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:

a. In the introduction (page 7 lines 7-14):

"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 #2

1. The Abstract is too long. Goal, methods, and essential findings should be given only. No separating paragraphs, just about 20 lines.

Answer

In response to this comment the abstract was shortened and the spacing between paragraphs was removed.

2. The introduction is lengthy and circumvents the recent and relevant literature from AMMA (DABEX, DODO) in JGR 2008 and SAMUM in Tellus 2009 and 2011. These campaigns show all the aerosol layering pattern during winter and summer mode (over central, western Africa including Cape Verde). Taking the CALIPSO observations of Saharan dust pattern over Africa and the Atlantic into account (JGR, 2008) there is almost a complete vertically resolved characterization of dust features in winter and summer including the documentation of complex mixing and layering of dust and smoke at heights above 1000 m in winter.

Answer

As requested, the revised introduction includes major revisions, especially in paragraph 5 (page 5 line 14- page 6 line 2 in the revised manuscript) and paragraph 7 (page 6 lines 12-22 in the revised manuscript). In addition, relevant literatures from recent campaigns are now cited in the following places:

a. Page 4 line 20- page 5 line 1:

"Dust transport over the Atlantic has been extensively studied, using a variety of sensors… field experiments (e.g.: Reid et al., 2003; Ansmann et al., 2011)".

b. Page 5 lines 18-20:

"It supports favorable conditions for dust emission, mainly over the northwestern part of Africa, such as enhanced surface gustiness (Engelstaedter and Washington, 2007) and cold-pool outflow (Bou Karam et al., 2008)".

c. Page 7 lines 4-5:

"The dust, partly mixed with biomass burning smoke (Formenti et al., 2008)…"

d. Page 7 lines 7-14:

"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)".

e. Page 9 lines 14-17:

"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)".

f. Page 9 lines 19-21:

"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)".

3. Page 23517: Especially here the DABEX and SAMUM literature should be cited. There are so many papers in the respective special issues describing the layering and mixing of dust and smoke.

<u>Answer</u>

Reviewers' suggestion was adopted. The new literature was inserted to the revised manuscript. Please see the previous answer for all the relevant parts in the revised manuscript.

4. Page 23519: I am not sure that such a complex mixture of dust and smoke as documented in the DABEX and SAMUM 2 papers allow such a simple separation of dust and smoke as done here. How large can the smoke-related AODs be? Please, provide some numbers! To my opinion the retrieved dust-related AODs in winter can easily be wrong by 0.1-0.3 because of an erroneous smoke AOD subtraction.

Answer

In the opening part of this document ("Preamble to specific responses") we address with many details this issue of uncertainty in the estimation of dust AOD and the estimated bias due to biomass smoke contamination during the boreal winter. We conducted a sensitivity analysis and it suggests that more than 85% of the results are bounded within an error of ±18%. A deep discussion regarding this subject was added to the manuscript (please see opening statement for the relevant added sections).

5. Page 23520: Hovmoller diagrams: Provide reference!

Answer

A reference was provided as requested.

6. Page 23521: Again, hot spots (spikes, mostly in winter) in the AOD time series can easily be produced by strong smoke events. There is no reason to speculate that all these spikes are caused by dust outbreak events. I do not trust Figure 3a, and consequently I do not agree with the (speculative) discussion on the following pages. This problem must at least be mentioned in the revised version. The variability in Figs 4 and 5 is especially high in winter (smoke impact).

Answer

As this comment is related to the most important point of the review, we addressed the problem in detailed manner in the opening part of this document. Since smoke event tend to generate less spiky signal compared to dust (smoke fronts are less profound) we believe that the biomass smoke contamination will likely to affect as a bias of all measurements.

This phenomenon of high variability in τ_d during the boreal winter was already addressed by previous studies that used other sensors to derive the dust loading (e.g.: Chiapello and Moulin, 2002). The coherency with the Bodélé pattern of emission, which is driven by the matching of the spikes, is another supporting evidence to the limited effect of biomass smoke contamination.

7. Page 23522: The authors finally state that the found 3-5 days lag suggests that the Bodele is a key source. May be! However, all emitted Saharan dust needs about 3-5 days to show up over the Atlantic Ocean (after crossing the burning areas in central, western Africa), and the highest AODs are found around the central western African coast (Figure 1, south of 10 degrees N). And all the winter AODs are highly influenced by smoke, as already mentioned several times.

Answer

We agree with the referee that 3-5 days is the time needed for most emitted dust to reach the ocean. Nevertheless, results of cross-correlation between AOD over other dust sources and our study area show almost no correlation. Two examples for such cross correlation between time series of AOD over dust sources (in northwest Africa and in Libya, based on Formenti et al., 2011 and the reference therein) and τ_d over the ocean are shown in Fig. 3a and b.

Keeping in mind that the area around the Bodélé where AOD data was collected (~140,000 km^2) occupies less than one percent of the Atlantic area in which the AOD of dust is averaged for (more than 15×10^6 km^2), as well as the perfect timing in the year, we hope that the referee appreciates this unique correlation.

8. If the correlation study would exclude the zones near Africa, and thus consider areas over the Atlantic only, e.g., 20 W, the correlation results with focus on the role of Bodele would be more convincing.

Answer

In order to convince the referee about the uniqueness of the Bodélé, we show result of cross-correlation between the Bodélé AOD loading (for the area between15º-18º N and 15º-19º E, similar to the time series we used in the manuscript) and τ_d over the Atlantic Ocean for the area westwards of 20° W (Fig 4a and b). The correlation between AOD over the Bodélé and τ_d over the oceanic area westwards of 20° W shows large increase in correlation in about 3 to 5 days delay, in agreement with the results shown for the entire study area (Fig. 7 in the revised manuscript).

Three months running correlation analysis between the Bodélé dust loading and τ_d over the oceanic area westwards of 20° W (black line in Fig. 5) shows that the maximum correlation appears during the boreal winter, in perfect agreement to the result shown in Fig. 8 in the revised manuscript.

9. As a conclusion, the impression is left: Many parts of the paper are speculative. Strong uncertainties in the AODs caused by a high uncertainty in the subtraction of smoke contributions are ignored.

Answer

The issue of mixing of the two types of aerosol and the contamination by biomass smoke is the most important point of the review, and it is addressed now in much more details in the revised manuscript. We conducted a sensitivity test to our separation algorithm and we present its results. We think that the issue of biomass smoke contamination is not ignored any more and is addressed in details in the opening part of this document and in the revised manuscript.

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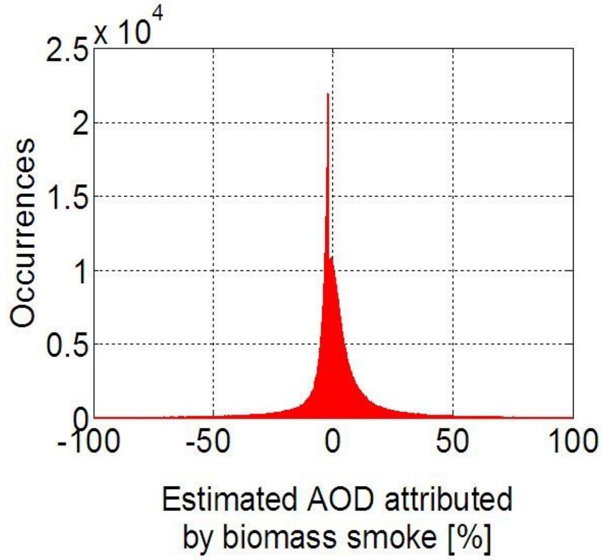
Figures

a.



b.

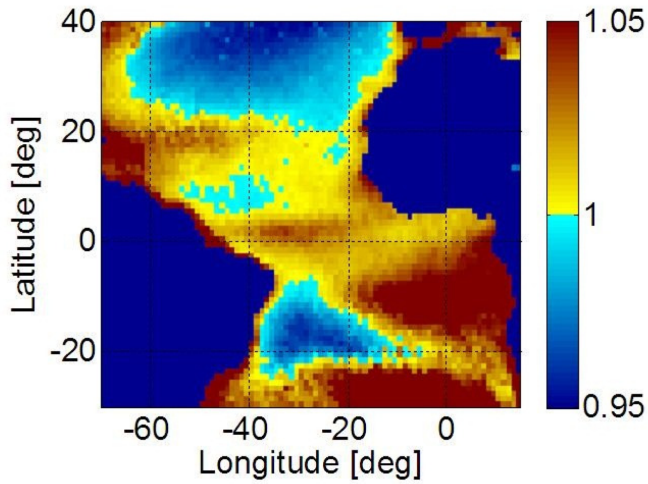


Fig. 1: (a) Frequency of occurrence and (b) spatial distribution of estimated bias in the derived τ_d (for the months Dec.-Feb) due to biomass smoke contamination. The Fig. are based on two runs of our algorithm (one with f_s=0.9, as in the paper and the second with f_s=1) and the ratio between them,

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

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

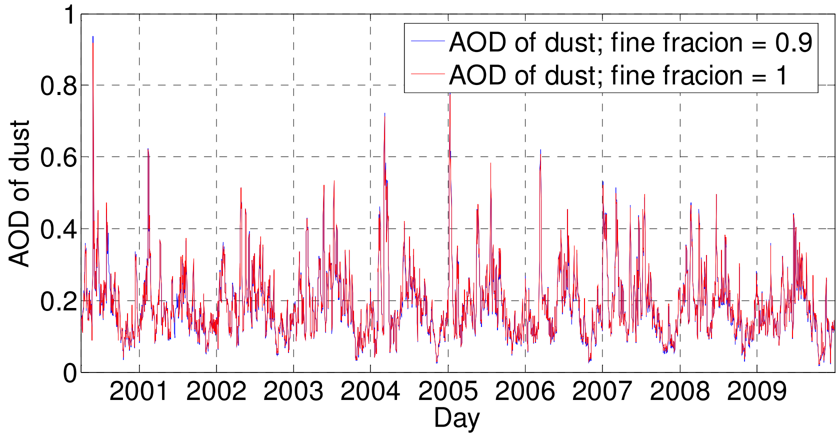


Fig. 2: Daily τ_d averaged over the study area plotted as a function of time for the years 2000-2009, as calculated by the algorithm using f_s =0.9 (as shown in the manuscript, blue curve) and using f_s =1 (red curve).

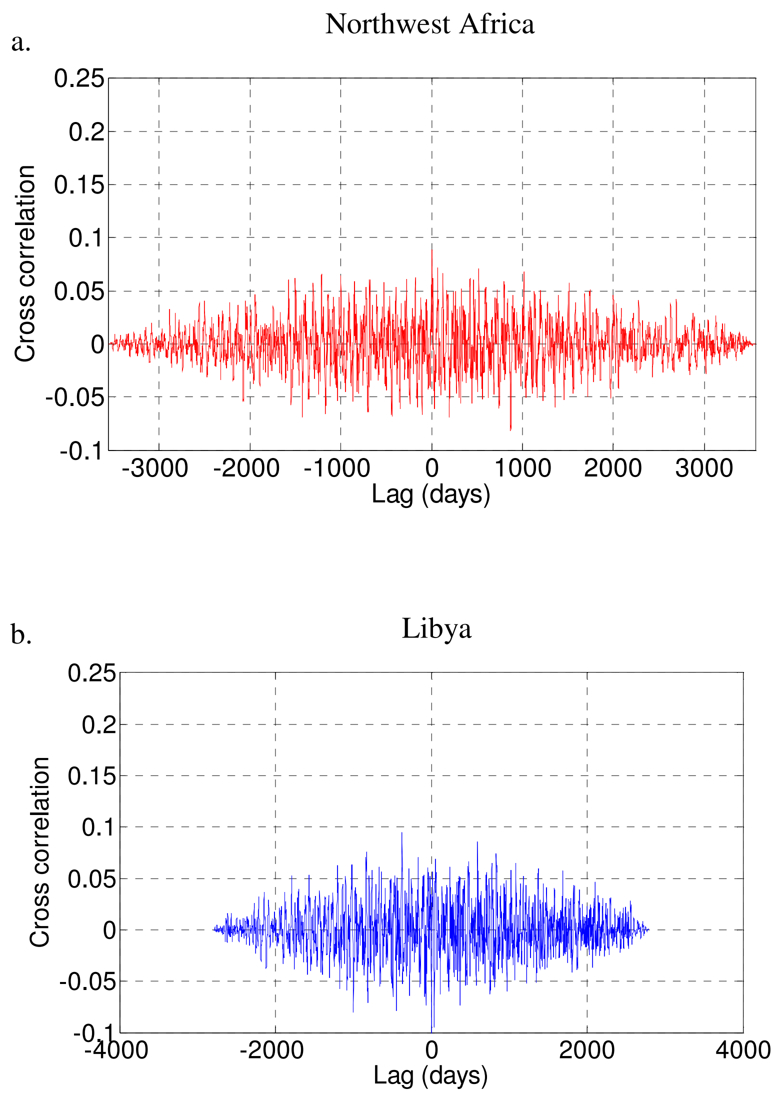
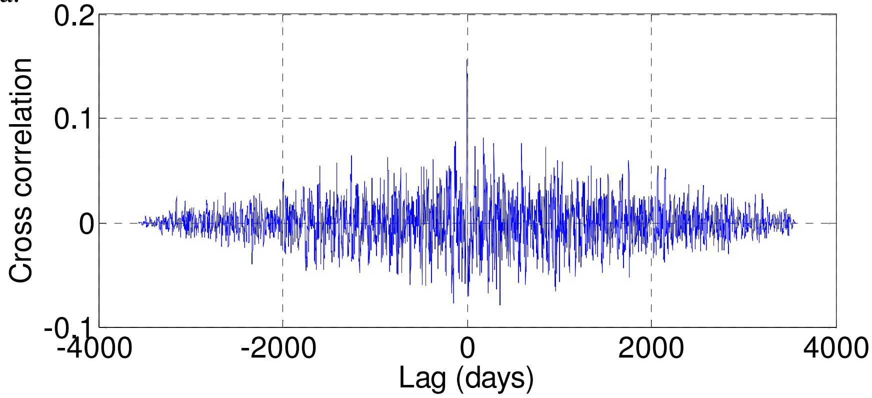


Fig. 3: a. Cross-correlation between τ_d over the Atlantic Ocean (our study area) and AOD over northwest Africa (for the area between 25º-22ºN and 3Wº-1ºE); b. the same as a but for AOD over Libya (for the area between 29º-23ºN and 17ºE-22Wº). The seasonal signal was removed from all time series.

a.



b.

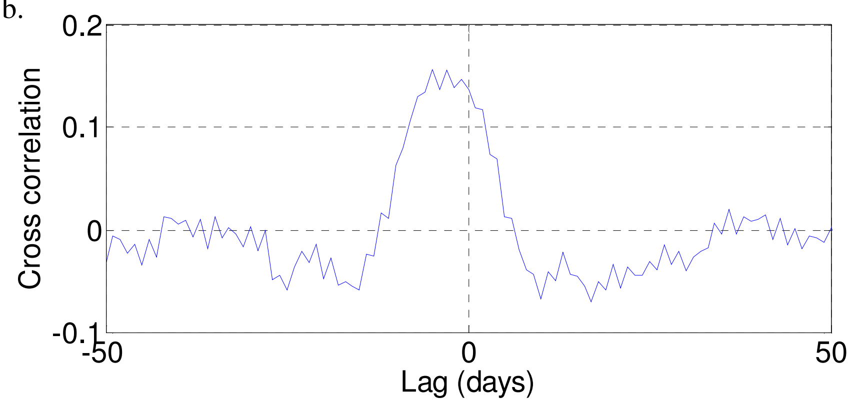


Fig. 4: a. Cross-correlation between the Bodélé AOD and τ_d over the Atlantic Ocean, <u>for the area westwards to 20°W</u>, after subtracting the seasonal signal; b. Enlargement of Fig. a for 50 days lag.

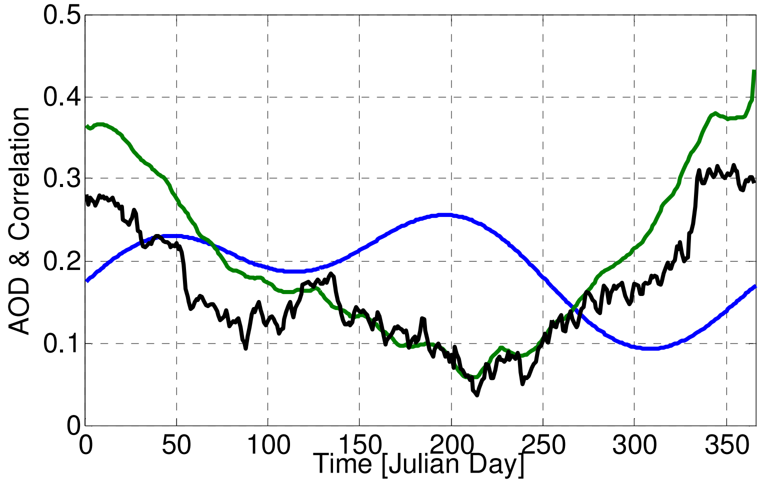


Fig. 5: Three months running correlation analysis between the Bodélé dust loading and the Atlantic AOD signal for the entire study area (green) and for the oceanic area *<u>westwards to 20°W</u>* *(black). The corresponding low pass dust loading over the Atlantic is shown in blue.*