Response to the comments of the Anonymous Referee #3

Sič et al.

September 20, 2016

We thank the reviewer for detailed and thoughtful comments that make the article clearer and more complete. Following the suggestions, we changed and added some information, and put more emphasis and explanations where it was necessary.

First of all, we would like to explain the "lack" of aerosol speciation influence by the assimilation in this study. First, the reviewer called it as a negative result, but later in Comment 3 the reviewer correctly interpreted what could be the reason of such system behaviour. The repartition of the increment that is explained in the Section 3.2 applies to all bins and species (in total) that are present in the study. Although, for example, the desert dust was the dominant aerosol type during the studied dust episode, the increment is repartitioned according to relative mass fractions of all bins present in the model (4 species with 6 bins each). Thus, the speciation of aerosol species and sizes that is present in the model field before an assimilation cycle remains the same also after the cycle and the assimilation does not change relative ratio between species and their sizes.

To change the ratio between species before and after an assimilation cycle, it would be necessary to have as a different control variable, like the 3D concentration of all 24 bins (4 species with 6 bins), and not the total concentration (all bins summed) as we have. In that case the system with its tangent linear, and adjoint code would automatically decide about the increment of each species (bin to be more precise) separately. The problem with this approach is that if we assimilate aerosol optical depth, in observations there is no information about separate species and the system would decide about repartition between different species without real foundations based on observations. To have this approach fulfil its potential it would be necessary to assimilate the speciated aerosol observations.

In the text we wrote that "AOD assimilation can also impact aerosol size and type for the same reason, but this was not evident in this experiment", thinking only about the effect of the multicycle assimilation and the model propagation of the increment. The eventual change of aerosol size and type depends on the model propagation by winds and in the same time on the differential effects of the physical processes on aerosol size and type; and it is expected to be smaller and more difficult to detect than the profile improvement which depends only on the model propagation of the increment. Its detection would necessitate more type and size dependent measurements, and the secondary aerosols in the model, and therefore it was out of the scope of this study.

We modified the text with the increment repartition to make it clear.

Concerning specific comments, below we reply point to point to the reviewer comments.

> 1. Negative results w.r.t. aerosol size and type should also be mentioned in the abstract.

As the reviewer correctly interpreted in Comment 3, it is the increment repartition in the system that fix the relative proportion between aerosols inthe model, including size and type.

> 2. P. 2, line 14: please provide at least one general reference about D.A. for atmospheric models.

We added two of pioneering references related to the assimilation of the atmospheric constituents, Fisher and Lary (QJRMS, 1995) and Elbern et al. (JGR, 1997)

> 3. Section 3.2: please clarify the repartition of the analysis increment into aerosol types. The end of the section states "...we decided to keep constant the relative mass contributions. After the analysis increment is calculated, it is repartitioned to the different bins in the model according to their background fractions of the total aerosol mass." Does this apply to aerosol types as well? This could be a key information w.r.t. the negative results which are suggested for addition in the discussion.

We agree with the reviewer, this is a key information. Since we keep constant relative amount of all bins (types included) we limited the change of the size or type by assimilation, as explained above in more details.

> 4. Equation (7): AOD is obtained by summation over all size bins and levels { but where is the summation over aerosol types? Shouldn't extinction crosssections be explicitly noted as depending on aerosol type as well?

We mean by "all bins", all different bins of all available aerosol types in the model. And, therefore, extinction cross-sections directly depend on aerosol type as well. We clarified this in the revised version of the manuscript (P6 L24 L30).

> 5. P. 8, lines 8-10: specifying correlation lengths in terms of geographic degrees leads to assimilation increments which become increasingly smaller in longitude as the assimilated observation gets closer to the poles. This is not a concern for the present study which is limited to the Mediterranean domain, but may be worth mentioning nonetheless.

The horizontal correlation lengths in the model are constant over the whole domain. The value in degrees is converted in kilometres (at equator), and then applied to the whole domain. Therefore, in the revised text, instead of presenting it in degrees we have changed it to kilometres (P8 L25).

> 6. Section 6.2: this could be the best place to document the negative result w.r.t. aerosol type, e.g. with a timeseries figure similar to figure 3, but showing the contribution of each aerosol type to AOD in the assimilation experiment. If I understand well, all aerosol types increased during the desert dust episode { contrarily to expectations.

As explained in the general comments section, the result is the direct outcome of the choice to assimilate total concentrations and repartition the increment to all bins (types included). Therefore, during the desert dust episode, by assimilation we increase/decrease all aerosols present in a grid boxes.

> 7 Figures 1 and 9: a novice reader could confuse the "forecast" results with those of the direct model run. Consider using instead the words "one-hour forecast" or even "first guess".

We agree with the reviewer, and we modified the text as suggested (Figs. 1 and 9).

> 8 P. 14, lines 8-9: "The stations in the east, like in Lampedusa and Cyprus, were not influenced by these dust events. They are mostly influenced by sea salt aerosols, and the data assimilation also here has a very positive impact." This could suggest that DA has a very positive impact on the amount of sea salt aerosols at these stations. I understand that this is not the case, so a more precise formulation is necessary, e.g.: They are mostly influenced by sea salt aerosols, and the data assimilation has a very positive impact on AOD at these stations as well.

The assimilation can increase/decrease the amount of aerosols already present in a grid box. In the case of Lampedusa and Cyprus stations, since we assimilate all aerosol types and since sea salt aerosols were dominant before the assimilation, it is mostly this type of aerosols that the model corrected.

> 9 P. 21: the discussion does not describe Fig. 10 clearly. For example, you write "...the assimilation on 26 and 27 June lowers the intensity in the higher layers..." but from pane c) it appears that assimilation lowers intensity in all layers on these days!? "On 30 June, the assimilation increases even more the relative difference between the layers. As a result, aerosol mixing ratios are larger in lower layers than in higher layers, while in the direct model run this is the opposite." I am unable to see this. At first sight this figure is contradictory with the AOD comparisons which showed elevated aerosol amounts in the analyses and observations. Specifically, the maps on 29 June (Fig. 2) showed that at the corresponding trajectory location (Fig. 10d: off the coast of Morocco) the AOD is larger in the analyses than in the direct run. Hence you should explain why the mass mixing ratio becomes smaller nonetheless. Overall, Fig. 10 is pretty but it shows only model results while the journal is AMT (not GMD). Since the result seems hard to discuss, you could as well drop it. This way the paper would finish with Fig. 9, which displays the most important outcome.

Since in Fig. 10 we can evidently see the profile change on the larger part of the plume trajectory, we decided to keep it, but we modified the text to clarify the message (P21 L28 - P22 L5). Figure 10 shows only the model runs, but it should be seen rather as the follow-up of Fig. 9. We explain the mechanisms on how assimilation can change the profiles, and one can see an obvious change of the aerosol profile and its evolution during the lifetime of the plume, from its emission to its dissipation.

We would like also to explain that Fig. 10 and Fig. 2 are in good agreement. On 29 June, in the analysis, the mass mixing ratios between the higher and lower layers are comparable, but the lower layers contain a lot more aerosol mass since the mass mixing ratio is an air density dependent unit. Thus, lower layers give majority of AOD in this case, and in the direct model run we see that mixing ratios are smaller near the surface than in the assimilation run.

We presented the figure in mass mixing ratio as this unit lowers the difference between higher and lower levels (because it is pressure dependent) and we can easily follow the evolution of the aerosols in the plume at higher levels and aerosols nearer to the surface in the same figure.

> 10 P. 23, line 2-3: "To take into account these uncertainties in the assimilation process, we defined the variances in the matrix B in such a way that it allows a margin for the model error (Talagrand, 2003)." This is not clear. Does it refer to setting the model errors twice as large as the observations (24% versus 12%) as explained p.8, lines 16? If yes, please rewrite more explicitly. If not, please expand section 3.4 accordingly

Yes, it refers to the percentages we used to define the variances. The text is corrected as suggested (P24 L3).

> 11 P. 23, lines 6-7: "One would be to add an additional term in the cost function where we would describe the errors in the model evolution." Please provide a reference on this technique. I believe that this it is named "Variational Bias Correction" at ECMWF.

We mentioned also the bias correction method at P24 L10, but what we had in mind is the method referred as the weak constrain 4D-VAR. There is really another term in the cost function J_q that accounts for the model errors with the corresponding model error covariance matrix Q besides the background term J_b and the observation term J_o. The bias correction method does not change the cost function, but the known biases in the model attributes to the observation error in the system who in such way account them as well.

> 12 P. 23, lines 12-18: One wonders why you give these details about error characterization in Benedetti et al. (2009). Do you mean that these implementation details differ from yours, yet did not prevent that "Their 4D-Var analysis results showed qualitatively a very similar impact of assimilation as in this study."? If yes, please state more clearly what are the differences between your implementation and theirs. You could also propose a few ideas about the added value of your approach compared with the implementation at ECMWF, e.g. computing cost of 3D-FGAT lower than 4D-Var ? Or shorter assimilation cycles (here 1 hour versus probably 12 hours at ECMWF) which could be necessary to improve vertical distribution as shown on Fig. 9c ?

The motivation to explicitly give details of the Benedetti et al. (2009) study is that our studies are similar as we both used total concentrations as the control variable, and the similar results show that the differences in our system do not influence qualitatively the analysis. Of course, we did not intend to go the detailed comparison, but to point out the similar performances in our system that are influenced by the choice of the control variable.

> 13 P. 23, lines 19-28: here you discuss the impact of assimilation on size distribution, but you chose to remove any such comparison from section 6. Hence this whole paragraph is not supported by any figure and actually becomes quite unclear. This is similar to the issue of aerosol type speciation which I raise in the general comments, but less important in my opinion. So you could either re-insert the size distribution results in section 6 and discuss them there, or drop this paragraph altogether.

Considering also a similar comment from the Reviewer 2, we have dropped this paragraph out of the manuscript.

> 14 P. 24, lines 25-28: you propose simultaneous assimilation of AOD by different satellite instruments, but in this case the inter-instrument biases need to be carefully considered and corrected first, before any assimilation.

We agree with this remark and we added it to the revised text (P25 L20).

> 15 P. 25, line 1: "Then with this information we could modify the size distribution and aerosol bin distribution in the model". What is the difference between size distribution and aerosol bin distribution? Couldn't this information simply allow you to modify the partitioning between aerosol types in the model?

We simplified the text, and we mention only the term 'size distribution'. This information from the multi-wavelength observations in certain cases could be used to partition between aerosol types as well.

> Minor comments...
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> Citation style is sometimes erroneous, with intext citation (LaTeX command citet) used instead of inparenthesis citation (LaTeX command citep). Examples: p. 2, lines 18-19; p. 5, line 13; p. 7, line 15.
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> P. 7, line 23: The χ^2 test does not "define" errors. Consider: "The χ^2 test is an a posteriori diagnostic which allows to check that the errors are properly specified. It checks if, for each assimilation window..."
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> P. 8, line 4: "Therefore, the possibly smaller AOD..."
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> P. 10, line 20: "The assimilated model can more readily lower the overestimated values than to elevate the underestimated values."
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> P. 18, last line: "To further explore this, we compare the modelled and the measured vertical profile follows". Please rewrite the sentence.
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> P. 20, line 5: "LOAC measurements acquired during the balloon flight (Fig. 9b) are colocated with..."

We have corrected all pointed errors.