Referee comment for

Aerosol data assimilation in the chemical-transport model MOCAGE during the TRAQA/ChArMEx campaign: Aerosol optical depth (Sič et al., A.M.T.D., 2016)

General comments

This is a well-designed and well-written paper describing a Data Assimilation (DA) system built on top of a chemical-transport model, its application to aerosol optical depth (AOD), and its performance through comparisons with assimilated and independent observations. As noted by the first reviewer already, the main achievement is the successful comparison with vertical profiles of concentrations obtained from in-situ balloon measurements. This shows, in a real-life case, how the assimilation of two-dimensional optical depths can improve the vertical distribution of aerosols. This positive result is convincingly attributed to the accumulation over time of assimilation increments peaking in layers which are correctly located by the model at emission time, and transported afterwards.

Section 7 needs revision to become clearer and more consistent with the results chosen for publication in section 6 (see specific comments 10-13). But this study has only one real weakness in my opinion: an insufficient discussion of the issue of speciation between different aerosol types - both in the description of the DA system and in the results section. One would have expected that the same mechanism improving vertical distribution would also improve aerosol speciation, especially in the present case study of a desert dust event originating in the Sahara. Yet it is simply stated in the conclusion that "*AOD assimilation can also impact aerosol size and type for the same reason, but this was not evident in this experiment*". It is important to fully document this negative result as well, especially w.r.t. aerosol type, in order to indicate the direction for future progress. The improvement of aerosol speciation thanks to DA of satellite AOD would indeed be a very desirable advance in this field.

Specific comments

1. Negative results w.r.t. aerosol size and type should also be mentioned in the abstract.
2. P. 2, line 14: please provide at least one general reference about D.A. for atmospheric models.
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.
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 cross-sections be explicitly noted as

depending on aerosol type as well?

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.

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.

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".

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.*

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.

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 1-6? If yes, please re-write more explicitly. If not, please expand section 3.4 accordingly.

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.

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 ?

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.

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.

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?

Minor comments

- Citation style is sometimes erroneous, with in-text citation (LaTeX command `\citet`) used insetad of in-parentheses citation (LaTeX command `\citep`). Exemples: p. 2, lines 18-19; p. 5, line 13; p. 7, line 15.
- 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...”
- P. 8, line 4: “*Therefore, the possibly smaller AOD...”*
- P. 10, line 20: “*The assimilated model can more readily lower the overestimated values than ~~to~~ elevate the underestimated values.”*
- P. 18, last line: “*To further explore this, we compare the modelled and the measured vertical profile follows”*. Please re-write the sentence.
- P. 20, line 5: “*LOAC measurements acquired during the* **balloon** *flight* **(Fig. 9b)** *are colocated with...*”