

Supplement of Atmos. Chem. Phys., 14, 5735–5747, 2014
<http://www.atmos-chem-phys.net/acp-14-5735-2014/>
doi:10.5194/acp-14-5735-2014-supplement
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Supplement of

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

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1 **Supplementary Material**

2

3 **Climatological evaluation of GEOS-Chem**

4 **Observational datasets**

5 The MODIS instrument provides near daily coverage of the globe and AOD retrievals at 550 nm
6 that are well validated against AERONET observations (Levy et al., 2010). The MODIS Deep Blue
7 retrieval is used to estimate AOD over bright surfaces (Hsu et al., 2004), providing complementary
8 observations in regions not covered by AERONET in the Sahara. In this study we use daily Level 3
9 MODIS observed AOD from the Aqua satellite (13:30 equatorial overpass, Collection 5) with the
10 Deep Blue retrieval where no standard retrieval is available.

11 The Aerosol Robotic Network (AERONET) is a well-established global network of sun photometers
12 used to derive aerosol optical depth (AOD) at a set of standard wavelengths (440, 675, 870 and
13 1020 nm) on a daily basis (Holben et al., 1998). Almuqantar measurements (a series of
14 measurements taken for multiple azimuth angles at the elevation of the sun) allow size and
15 scattering information to be estimated from the retrieved extinction, producing a measure of the
16 contribution of fine (< 1 μ m) and coarse aerosol to the AOD (Dubovik et al., 2002). In this study,
17 we use daily AOD retrievals from eight AERONET sites across West Africa (locations shown on Fig.
18 2) between 2004 and 2008 to evaluate the model representation of AOD close to source.

19

20 **Evaluation of GEOS-Chem dust scheme over North Africa**

21 Several previous studies have assessed the skill of the DEAD dust scheme in the GEOS-Chem
22 model and show reasonable agreement with satellite and surface observations both close to
23 source and downwind (Duncan Fairlie et al., 2007; Generoso et al., 2008; Johnson et al., 2010;

1 Ridley et al., 2012). Given the model alterations described in Sect. 2 (inclusion of a sub-grid wind
2 parameterization, a new source map and dynamic vegetation), we re-evaluate the model for
3 North Africa using AERONET observations and MODIS retrievals close to source and surface
4 concentrations downwind in Barbados. We use a five year simulation (2004 - 2008) with all
5 aerosol components included to allow for direct comparison with AOD observations from MODIS
6 and AERONET.

7 Figure S1 compares the normalized dust emissions for both the GOCART and geomorphic source
8 maps averaged over the period 2004 – 2008, both scaled to give the same total emissions for the
9 region. The key source regions of the Bodélé Depression and in Central Sahara are well
10 represented in both, with more weighting given to the former when using the geomorphic source
11 map. Relative to the potential source regions outlined in (Formenti et al., 2011), five of the six
12 regions are represented in the current dust scheme; however, emissions from the region at the
13 Mali – Niger border (20°N, 5°E) appear weak in both dust schemes. Another key difference is that
14 the area from which emissions can occur is broader in the geomorphic source map, with potential
15 emissions extending further south if vegetation cover permits. The dust emission frequency
16 derived from SEVIRI satellite observations by Schepanski et al. (2007) indicates that emissions are
17 possible from a broader region, in agreement with the geomorphic source map. While it has been
18 shown that geomorphic source maps can lead to erroneous dust emissions in the US as a result
19 of the vegetation-dependence (Fairlie et al., 2007; Zender et al., 2003), comparison between the
20 model and observations in Africa suggests that the new source function performs at least as well
21 as the original GOCART source function in this region.

22

1 Figure S2 compares the seasonal AOD (averaged over 2004 – 2008) observed by MODIS to the
2 GEOS-Chem simulation with the original and updated dust scheme. The spatial correlation
3 between model and MODIS coarse AOD over the dust source region in North Africa ($10^{\circ}\text{N} - 36^{\circ}\text{N}$,
4 $22.5^{\circ}\text{W} - 32.5^{\circ}\text{E}$) is slightly improved in all five years when using the updated dust scheme, from
5 0.72 to 0.78 in winter and from 0.63 to 0.67 in summer. This improvement is largely the result of
6 the reduction in emissions from the West Coast relative to those from the Bodélé Depression
7 produced by the new source function and vegetation modulation. However, the fraction of
8 modeled AOD at the Bodélé Depression is still low relative to the observations; this contributes
9 to the regional model AOD low biases of 21% and 38% in winter and summer, respectively. We
10 find that suppression of emissions by soil moisture and vegetation is limited in the Bodélé region,
11 suggesting that MERRA surface winds are too weak to generate the large flux of dust observed,
12 particularly in August and September. Biomass burning aerosol below approximately 12°N during
13 the winter and sea salt aerosol in coastal regions may both influence the agreement with MODIS
14 and AERONET, but we expect these effects to be small relative to the dust aerosol that accounts
15 for over 70% of the annual AOD between 10°N and 36°N in the model, also observed in (Formenti
16 et al., 2008).

17 Figure S3 shows comparison of daily measurements at eight AERONET sites with the updated
18 model AOD (including non-dust species) during the period 2004 – 2008. The temporal correlation
19 averaged across the sites (and weighted by the number of observations) increases from 0.75 to
20 0.78 during winter, and increases from 0.66 to 0.72 in summer when using the updated dust
21 scheme. The RMS error relative to AERONET is reduced during the summer at all eight sites but
22 worsened slightly at Sahelian sites during the winter using the updated dust scheme. We find

1 that including the sub-grid wind PDF reduces the RMS error across almost all sites and seasons
2 but the inclusion of the new source function and vegetation modulation masks this improvement
3 in winter, suggesting that wintertime source regions are now over-represented in the Sahel
4 (excluding the Bodélé Depression). While the total improvement relative to the observations is
5 small, the new dust emission scheme should be capturing more processes as it represents both
6 sub-grid winds and the modulation of dust emissions from vegetation changes. General
7 agreement with daily observations from the AERONET sites is good, with the majority of days
8 falling within a factor of 2 of the observations and seasonal correlation coefficients greater than
9 $r = 0.6$ for all but one location. In the Sahel, there is a tendency for the model to overestimate
10 the AOD during high aerosol loading (predominantly in winter) and underestimate the AOD in
11 summer, potentially from poor representation of dust emissions driven by local convective storm
12 down drafts, i.e. Haboobs (Marsham et al., 2011), or from poor representation of wind gustiness
13 and therefore a bias towards emission from synoptic air flow in the wintertime. Attribution of
14 the wintertime overestimation is confounded by the presence of biomass burning aerosol. In
15 addition, the AERONET sites are clustered in the Sahel biasing the comparison to this particular
16 region and underrepresenting the Sahara. Tamanrasset, located in the Sahara but perhaps not
17 representing the area well (Cuesta et al., 2008), indicates a similar picture to the Sahel with the
18 model AOD biased high during winter and low during summer.

19 Figure 5 shows the comparison of both the original and updated dust model with a monthly
20 climatology of surface dust concentrations at the Barbados site over the period 1982 - 2008. The
21 observed climatology shows a peak in the average concentration in June ($30 \mu\text{gm}^{-3}$) and a
22 minimum in December ($7.5 \mu\text{gm}^{-3}$), with greatest interannual variability between March and

1 June. The updated source map shifts the peak in modeled concentration from July to June, in
2 better agreement with the observations, and also improves the seasonality by increasing the
3 summertime peak average concentration from $21.0 \mu\text{gm}^{-3}$ to $26.0 \mu\text{gm}^{-3}$ and decreasing the
4 wintertime average concentration slightly.

5 We also investigated the possibility of using the SEVIRI dust source activation frequency (DSAF)
6 to constrain the emissions in the model. We scaled the wind threshold for emission such that the
7 model produced the best fit with the number of dust event days per month, derived from four
8 years of SEVIRI observations (Schepanski et al., 2007). While the wind threshold scaling boosted
9 emissions from the Bodélé Depression during summer and reduced them in coastal regions,
10 which would be expected to improve the model based on Fig. 3, the improvement to the model
11 dust AOD was minimal when compared with both MODIS and AERONET. This suggests that the
12 efficacy of DSAF for improving the simulation of dust may be limited by the inability to consider
13 the size of the dust event, in agreement with (Tegen et al., 2013). Scaling the wind threshold for
14 emission may be masking errors in MERRA surface winds rather than providing useful
15 information on surface susceptibility to emission; therefore, we have chosen not to include the
16 DSAF scaling in the simulations used in this study.

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

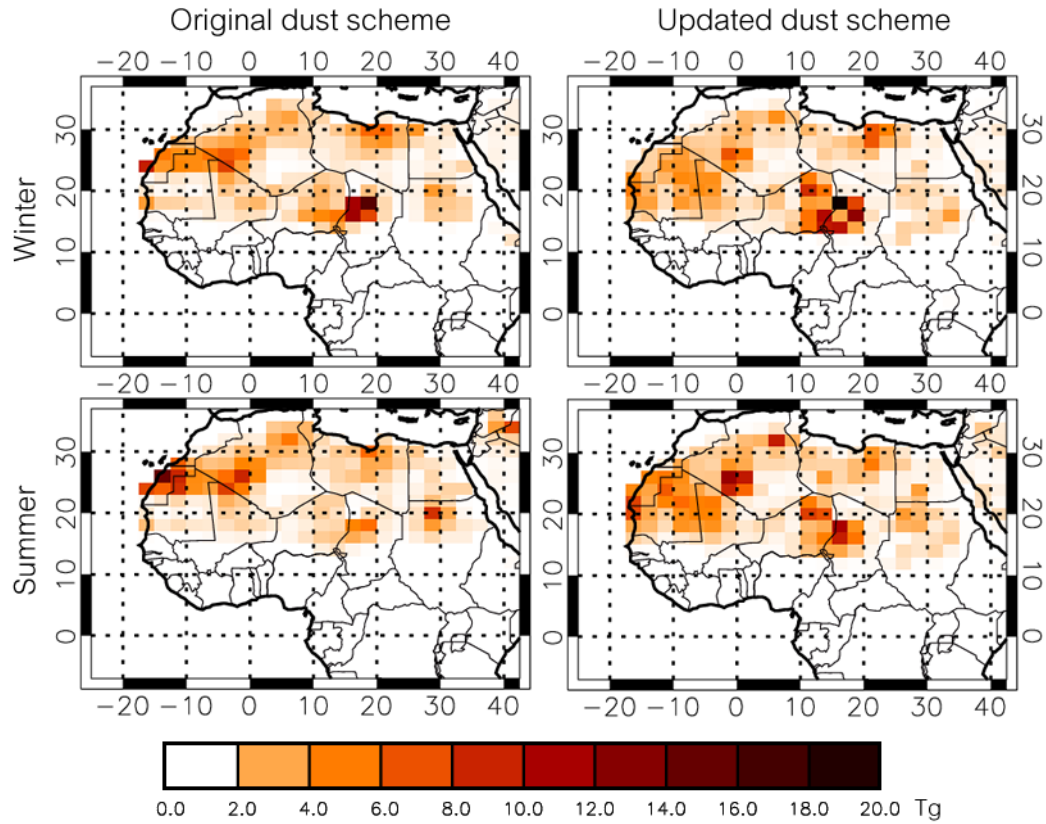
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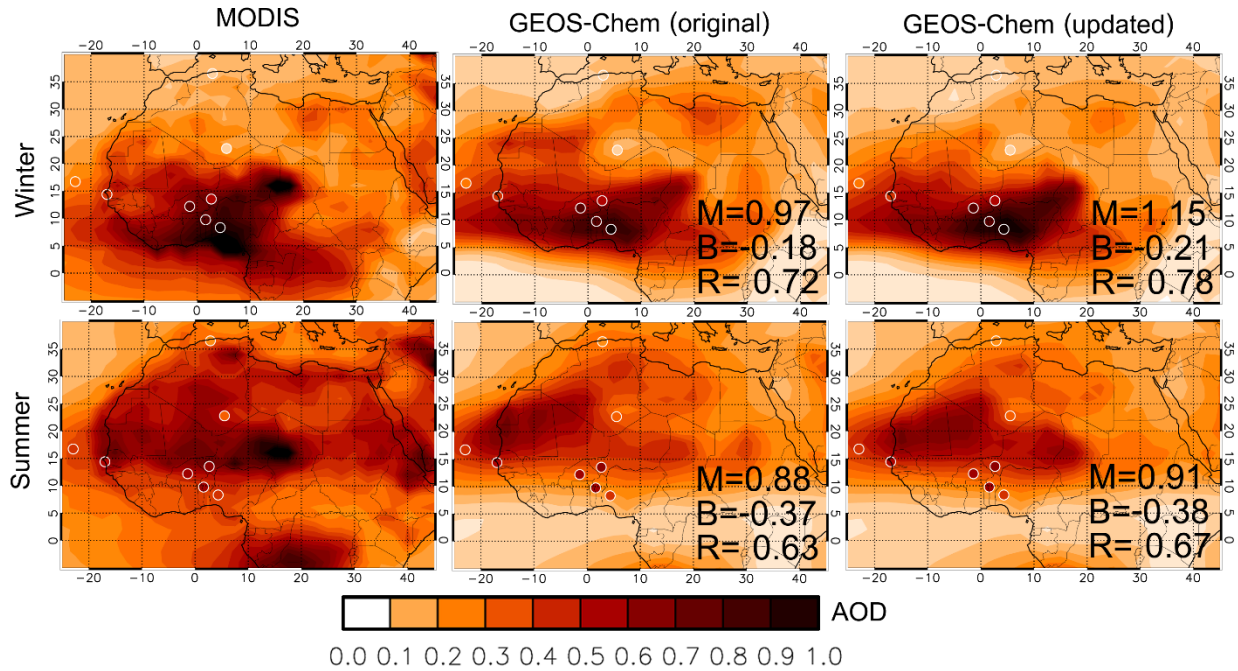
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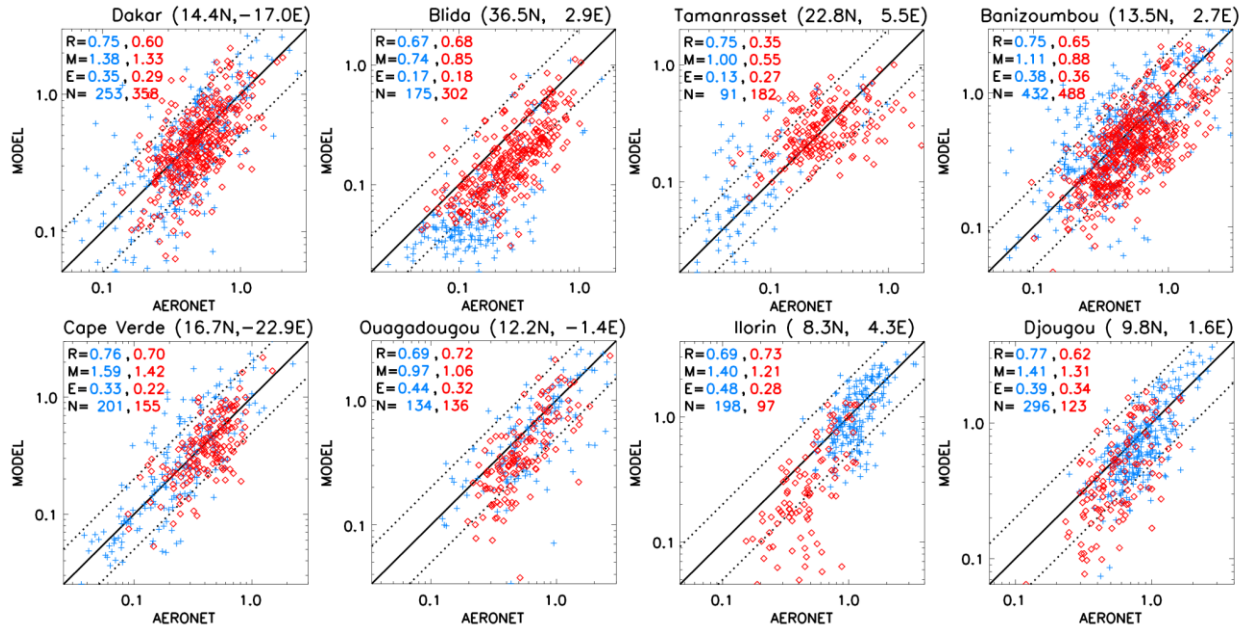
2 Figure S1 – Dust emissions averaged over the period 1982 – 2008 are shown for the region of
 3 interest based on (a) the GOCART dust source map, the default in GEOS-Chem, and (b) the newly
 4 implemented source map, derived from levelness and roughness surface properties (Koven et al.,
 5 2008) and responding to dynamic vegetation cover derived from the NDVI product from AVHRR.

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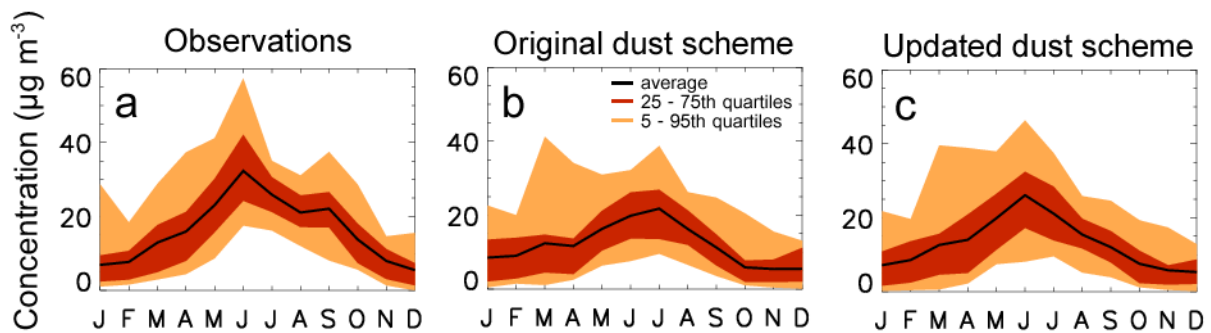


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2 Figure S2 – AOD averaged over 2004 to 2008 for winter (DJFM) and summer (AMJJAS) is shown
 3 for MODIS, the original model and the model with updated dust scheme. Average AERONET AOD
 4 for all available data is displayed in the circles. Both versions of the model have the same total
 5 dust emissions and identical emissions for all other aerosol. Regression coefficient (R), slope (M)
 6 and bias (B) with MODIS are indicated for both models.



1
 2 Figure S3 – Daily AOD between 2004 and 2008 from AERONET and the model (all aerosol) with
 3 updated dust scheme are displayed for 8 AERONET sites in Africa. Days during the winter and
 4 summer seasons are shown as blue crosses and red diamonds, respectively, the solid line
 5 represents the 1:1 agreement and dashed lines indicate the 2:1 boundaries. Regression
 6 coefficient (R), slope (M), RMS error (E) and number of data points (N) are displayed for winter
 7 (blue) and summer (red).



1 Figure S4 – Climatological (1982 – 2008) monthly surface dust concentrations are displayed for
2 (a) observations made at Ragged Point in Barbados, (b) the baseline GEOS-Chem model, and (c)
3 the GEOS-Chem model with sub-grid wind parameterization and Koven dust source map. The
4 black line indicates the monthly mean and the red and orange shading corresponds to the 25th
5 to 75th and 5th to 95th quantile ranges, respectively.

6