Response to the reviewer' comments

First, we wish to thank the referees for their beneficial comments that helped us improve the manuscript. Also, we are pleased that the reviewers found the paper interesting and well written. We begin with a general comment, referring to an important point raised by the reviewers regarding the separation between dust and biomass smoke aerosol during the boreal winter. Following this part are specific answers to all of the reviewer's comments point by point.

Preamble to specific responses

Both reviewers questioned the separation of dust and biomass smoke during the winter season. Indeed, the algorithm that estimates the Aerosol Optical Depth (AOD) of dust component (τ_d) using the total AOD (τ), the fine mode fraction (f) and the surface winds is an approximation that is not perfect for all specific cases. Dust and biomass smoke aerosols have a range of size distributions and this algorithm uses an approximation that was developed and tuned for the average values. Following the reviewers concerns we have conducted a sensitivity analysis to find the range of the possible errors in τ_d due to error in the fine mode fraction attributed to biomass smoke aerosol (f_s):

Recent field experiments and remote sensing studies proposed that f_s is near unity (e.g.: Johnson et al., 2008b; Capes et al., 2008; Eck et al., 2010). The value we used for this work is 0.9, based on studies by Kaufman et al., (2005a) and Yu at al., (2009) (and the reference therein). To the best of our knowledge, there are no studies suggesting that f_s can be smaller than 0.9. Therefore, to estimate the sensitivity of τ_d separation algorithm and to set bound on the possible error we re-ran the calculations with f_s of unity (meaning, no contribution to the coarse mode by biomass smoke aerosol) and compared the results to our original results (Fig. 1a and b and Fig. 2).

The sensitivity analysis during the boreal winter yields an estimated bias of less than a percent and standard deviation of ~18% in the τ_d due to biomass smoke contamination. Since the distribution of the differences in the τ_d estimations is sharper than a normal distribution, our sensitivity study suggests that more than 85% of the results are bounded within an error of ± 18%. In addition, Fig. 2 shows that both time series of τ_d (using f_s equal 1 or 0.9) over the ocean are very similar, with very minor differences.

Note that the dominance of dust in winter mixed plumes of dust and biomass smoke was described in previous studies. Formenti et al. (2008), for example, proposed that mineral dust was accounting for 72% of the estimated aerosol mass in aged elevated biomass burning layers, 91% in fresh biomass burning and up to 93% in plumes of mineral dust.

Moreover, insofar as our main results rely on the temporal and spatial signature of τ_d and the cross-correlation analysis, the biomass smoke contamination of the dust signal is somewhat circumvented by the use of these different tools. In particular, the spatial correlation between dust emitted from a specific source (e.g. Bodélé) and the plumes crossing the ocean would be greatly weakened by the spatially random biomass smoke contamination, if the fraction of the latter be significant. This is especially noteworthy for the time-lag of the cross-correlation which so nicely fits the duration of transatlantic transit.

Specific parts regarding this subject were added to the revise manuscript in the following sections:

<u>a. In the introduction (page 7 lines 7-14):</u>

"Results from recent field experiments show that the characteristic vertical structure of the aerosol column is dust in lower altitudes, up to ~2 km, and a mixed dust and biomass smoke layer at the upper few kilometers (Formenti et al., 2008; Johnson et al., 2008a; Ansmann et al., 2011; Knippertz et al., 2011; Weinzierl et al., 2011). They also show the dominance of dust in those winter plumes: mineral dust contributes 72% of the aerosol mass in aged elevated biomass burning layers, 91% in fresh biomass burning layers and up to 93% in plumes of mineral dust (Formenti et al., 2008)".

b. In the data section (page 9 line 14- page 10 line 12):

"The expected error in derivation of τ_d increases during the Sahelian biomass burning season (December to February), when the dust transport route passes over the biomass burning region and the dust is mixed with biomass smoke (e.g.: Formenti et al., 2008; Weinzierl et al., 2011).

Recent field experiments and remote sensing studies proposed that the biomass smoke fine mode fraction is near unity (e.g.: Johnson et al., 2008b; Capes et al., 2008; Eck et al., 2010). The value we used in this work is 0.9, based on studies by Kaufman et al., (2005a) and Yu at al., (2009) and the reference therein. To the best of our knowledge, there are no studies suggesting that biomass smoke fine mode fraction can be smaller than 0.9. Therefore, to estimate the sensitivity of τ_d separation algorithm and to bound the possible error we re-ran the calculations with biomass smoke fine mode fraction of unity (meaning, no contribution to the coarse mode by biomass smoke aerosol) and compared the results to our original results. The sensitivity analysis during the boreal winter yields an estimated bias of less than a percent and standard deviation of ~18% in the τ_d due to biomass smoke contamination. Since the distribution of the differences in τ_d estimations is sharper than a normal distribution, our sensitivity study suggests that more than 85% of the results are bounded within an error of ± 18%. Additional possible source of error in our analysis can be an overestimation of τ by ~0.02 due to cloud-contamination (Kaufman et al., 2005b). Nevertheless, by averaging τ_d over large area and focusing on the low frequencies of the annual dust's cycle, we expect the above errors to be insignificant".

c. In the discussion (page 19 lines 9-16):

"This correlation analysis is an additional evidence for the preponderance of dust in winter plumes. The high correlation between a specific source (e.g. Bodélé) and the plumes arriving at the middle of the ocean should be obscured by a spatially random biomass smoke contamination, if the fraction of the latter be significant. This is particularly noteworthy for the 3-5 days time lag of the cross-correlation which so nicely fits the duration of transatlantic transit. Thus, insofar as our main results rely on the cross-correlation analysis, the biomass smoke contamination of the dust signal is somewhat circumvented and constrained by the temporal signature".

Specific answers to referee #1

1. I would suggest that they include four panels representing four seasons to show seasonal variations of dust AOD. This would help readers better understand Figure 2.

Answer

The four panels were inserted into the revised manuscript as requested (Fig. 3a-d here and Fig. 1a-d in the revised manuscript).

2. I notice that the dust AOD in the Gulf of Guinea is impressively high. I assume this high dust AOD occurs predominantly in winter (or during the southern-route period). Given that smoke is often mixed with dust (e.g., with smoke over dust as observed during AMMA and DABEX) in this region, can the high dust AOD partly reflect difficulty and uncertainty of using MODIS total AOD and fine-mode fraction to derive dust AOD? How does this uncertainty affect your results about the characteristics of the southern route? They may consider of adding some discussions about this uncertainty.

Answer

We consider these comments critically important and therefore answered them in details in the opening (preamble to specific responses).

We will add here that in cases of pure biomass smoke plumes, such as in the case near 10º S and 10º E during the boreal summer (Fig. 1c), when using one average value for biomass smoke an additive error can accumulate. For that reason we did not include this zone in our area of interest and as shown in the opening statement we estimate the possible error around 18% with little bias. Thanks to this comment, this issue is now mentioned in the revised manuscript in several places, as mentioned in the opening statement.

3. Figure 2(b): It is not clear to me what they are referring to by saying “the apparent line near 16W ”. What does X-axis (longitude of 20 , 40, 60 degree) really mean?

<u>Answer</u>

The Hovmöller diagram (Fig. 3b in the revised manuscript) averges the AOD of dust over the study area for all longitudes per day. Therefore the X axis in this Fig. is longitudinal degrees along the study area.

During the northern-route period (NRP) the dust is emitted to the Atlantic mostly from the Saharan coast, crossing the west shoreline of Africa over longitude ~16° W. During the southern-route period (SRP) the dust is emitted from an eastern (and southern) position, over the Gulf of Guinea. Since our study area is only over the ocean, the NRP months in the Hovmöller diagram present a combination of relatively clean conditions over the eastern part of the ocean (the southern regions of the study area, over the Gulf of Guinea, where there is no emission of dust during the summer) and dusty conditions on the western part of the ocean. It creates a line in the NRP months in the Hovmöller diagram along longitude ~16° W (Fig 3b in the revised manuscript).

Following the reviewer's question, the following paragraph was added to the manuscript in page 12 lines 4-8:

"Focusing on oceanic regions only and averaging for all latitudes of the study area creates apparent discontinuity. During the NRP the dust arrives the Atlantic Ocean from the Saharan coast which is located in a western position compared to the Gulf of Guinea. Therefore during this season (NRP) relatively clean ocean is averaged over the Gulf. This creates the apparent discontinuity in the dust loading east-west gradient".

4. p.23517, line 1: “(Sundar et al., 2010)” should be “(Christopher and Jones, 2010)”. Also the full citation in the reference list should be corrected.

Answer

The citation and the reference were corrected.

5. p.23519, line 9-10: why may the dust AOD be under or over estimated in occasions of high or low dust loading, respectively? Please explain.

Answer

We assume that there are occasions when the prevalent conditions of aerosol, as estimated in our algorithm, fail to describe the true optical properties of the observed aerosol. For example, when $\tau \gg 1$ we expect to underestimate τ_d. Thanks to this comment the unclear sentence was clarified in the revised manuscript in page 9 lines 9-13 as follows:

"Note that this algorithm for extracting τ_d is based on some assumptions regarding the prevalent conditions of dust, maritime and anthropogenic aerosol loading that were estimated over specific regions where each type of aerosol is concentrated. Since the prevalent conditions represent average aerosol loading, we expect that τ_d may be under (over) estimated on occasions of high (low) dust loading".

References

1. Ansmann, A., Petzold, A., Kandler, K., Tegen, I., Wendisch, M., D Muller, Weinzierl B., Muller, T., and Heintzenberg, J.: Saharan Mineral Dust Experiments SAMUM-1 and SAMUM-2: what have we learned?, Tellus 63B, 403–429, doi: 10.1111/j.1600-0889.2011.00555.x, 2011.

2. Capes, G., Johnson, B., McFiggans, G., Williams, P. I., Haywood J., and Coe, H.: Aging of biomass burning aerosols over West Africa: Aircraft measurements of chemical composition, microphysical properties, and emission ratios, Journal of Geophysical Research (Atmos.), 113, D00C15, doi:10.1029/2008JD009845, 2008.

3. Eck, T. F., Holben, B. N., Sinyuk, A., Pinker, R. T., Goloub, P., Chen, H., Chatenet, B., Li, Z., Singh, R. P., Tripathi, S. N., Reid, J. S., Giles, D. M., Dubovik, O., O'Neill, N. T. Smirnov, A. Wang, P., and Xia, X.: Climatological aspects of the optical properties of fine/coarse mode aerosol mixtures, Journal of Geophysical Research, 115, D19205, doi:10.1029/2010JD014002, 2010.

4. Formenti, P., Rajot, J. L., Desboeufs, K., Caquineau, S., Chevaillier, S., Nava, S., Gaudichet, A., Journet, E., Triquet, S., Alfaro, S., Chiari, M., Haywood, J. M., Coe, H., and Highwood, E. J.: Regional variability of the composition of mineral dust from Western Africa: results from the AMMA SOP0/DABEX and DODO, Journal of Geophysical Research, 113, D00C13, doi:10.1029/2008JD009903, 2008.

5. Johnson, B. T., Heese, B., McFarlane, S. A., Chazette, P., Jones, A., and Bellouin, N.: Vertical distribution and radiative effects of mineral dust and biomass burning aerosol over west Africa during DABEX, Journal of Geophysical Research, 113, D00C12, doi:10.1029/2008JD009848, 2008a.

6. Johnson, B. T., Osborne, S. R., Haywood, J. M., and Harrison, M. A. J.: Aircraft measurements of biomass burning aerosol over West Africa during

DABEX, Journal of Geophysical Research, 113, D00C06, doi:10.1029/2007JD009451, 2008b.

7. 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 Spectroradiometer (MODIS) spacecraft over the Atlantic Ocean, Journal of Geophysical Research, 110, D10S12, 2005a.

8. Kaufman, Y. J., Remer, L. A., Tanre, D., Li, R. R., Kleidman, R., Mattoo, S., Levy, R., Eck, T., Holben, B. N., Ichoku, C., Martins, J., and Koren, I.: A critical examination of the residual cloud contamination and diurnal sampling effects on MODIS estimates of aerosol over ocean, IEEE Transactions on Geoscience and Remote Sensing, 43, 2886-2897, 2005b.

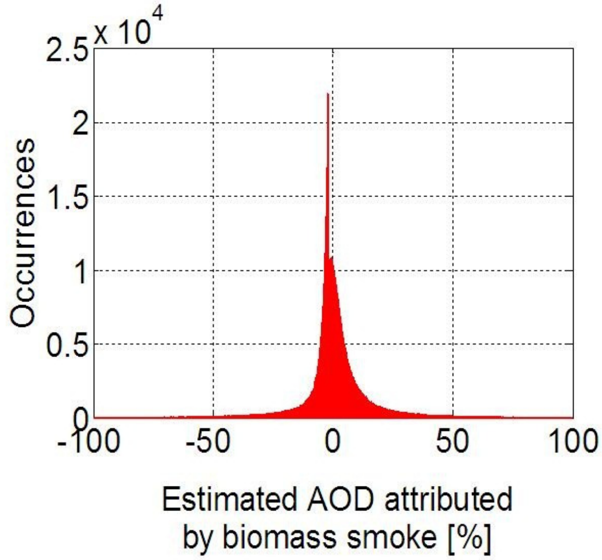
9. Knippertz, P., Tesche, M., Heinold, B., Kandler, K., Toledano, C.. and Esselborn, M.: Dust mobilization and aerosol transport from West Africa to Cape Verde: a meteorological overview of SAMUM-2, Tellus 63B, 430–447, doi: 10.1111/j.1600-0889.2011.00544.x, 2011.

10. Yu, H. B., Chin, M, Remer, L. A., Kleidman, R. G., Bellouin, N., Bian, H. S., and Diehl, T.: Variability of marine aerosol fine mode fraction and estimates of anthropogenic aerosol component over cloud-free oceans from the Moderate Resolution Imaging Spectroradiometer (MODIS), Journal of Geophysical Research (Atmos.), 114, D10206, 2009.

11. Weinzierl, B., Sauer, D., Esselborn, M., Petzold, A., Veira, A., Rose, M., Mund, S., Wirth, M., Ansmann, A., Tesche, M., Gross, S., and Freudenthaler, V.: Microphysical and optical properties of dust and tropical biomass burning aerosol layers in the Cape Verde region - an overview of the airborne in situ and lidar measurements during SAMUM-2, Tellus, 63B, 589–618, doi: 10.1111/j.1600-0889.2011.00566.x, 2011.

Figures

a.



b.

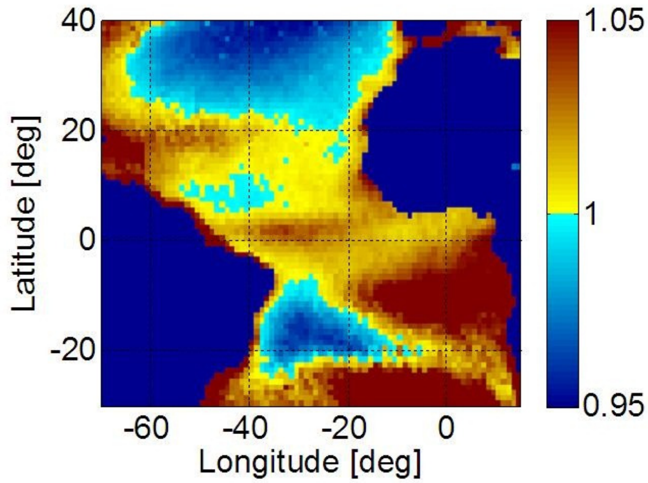


Fig. 1: (a) Frequency of occurrence and (b) spatial distribution of estimated bias in the derived τ_d (for the months Dec.-Feb) due to biomass smoke contamination. The Fig. are based on two runs of our algorithm (one with f_s=0.9, as in the paper and the second with f_s=1) and the ratio between them,

$$\frac{\tau_d(f_s = 1) - \tau_d(f_s = 0.9)}{\tau_d(f_s = 1)}$$

where f_s is fine mode fraction attributed to biomass smoke aerosol.

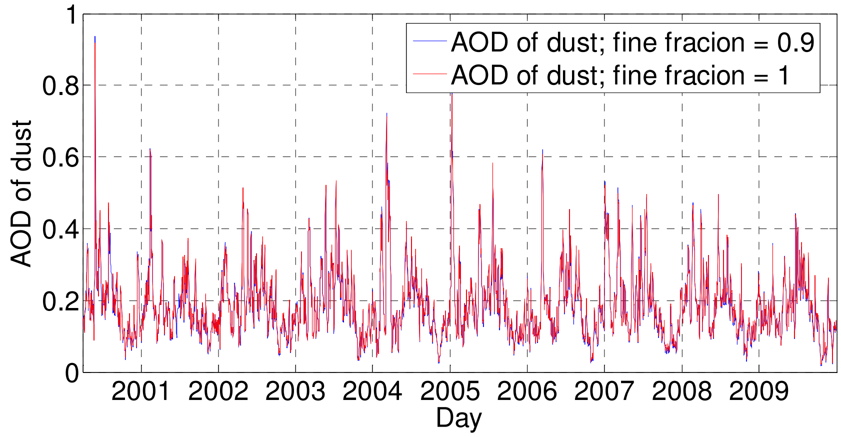


Fig. 2: Daily τ_d averaged over the study area plotted as a function of time for the years 2000-2009, as calculated by the algorithm using f_s =0.9 (as shown in the manuscript, blue curve) and using f_s =1 (red curve).

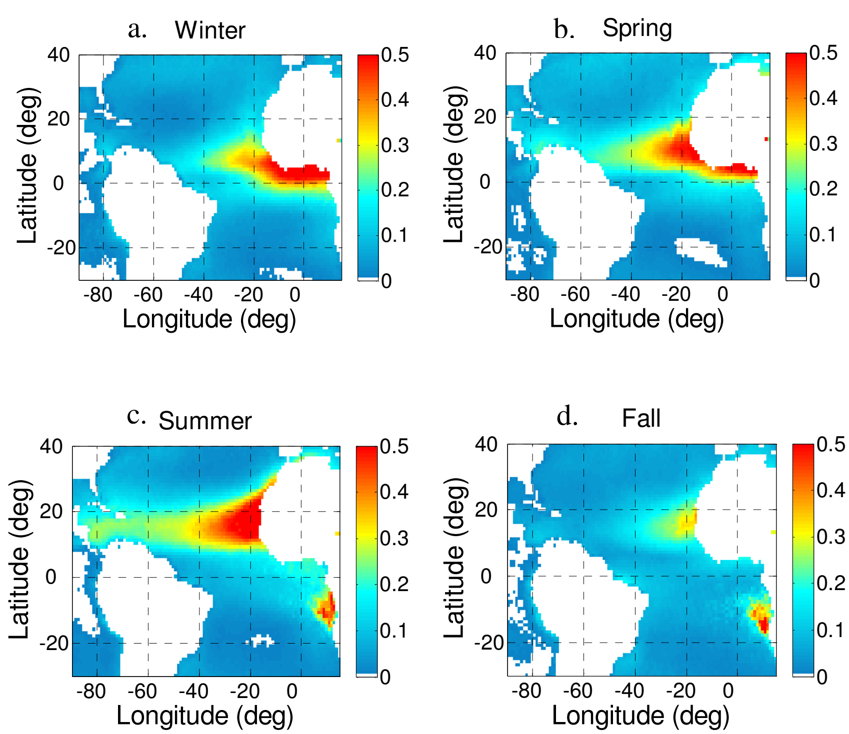


Fig. 3: Seasonal averaged values of daily τ_d for the (a) winter (Dec.-Feb.), (b) spring (Mar.-May.),(c) summer (Jun.-Aug.) and (d) fall (Sep.-Nov.), between the years 2000-2009.