and model in the South America region (r = 0.70) indicating that the decreasing dust trends are present and captured by the model in this season as well as the other seasons.

Figure 7 shows the anomaly in monthly dust concentration measured at Barbados alongside the modeled surface concentration anomaly. We find that seasonal correlation between the dust 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, respectively. A decreasing trend in surface con-

- r = 0.44 during winter and summer, respectively. A decreasing trend in surface concentration measured at Barbados is found during the summer $(-3.5 \pm 1.3 \,\mu gm^{-3} per decade)$ and is also present in the model surface concentration $(-5.2 \pm 1.1 \,\mu gm^{-3} per decade)$. No trend is present in either observations or model during the winter, consistent with the Caribbean regional DAOD (Fig. 6).
- ¹⁵ 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.

4.3 Attribution of variability and trends in dustiness

from surface roughness changes, discussed further below.

5

To attribute the driving forces behind the inter-annual variability in African dust near
 source and downwind, four 27 yr simulations are performed with inter-annual variability removed from surface winds, transport, precipitation, or vegetation. The inter-annual 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). 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 yr 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



The variability caused by each of the three factors is inferred by assessing the reduction in the variance of the DAOD in each region relative to the original model. This method does not account 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 %) when removing the interannual variability using data from 2004 instead of 1988. Figure 8 shows the apportionment of variability in DAOD to the three factors tested for summer and winter. In 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 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 of the variability in DAOD in all regions during the winter, but becomes more important downwind during the summer. We find that precipitation primarily affects the variability in 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 on the variability in DAOD suggesting that the changes in dust source region resulting from vegetation cover changes are unimportant. The seasonal NAO index correlates with the modeled and observed DAOD in winter (r = 0.72 and r = 0.56, respectively) but only weakly in the model in summer (r = 0.17 and r = 0.26).
- ²⁰ This indicates that some of the interannual variability in processes affecting the dust export during winter, but not summer, are captured by the NAO climate index.

The same four simulations described above are used to assess the cause of trends in DAOD. Figure 9 shows the resulting annual DAOD anomaly in each region for each simulation. In the Coastal Africa region it is clear that removing inter-annual variabil-

ity in 10 m winds almost entirely removes the trend in DAOD. Further downwind the inter-annual variability in other meteorology contributes between 30–50 % of the trend; however, the surface wind at source remains the 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 removal down-



wind, more so in the more northerly outflow regions. We find that the direct effect of vegetation changes on dust emission in the model has a negligible impact on the trend in dustiness over the Atlantic. Although surface winds have been inferred as the likely cause of the observed reduction in Atlantic dust loading (Chin et al., 2013) this is the first time that the link has been quantified.

5

10

While we do find a positive correlation between the NAO index and the DAOD in the outflow regions during winter, we find no significant trend in the NAO index between 1982 and 2008, therefore the question remains as to what is driving the stilling of winds over Africa. We have shown that changes in vegetation are unlikely to *directly* influence dust emission via changes in source regions but they may still indirectly affect the emissions via stilling of the winds. If vegetation changes are driving the decrease in winds responsible for the change in dustiness over the Atlantic then any increase in vegetation cover is expected to be coincident with the decrease in winds (Bichet et al., 2012).

- Figure 10a shows the change in the bareness fraction (a reduction in bareness fraction indicating a "greening") derived from AVHRR between 1982–1986 and 2002–2006 for summer and winter. Figure 10b shows the change in surface winds apparent in the MERRA reanalysis (blue-red shading) with black contours overlaid, indicating the regions in which dust emissions decrease by more than 0.5 Tg per grid box. For both
- seasons we see that there is a limited amount of overlap between the location in which vegetation increases and the 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 when dust is transported from elsewhere are excluded and account for between 50 % and 90 % of all dust events at
- the Sahel weather stations (S. Cowie, personal communication, 2013). The reanalysis winds are unlikely to capture the full extent of wind stilling from surface roughness changes and 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 regions other than the Sahel are controlling the trends in



Atlantic DAOD. The lack of spatial correspondence between the greening and the wind stilling suggest that vegetation is not driving the change in model surface winds and perhaps both are the result of a larger-scale climatic change. Indeed, shifts in dustiness over the past 20 000 yr also suggest that large changes in dust emission in the past are primarily driven by changes in large-scale winds, rather than vegetation and precipitation changes (McGee et al., 2010).

4.4 Reliability of surface wind trends in reanalyses

5

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 NCEP and ERA-Interim reanalysis products. Figure 11 shows the annual trend in winds for the three 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 a significant stilling between 10–20° N and a strengthening of the wind in the Gulf of Guinea and south of the

- equator. Across North Africa, all three reanalyses show significant stilling in regions 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 reanalysis wind trends are weaker than trends in surface observations (Cowie et al., 2013; Vautard et al., 2010) and therefore the stronger trend in MERRA is
- 20 expected to agree better with the surface observations. There is some disagreement in the latitude and strength of the stilling over the dust source regions but the consistency in the significant stilling trends between reanalysis products bolsters confidence in the MERRA surface wind trends.

4.5 Radiative effects of dust variability and trends

²⁵ The considerable interannual changes in the Atlantic dustiness seen in Fig. 6 are likely to have a significant impact on the radiative balance over this region. We use the



GEOS-Chem model coupled with RRTMG (Heald et al., 2013) to quantify the change in radiative flux at the surface resulting from the dust aerosol over the 27 yr period (for all-sky conditions).

- Figure 12 shows the average seasonal direct radiative effect (DRE) of dust over
 Africa and the Atlantic for the whole 27 yr 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 all-sky radiative effect from African dust is -0.18 Wm⁻² at TOA, accounting for 46% of the total 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 of +0.4 ± 0.1 Wm⁻² and +3.1 ± 0.7 Wm⁻², respectively. This region covers less that 5% of the Earth's 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 atmosphere owing to dust. When the dust outflow is over the Sahara (primarily during summer) the airborne dust can be darker than the surface between the TOA and producing a warming effect
- beneath, decreasing the amount of outgoing radiation and producing a warming effect at TOA (Fig. 12).

While direct comparison with previous estimates is difficult due 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). Evan and Mukhopadhyay (2010) estimate the radiative effect of dust over the same region based on the same satellite DAOD used in this study, but extended back to 1955. We find that our estimate of annual mean DRE is
- ²⁵ slightly larger than that derived from the satellite observations $(-1.7\pm0.7 W m^{-2} at TOA and -5.4\pm2.5 W m^{-2} at the surface for the 53 yr climatological average). The seasonal cycle in modelled DRE is weaker than in Evan and Mukhopadhyay (2010) owing to a stronger DRE in winter in the model. Comparison with AERONET and MODIS AOD does suggest that wintertime emissions may be biased high in the model, especially$



towards the west coast; however, overestimating the biomass burning aerosol contribution when deriving DAOD from the satellite retrievals would cause an underestimation of dust DRE in the winter months when biomass burning peaks in West Africa.

- Balkanski et al. (2007) show that significant uncertainty arises from the choice of refractive index and that, based on AERONET observations, dust is typically less absorbing than previously assumed. The refractive indices used in this model have been updated to reflect this (e.g. 1.56–0.0014i at 550 nm) and are within the range of uncertainty of the AERONET observations. We simulate atmospheric heating greater than the best fit scenario of Balkanski et al. (2007) but less than the typical over-prediction
- ¹⁰ when using values from (Patterson et al., 1977), indicating that the refractive index of dust at longer wavelengths may still be overestimated. Aerosol size, absorption, 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 ra-¹⁵ diative 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.

20

The inset charts within Fig. 12 show the modeled time series of seasonal DRE for the region $0-30^{\circ}$ 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 cooling varies by up to 8 Wm^{-2} in winter and 6 Wm^{-2} in

summer, and a more modest 4 Wm^{-2} and 1.5 Wm^{-2} at TOA in winter and summer, respectively.

²⁵ Annually, warming trends of +1.27 Wm⁻² and +0.37 Wm⁻² per decade are observed at the surface and TOA, respectively, over the region including both the ocean and land from 1982 to 2008. The seasonal trends are similar in magnitude with both winter and summer trends significant (95% confidence). Similar trends in DRE exist over both ocean and land at the surface (+1.23 Wm⁻² and +1.32 Wm⁻² per decade, respec-



tively) but at TOA there are marked differences between the trends over ocean and land $(+0.60 \text{ Wm}^{-2} \text{ and } +0.00 \text{ Wm}^{-2} \text{ per decade})$ as a result of surface albedo and the high concentration of large dust particles (increasing the LW warming effect). The regional trend constitutes an increase in DRE over the past three decades that is com-

⁵ parable in magnitude to the regional increase in CO₂ forcing since 1750 (IPCC, 2013). This illustrates the strong radiative perturbation potential of dust: climatic changes that are affecting the emission of African dust are likely to have significant impacts upon the radiative balance over the Atlantic that are not accounted for in the traditional radiative forcing metric.

10 5 Potential mechanism to explain the trends in African dust

Thus far, we have shown that the trends 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 (Chin et al., 2013; Shao et al., 2013; Wang et al., 2012). Several studies have shown a southward displacement of the inter-tropical con-
- vergence zone (ITCZ) in response to a decrease in the North Atlantic SST (Broccoli et al., 2006; Rotstayn and Lohmann, 2002; Williams et al., 2001). While these studies generally consider the implications in terms of the change in precipitation and drought in the Sahel, the location of the ITCZ is associated with changes in the wind strength and direction over North Africa as well (Doherty et al., 2012; Fontaine et al., 2011).
- ²⁵ A warming of the North Atlantic is expected to produce a northward shift (or broadening) in the ITCZ, bringing more precipitation to the Sahel and also associated with decreasing wind strength, and therefore both a vegetative greening and a reduction in



dust emissions, as observed. This presents a potentially important connection between anthropogenic aerosol loading in the Northern mid-latitudes and changes in African dust emissions.

- While it is beyond the scope of this paper to investigate the cause of large-scale
 ⁵ wind changes (GEOS-Chem is not a coupled chemistry-climate model), we present a brief analysis of surface winds from 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 Mid-Atlantic and compare these with the average trend from the three meteorological reanalysis. The surface
 ¹⁰ wind trends in the CMIP5 models over Africa and the Atlantic do not match the reanalysis products, with spatial correlations varying between –0.3 and 0.3. However, 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 (AIE) and surface wind trends we group the models based upon whether or
- not they include aerosol feedback upon cloud properties, i.e. the aerosol indirect effect, required to model observed SST changes in Booth et al. (2012). Eight are found to include a parameterization for the feedback and seven do not. Using bootstrapping, we randomly sample three models 500 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 13 shows the distribution of correlation coefficients for all samples and for subsets with only those models 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 wind trends is better than 91 % of the other random samples, with a mean correlation of 0.16 (-0.05 to
- ²⁵ 0.29). Furthermore, when the sample contains only those models known to not contain the aerosol indirect effect the correlation is worse than 90% of the random samples, correlation 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) and to the mean of all the randomized samples (0.05),



indicating there may be a link between simulating the indirect effect of aerosols and better representation of trends in surface winds, as represented in assimilated meteorology.

6 Summary and conclusions

- ⁵ Using a host of observations and the GEOS-Chem model, including an improved representation of sub-grid surface wind and dust source regions, we have assessed the variability and trends in Atlantic dust loading, the resulting direct radiative effect and quantified the drivers. Substantial interannual changes in surface insolation of over 6.0 Wm⁻², averaged over the mid-Atlantic, are modeled. These 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 dust emissions in climate models. We find that the interannual variability in dust is primarily controlled by changes in surface winds over Africa, accounting for 60–80 % of the
- interannual variability in dust AOD off the coast of Africa. Further downwind transport and, to a lesser extent precipitation, contribute to the variability in dust AOD (30–60% and 0–15%, respectively, depending upon season).

Satellite observations across the Atlantic show a significant downward trend in DAOD since the 1980s, persisting through both summer and winter seasons. Decreasing trends are also observed in surface dust concentrations at Barbados during summer.

- ²⁰ The GEOS-Chem model captures these broad trends in dustiness over the Atlantic and using this model we estimate that these lead to an annually averaged increase of $+1.23 \text{ Wm}^{-2}$ per decade over the surface of the mid-Atlantic since 1982. The model indicates these 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
- ²⁵ the change in African dust emissions since the 1980s cannot be directly attributed to vegetation changes (or anthropogenic land use changes); a vegetation feedback on



dust emission via surface roughness may be valid on a local scale but less important for large dust sources responsible for the trends in dustiness over the 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

- ⁵ suggest that the wind stilling over Africa reducing dustiness over the Atlantic in recent decades may be a further consequence of these interactions. The CMIP5 models do not capture the wind trend over Africa, preventing conclusive evidence of this mechanism. However, the CMIP5 models that include aerosol indirect effects show significantly 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 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 anthropogenic aerosol in the Atlantic, which is not captured by the aerosol radiative forcing metric.
- Acknowledgements. The authors would like to thank C. Koven for the dust source function, A. Evan for the satellite-derived dust AOD product, J. Marsham and S. Cowie for providing data on the dust source classification, K. Schepanski for supplying dust activation data derived from SEVIRI, and O. Doherty for discussions and data relating to the ITCZ. This work was funded by the MIT Charles E. Reed Faculty Initiative Fund. The Barbados research is funded
 with grants to J. M. Prospero from the National Science Foundation AGS-0962256 and NASA NNX12AP45G.

References

25

- Alexander, B., Park, R. J., Jacob, D. J., Li, Q. B., Yantosca, R. M., Savarino, J., Lee, C. C. W., and Thiemens, M. H.: Sulfate formation in sea-salt aerosols: constraints from oxygen isotopes, J.
- Geophys. Res.-Atmos., 110, D10307, doi:10.1029/2004JD005659, 2005.
 Balkanski, Y., Schulz, M., Claquin, T., and Guibert, S.: Reevaluation of Mineral aerosol radiative forcings suggests a better agreement with satellite and AERONET data, Atmos. Chem. Phys., 7, 81–95, doi:10.5194/acp-7-81-2007, 2007.



Ben-Ami, Y., Koren, I., Altaratz, O., Kostinski, A., and Lehahn, Y.: Discernible rhythm in the spatio/temporal distributions of transatlantic dust, Atmos. Chem. Phys., 12, 2253–2262, doi:10.5194/acp-12-2253-2012, 2012.

Bichet, A., Wild, M., Folini, D., and Schär, C.: Causes for decadal variations of wind speed over

⁵ land: sensitivity studies with a global climate model: decadal variations of land wind speed, Geophys. Res. Lett., 39, L11701, doi:10.1029/2012GL051685, 2012.

Booth, B. B. B., Dunstone, N. J., Halloran, P. R., Andrews, T., and Bellouin, N.: Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability, Nature, 484, 228–232, 2012.

¹⁰ Broccoli, A. J., Dahl, K. A., and Stouffer, R. J.: Response of the ITCZ to Northern Hemisphere cooling, Geophys. Res. Lett., 33, L01702, doi:10.1029/2005GL024546, 2006.

Chiang, J. C. H. and Friedman, A. R.: Extratropical cooling, interhemispheric thermal gradients, and tropical climate change, Annu. Rev. Earth Pl. Sc., 40, 383–412, 2012.

Chiapello, I.: Understanding the long-term variability of African dust transport across the

¹⁵ Atlantic as recorded in both Barbados surface concentrations and large-scale Total Ozone Mapping Spectrometer (TOMS) optical thickness, J. Geophys. Res., 110, D18S10, doi:10.1029/2004JD005132, 2005.

Chiapello, I. and Moulin, C.: TOMS and METEOSAT satellite records of the variability of Saharan dust transport over the Atlantic during the last two decades (1979–1997), Geophys. Res.

²⁰ Lett., 29, 17-1–17-4, 2002.

Cowie, S. M., Knippertz, P., and Marsham, J. H.: Are vegetation-related roughness changes the cause of the recent decrease in dust emission from the Sahel?, Geophys. Res. Lett., 40, 1868–1872, 2013.

Cuesta, J., Edouart, D., Mimouni, M., Flamant, P. H., Loth, C., Gibert, F., Marnas, F., Bouklila, A.,

- Kharef, M., OuchèNe, B., Kadi, M., and Flamant, C.: Multiplatform observations of the seasonal evolution of the Saharan atmospheric boundary layer in Tamanrasset, Algeria, in the framework of the African Monsoon Multidisciplinary Analysis field campaign conducted in 2006, J. Geophys. Res.-Atmos., 113, D00C07, doi:10.1029/2007JD009417, 2008. Doherty, O. M., Riemer, N., and Hameed, S.: Control of Saharan mineral dust transport to Bar-
- bados in winter by the Intertropical Convergence Zone over West Africa: winter dust in barbados and the ITCZ, J. Geophys. Res.-Atmos., 117, D19117, doi:10.1029/2012JD017767, 2012.



- Drury, E., Jacob, D. J., Spurr, R. J., Wang, J., Shinozuka, Y., Anderson, B. E., Clarke, A. D., Dibb, J., McNaughton, C., and Weber, R.: Synthesis of satellite (MODIS), aircraft (ICARTT), and surface (IMPROVE, EPA-AQS, AERONET) aerosol observations over Eastern North America to improve MODIS aerosol retrievals and constrain surface aerosol concentrations
- and sources, J. Geophys. Res., 115, D14204, doi:10.1029/2009JD012629, 2010. Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanre, D., and Slutsker, I.: Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atmos. Sci., 59, 590–608, 2002.

Duncan Fairlie, T., Jacob, D. J., and Park, R. J.: The impact of transpacific transport of mineral dust in the United States, Atmos. Environ., 41, 1251–1266, 2007.

10

20

Dunion, J. P. and Velden, C. S.: The impact of the Saharan air layer on Atlantic tropical cyclone activity, B. Am. Meteorol. Soc., 85, 353–365, 2004.

Evan, A. T. and Mukhopadhyay, S.: African dust over the Northern Tropical Atlantic: 1955–2008, J. Appl. Meteorol. Clim., 49, 2213–2229, 2010.

- Evan, A. T., Dunion, J., Foley, J. A., Heidinger, A. K., and Velden, C. S.: New evidence for a relationship between Atlantic tropical cyclone activity and African dust outbreaks, Geophys. Res. Lett., 33, L19813, doi:10.1029/2006GL026408, 2006.
 - Evan, A. T., Vimont, D. J., Heidinger, A. K., Kossin, J. P., and Bennartz, R.: The role of aerosols in the evolution of Tropical North Atlantic ocean temperature anomalies, Science, 324, 778– 781, 2009.
 - Evan, A. T., Foltz, G. R., Zhang, D., and Vimont, D. J.: Influence of African dust on oceanatmosphere variability in the tropical Atlantic, Nat. Geosci., 4, 762–765, 2011.
 - Fairlie, T. D., Jacob, D. J., and Park, R. J.: The impact of transpacific transport of mineral dust in the United States, Atmos. Environ., 41, 1251–1266, 2007.
- Fontaine, B., Roucou, P., Gaetani, M., and Marteau, R.: Recent changes in precipitation, ITCZ convection and northern tropical circulation over North Africa (1979–2007), Int. J. Climatol., 31, 633–648, 2011.
 - Formenti, P., Schütz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, K., Petzold, A., Scheuvens, D., Weinbruch, S., and Zhang, D.: Recent progress in understanding physical
- and chemical properties of African and Asian mineral dust, Atmos. Chem. Phys., 11, 8231– 8256, doi:10.5194/acp-11-8231-2011, 2011.
 - Generoso, S., Bey, I., Labonne, M., and Bréon, F.-M.: Aerosol vertical distribution in dust outflow over the Atlantic: comparisons between GEOS-Chem and Cloud-Aerosol Lidar



3608

Hsu, N. C., Herman, J. R., and Weaver, C.: Determination of radiative forcing of Saharan dust using combined TOMS and ERBE data, J. Geophys. Res.-Atmos., 105, 20649–20661, 2000.

effect of aerosols, Atmos. Chem. Phys. Discuss., 13, 32925-32961, doi:10.5194/acpd-13-32925-2013, 2013. Henze, D. K., Seinfeld, J. H., Ng, N. L., Kroll, J. H., Fu, T.-M., Jacob, D. J., and Heald, C. L.: 20 Global modeling of secondary organic aerosol formation from aromatic hydrocarbons: high-

vs. low-yield pathways, Atmos. Chem. Phys., 8, 2405–2420, doi:10.5194/acp-8-2405-2008,

Heald, C. L., Ridley, D. A., Kroll, J. H., Barrett, S. R. H., Cady-Pereira, K. E., Alvarado, M. J., and Holmes, C. D.: Beyond direct radiative forcing: the case for characterizing the direct radiative

Haywood, J., Francis, P., Dubovik, O., Glew, M., and Holben, B.: Comparison of aerosol size distributions, radiative properties, and optical depths determined by aircraft observations and Sun photometers during SAFARI 2000, J. Geophys. Res., 108, 8471, doi:10.1029/2002JD002250, 2003,

Blue aerosol products, Rev. Geophys., 50, RG3005, doi:10.1029/2012RG000388, 2012.

Ginoux, P., Prospero, J. M., Torres, O., and Chin, M.: Long-term simulation of global dust distribution with the GOCART model: correlation with North Atlantic Oscillation, Environ. Modell. Softw., 19, 113–128, 2004. Ginoux, P., Prospero, J. M., Gill, T. E., Hsu, N. C., and Zhao, M.: Global-scale attribution of

Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S. J.: Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res.-Atmos., 106, 20255-20273, 2001.

doi:10.1029/2008JD010154, 2008.

108. 8578. doi:10.1029/2002JD002552. 2003.

5

10

15

25

30

2008.

viron., 66, 1–16, 1998.

and Infrared Pathfinder Satellite Observation (CALIPSO), J. Geophys. Res., 113, D24209,

Paper **Attribution of African** mineral dust trends D. A. Ridley et al. **Discussion** Paper anthropogenic and natural dust sources and their emission rates based on MODIS Deep **Title Page** Abstract Introduction Conclusions References **Figures** Tables Discussion Paper Highwood, E. J.: Radiative properties and direct effect of Saharan dust measured by the C-130 Back Close aircraft during Saharan Dust Experiment (SHADE): 2. terrestrial spectrum, J. Geophys. Res., Full Screen / Esc Holben, B. N., Eck, T. F., Slutsker, I., Tanre, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J. A., **Discussion** Pape Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A.: AERONET - a fed-**Printer-friendly Version** erated instrument network and data archive for aerosol characterization. Remote Sens, En-Interactive Discussion

ACPD

cussion

14, 3583–3627, 2014

- Hsu, N. C., Tsay, S. C., King, M. D., and Herman, J. R.: Aerosol properties over bright-reflecting source regions, IEEE T. Geosci. Remote, 42, 557–569, 2004.
- Hsu, N. C., Gautam, R., Sayer, A. M., Bettenhausen, C., Li, C., Jeong, M. J., Tsay, S.-C., and Holben, B. N.: Global and regional trends of aerosol optical depth over land and ocean
- ⁵ using SeaWiFS measurements from 1997 to 2010, Atmos. Chem. Phys., 12, 8037–8053, doi:10.5194/acp-12-8037-2012, 2012.
 - Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R.,
- Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C. S.: Global dust model intercomparison in AeroCom phase I, Atmos. Chem. Phys., 11, 7781– 7816, doi:10.5194/acp-11-7781-2011, 2011.
 - Hurrell, J. W.: Decadal trends in the North Atlantic oscillation: regional temperatures and precipitation, Science, 269, 676–679, 1995.
- ¹⁵ Hwang, Y.-T., Frierson, D. M. W., and Kang, S. M.: Anthropogenic sulfate aerosol and the southward shift of tropical precipitation in the late 20th century, Geophys. Res. Lett., 40, 2845–2850, 2013.
 - Jaeglé, L., Quinn, P. K., Bates, T. S., Alexander, B., and Lin, J.-T.: Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations, Atmos. Chem.
- ²⁰ Phys., 11, 3137–3157, doi:10.5194/acp-11-3137-2011, 2011.
 - Johnson, M. S., Meskhidze, N., Solmon, F., Gassó, S., Chuang, P. Y., Gaiero, D. M., Yantosca, R. M., Wu, S., Wang, Y., and Carouge, C.: Modeling dust and soluble iron deposition to the South Atlantic Ocean, J. Geophys. Res., 115, D15202, doi:10.1029/2009JD013311, 2010.
- Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D., and Rudich, Y.: The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean, P. Natl. Acad. Sci. USA, 102, 11207–11212, 2005a.
 - Kaufman, Y. J., Koren, I., Remer, L. A., Tanre, D., Ginoux, P., and Fan, S.: Dust transport and deposition observed from the Terra-Moderate Resolution Imaging Spectroradiome-
- ter (MODIS) spacecraft over the Atlantic ocean, J. Geophys. Res.-Atmos., 110, D10S12, doi:10.1029/2003JD004436, 2005b.



Kim, D., Chin, M., Bian, H., Tan, Q., Brown, M. E., Zheng, T., You, R., Diehl, T., Ginoux, P., and Kucsera, T.: The effect of the dynamic surface bareness on dust source function, emission, and distribution, J. Geophys. Res.-Atmos., 118, 871–886, 2013.

Kopke, P., Hess, M., Schult, I., and Shettle, E. P.: Global Aerosol Data Set, Max Planck Inst. für Meteorol., Hamburg, Germany, 1997.

Koren, I., Feingold, G., and Remer, L. A.: The invigoration of deep convective clouds over the Atlantic: aerosol effect, meteorology or retrieval artifact?, Atmos. Chem. Phys., 10, 8855–8872, doi:10.5194/acp-10-8855-2010, 2010.

5

10

Koven, C. D. and Fung, I.: Identifying global dust source areas using high-resolution land surface form, J. Geophys. Res.-Atmos., 113, D22204, doi:10.1029/2008JD010195, 2008.

Levy, R. C., Remer, L. A., Kleidman, R. G., Mattoo, S., Ichoku, C., Kahn, R., and Eck, T. F.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmos. Chem. Phys., 10, 10399–10420, doi:10.5194/acp-10-10399-2010, 2010.

Liao, H., Henze, D. K., Seinfeld, J. H., Wu, S. L., and Mickley, L. J.: Biogenic secondary organic aerosol over the United States: comparison of climatological simulations with observations, J.

- Geophys. Res.-Atmos., 112, D06201, doi:10.1029/2006JD007813, 2007.
 Marsham, J. H., Knippertz, P., Dixon, N. S., Parker, D. J., and Lister, G. M. S.: The importance of the representation of deep convection for modeled dust-generating winds over West Africa during summer, Geophys. Res. Lett., 38, L16803, doi:10.1029/2011GL048368, 2011.
- Martin, R. V., Jacob, D. J., Yantosca, R. M., Chin, M., and Ginoux, P.: Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, J. Geophys. Res.-Atmos., 108, 4097, doi:10.1029/2002JD002622, 2003.
 - Mbourou, G. N., Bertrand, J. J., and Nicholson, S. E.: The diurnal and seasonal cycles of windborne dust over Africa north of the equator, J. Appl. Meteorol., 36, 868–882, 1997.
- McGee, D., Broecker, W. S., and Winckler, G.: Gustiness: the driver of glacial dustiness?, Quaternary Sci. Rev., 29, 2340–2350, 2010.
 - McGee, D., deMenocal, P. B., Winckler, G., Stuut, J. B. W., and Bradtmiller, L. I.: The magnitude, timing and abruptness of changes in North African dust deposition over the last 20 000 yr, Earth Planet. Sc. Lett., 371–372, 163–176, 2013.
- Mian Chin, Diehl, T., Tan, Q., Prospero, J. M., Kahn, R. A., Remer, L. A., Yu, H., Sayer, A. M., Bian, H., Geogdzhayev, I. V., Holben, B. N., Howell, S. G., Huebert, B. J., Hsu, N. C., Kim, D., Kucsera, T. L., Levy, R. C., Mishchenko, M. I., Pan, X., Quinn, P. K., Schuster, G. L., Streets, D. G., Strode, S. A., Torres, O., and Zhao, X.-P.: Multi-decadal variations of atmo-



spheric aerosols from 1980 to 2009: sources and regional trends, Atmos. Chem. Phys. Discuss., 13, 19751–19835, doi:10.5194/acpd-13-19751-2013, 2013.

- Miller, R. L., Tegen, I., and Perlwitz, J.: Surface radiative forcing by soil dust aerosols and the hydrologic cycle, J. Geophys. Res., 109, D04203, doi:10.1029/2003JD004085, 2004.
- Moulin, C., Lambert, C. E., Dulac, F., and Dayan, U.: Control of atmospheric export of dust from North Africa by the North Atlantic oscillation, Nature, 387, 691–694, 1997.
 - Mulitza, S., Heslop, D., Pittauerova, D., Fischer, H. W., Meyer, I., Stuut, J.-B., Zabel, M., Mollenhauer, G., Collins, J. A., Kuhnert, H., and Schulz, M.: Increase in African dust flux at the onset of commercial agriculture in the Sahel region, Nature, 466, 226–228, 2010.
- Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J.-H., Zhang, K., Zhang, H., and Zhou, C.; Radiative forcing of the direct aerosol effect from AeroCom Phase
- ¹⁵ Zhang, H., and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, Atmos. Chem. Phys., 13, 1853–1877, doi:10.5194/acp-13-1853-2013, 2013.
 Nakamae, K. and Shiotani, M.: Interannual variability in Saharan dust over the North Atlantic Ocean and its relation to meteorological fields during northern winter, Atmos. Res., 122, 336–346, 2013.
- ²⁰ Olsson, L., Eklundh, L., and Ardö, J.: A recent greening of the Sahel trends, patterns and potential causes, J. Arid Environ., 63, 556–566, 2005.
 - Park, R. J., Jacob, D. J., Chin, M., and Martin, R. V.: Sources of carbonaceous aerosols over the United States and implications for natural visibility, J. Geophys. Res.-Atmos., 108, 4355, doi:10.1029/2002JD003190, 2003.
- Park, R. J., Jacob, D. J., Field, B. D., Yantosca, R. M., and Chin, M.: Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: implications for policy, J. Geophys. Res.-Atmos., 109, D15204, doi:10.1029/2003JD004473, 2004.
 Patterson, E. M., Gillette, D. A., and Stockton, B. H.: Complex index of refraction between 300 and 700 nm for Saharan aerosols, J. Geophys. Res., 82, 3153–3160, 1977.
- Prospero, J. M.: Long-term measurements of the transport of African mineral dust to the Southeastern United States: implications for regional air quality, J. Geophys. Res.-Atmos., 104, 15917–15927, 1999.



- Prospero, J. M. and Lamb, P. J.: African droughts and dust transport to the Caribbean: climate change implications, Science, 302, 1024–1027, 2003.
- Prospero, J. M., Glaccum, R. A., and Nees, R. T.: Atmospheric transport of soil dust from Africa to South-America, Nature, 289, 570–572, 1981.
- ⁵ Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Gill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 1002, doi:10.1029/2000RG000095, 2002.

Prospero, J. M., Collard, F. X., Molinié, J., and Jeannot, A.: Characterizing the annual cycle

of African dust transport to the Caribbean Basin and South America and its impact on air quality and the environment, Global Biogeochem. Cy., submitted, 2014.

Ridley, D. A., Heald, C. L., and Ford, B.: North African dust export and deposition: a satellite and model perspective, J. Geophys. Res., 117, D02202, doi:10.1029/2011JD016794, 2012.
Ridley, D. A., Heald, C. L., Pierce, J. R., and Evans, M. J.: Toward resolution-independent

- dust emissions in global models: impacts on the seasonal and spatial distribution of dust, Geophys. Res. Lett., 40, 2873–2877, 2013.
 - Riemer, N., Doherty, O. M., and Hameed, S.: On the variability of African dust transport across the Atlantic, Geophys. Res. Lett., 33, L13814, doi:10.1029/2006GL026163, 2006.

Rotstayn, L. D. and Lohmann, U.: Tropical rainfall trends and the indirect aerosol effect, J. Climate, 15, 2103–2116, 2002.

20

30

Savoie, D. L., Prospero, J. M., and Saltzman, E. S.: Non-sea-salt sulfate and nitrate in trade wind aerosols at Barbados: evidence for long-range transport, J. Geophys. Res.-Atmos., 94, 5069–5080, 1989.

Schepanski, K., Tegen, I., Laurent, B., Heinold, B., and Macke, A.: A new Saharan dust source

activation frequency map derived from MSG-SEVIRI IR-channels, Geophys. Res. Lett., 34, L18803, doi:10.1029/2007GL030168, 2007.

Schepanski, K., Tegen, I., and Macke, A.: Comparison of satellite based observations of Saharan dust source areas, Remote Sens. Environ., 123, 90–97, 2012.

Shao, Y., Klose, M., and Wyrwoll, K.-H.: Recent global dust trend and connections to climate forcing, J. Geophys. Res.-Atmos., 118, 11107–11118, 2013.

Sinyuk, A., Torres, O., and Dubovik, O.: Combined use of satellite and surface observations to infer the imaginary part of refractive index of Saharan dust, Geophys. Res. Lett., 30, 1081, doi:10.1029/2002GL016189, 2003.



- Stephenson, D. B., Pavan, V., and Bojariu, R.: Is the North Atlantic Oscillation a random walk?, Int. J. Climatol., 20, 1–18, 2000.
- Stier, P., Schutgens, N. A. J., Bellouin, N., Bian, H., Boucher, O., Chin, M., Ghan, S., Huneeus, N., Kinne, S., Lin, G., Ma, X., Myhre, G., Penner, J. E., Randles, C. A., Sam-
- set, B., Schulz, M., Takemura, T., Yu, F., Yu, H., and Zhou, C.: Host model uncertainties in aerosol radiative forcing estimates: results from the AeroCom Prescribed intercomparison study, Atmos. Chem. Phys., 13, 3245–3270, doi:10.5194/acp-13-3245-2013, 2013.

Stocker, T. F., Qin, D., Plattner, G. K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P. M., n.d. IPCC: Summary for policymakers, in: Climate Change 2013: the

- ¹⁰ Physical Science Basis, Cambridge University Press, Cambridge, UK and New York, NY, USA, 2013.
 - Tegen, I., Schepanski, K., and Heinold, B.: Comparing two years of Saharan dust source activation obtained by regional modelling and satellite observations, Atmos. Chem. Phys., 13, 2381–2390, doi:10.5194/acp-13-2381-2013, 2013.
- ¹⁵ Twohy, C. H., Kreidenweis, S. M., Eidhammer, T., Browell, E. V., Heymsfield, A. J., Bansemer, A. R., Anderson, B. E., Chen, G., Ismail, S., DeMott, P. J., and Van Den Heever, S. C.: Saharan dust particles nucleate droplets in eastern Atlantic clouds, Geophys. Res. Lett., 36, L01807, doi:10.1029/2008GL035846, 2009.

Vautard, R., Cattiaux, J., Yiou, P., Thépaut, J.-N., and Ciais, P.: Northern Hemisphere atmo-

- ²⁰ spheric stilling partly attributed to an increase in surface roughness, Nat. Geosci., 3, 756– 761, 2010.
 - Viana, M., Querol, X., Alastuey, A., Cuevas, E., and Rodrìguez, S.: Influence of African dust on the levels of atmospheric particulates in the Canary Islands air quality network, Atmos. Environ., 36, 5861–5875, 2002.
- ²⁵ Wang, C., Dong, S., Evan, A. T., Foltz, G. R., and Lee, S.-K.: Multidecadal covariability of North Atlantic sea surface temperature, African dust, Sahel rainfall, and Atlantic hurricanes, J. Climate, 25, 5404–5415, 2012.

30

Williams, K. D., Jones, A., Roberts, D. L., Senior, C. A., and Woodage, M. J.: The response of the climate system to the indirect effects of anthropogenic sulfate aerosol, Clim. Dynam., 17, 845–856. 2001.

Yoshioka, M., Mahowald, N. M., Conley, A. J., Collins, W. D., Fillmore, D. W., Zender, C. S., and Coleman, D. B.: Impact of desert dust radiative forcing on Sahel precipitation: relative



fall and Atlantic hurricanes, Geophys. Res. Lett., 33, L17712, doi:10.1029/2006GL026267,

Zhang, R. and Delworth, T. L.: Impact of Atlantic multidecadal oscillations on India/Sahel rain-

importance of dust compared to sea surface temperature variations, vegetation changes,

Zender, C. S., Bian, H. S., and Newman, D.: Mineral Dust Entrainment and Deposition (DEAD) model: description and 1990s dust climatology, J. Geophys. Res.-Atmos., 108, 4416,

Zender, C. S., Newman, D., and Torres, O.: Spatial heterogeneity in aeolian erodibility: uniform,

topographic, geomorphic, and hydrologic hypotheses, J. Geophys. Res.-Atmos., 108, 4543,

and greenhouse gas warming, J. Climate, 20, 1445–1467, 2007.

Zhao, T. X.-P., Laszlo, I., Guo, W., Heidinger, A., Cao, C., Jelenak, A., Tarpley, D., and Sullivan, J.: Study of long-term trend in aerosol optical thickness observed from operational AVHRR satellite instrument, J. Geophys. Res.-Atmos., 113, D07201,

doi:10.1029/2007JD009061.2008. 15

doi:10.1029/2002JD002775, 2003a.

doi:10.1029/2002JD003039. 2003b.

5

10

2006.



3614





Fig. 1. Dust emissions averaged over the period 1982–2008 are shown for the region of interest based on **(a)** the GOCART dust source map, the default in GEOS-Chem, and **(b)** the newly implemented source map, derived from levelness and roughness surface properties (Koven et al., 2008) and responding to dynamic vegetation cover derived from the NDVI product from AVHRR.



Fig. 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.





Fig. 3. AOD averaged over 2004 to 2008 for winter (DJFM) and summer (AMJJAS) is shown for MODIS, the original model and the model with updated dust scheme. Average AERONET AOD for all available data is displayed in the circles. Both versions of the model have the same total dust emissions and identical emissions for all other aerosol. Regression coefficient (R), slope (M) and bias (B) with MODIS are indicated for both models.





Fig. 4. Daily AOD between 2004 and 2008 from AERONET and the model (all aerosol) with updated dust scheme are displayed for 8 AERONET sites in Africa. Days during the winter and summer seasons are shown as crosses and diamonds, respectively, and grey shading indicates the 2 : 1 boundaries. Regression coefficient (R), slope (M), RMS error (E) and number of data points (N) are displayed for winter (blue) and summer (red).





Fig. 5. Climatological (1982–2008) monthly surface dust concentrations are displayed for **(a)** observations made at Ragged Point in Barbados, **(b)** the baseline GEOS-Chem model, and **(c)** the GEOS-Chem model with sub-grid wind parameterization and Koven dust source map. The black line indicates the monthly mean and the red and orange shading corresponds to the 25th to 75th and 5th to 95th quantile ranges, respectively.





Fig. 6. Seasonal dust AOD anomalies derived from AVHRR PATMOS-x (black) and modeled (grey) 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.





Fig. 7. Seasonally-averaged surface concentrations from Barbados (black) and modeled (grey) 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.





Fig. 8. Attribution of the inter-annual 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).





Fig. 9. Annual dust AOD (DAOD) anomalies are shown for the updated model (grey, same as the model in Fig. 6), with interannual variability in 10 m winds removed (blue), with interannual variability in all meteorology, except surface winds, removed (red), 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 grey line is almost always obscured by the green line.







Fig. 10. 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 10 m wind speed. Black contours outline regions where emissions decrease by more than 0.5 Tg per grid box.





Fig. 11. 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.



Fig. 12. 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 $0-30^{\circ}$ N, 50° W– 15° E. The thin black line indicates the trend over the simulation period.





Fig. 13. 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.

