

Interactive comment on “Simplified aerosol modeling for variational data assimilation” by N. Huneus et al.

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We thank the reviewer for the useful comments. The main changes made to the manuscript include i) restructuring the paper according to the reviewer's comments; including in section 3 the description and the validation of SPLA and leaving for section 4 the description of the tangent linear and adjoint models; ii) expanding the scope of the paper by not limiting it to MODIS AOD but to any equivalent satellite AOD product within a given range of wavelength; and iii) illustrating the impact on linearity using a test with respect to sulphur chemistry. We believe that we have responded satisfactorily to the reviewer's comments, which has contributed to an improved manuscript. Detailed answers to the reviewers' comments follow:

1. Comments

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Abstract : The summary of the comparison between the reduced and full model glosses over some of the differences noted in the manuscript and conclusions. I think space allows for a few more details to be presented regarding the areas and species for which the reduced model is most/least consistent with the full model as well as the observations.

Changed accordingly.

641 or 642: It's probably worth mentioning the work of Collins et al. (2001) somewhere, which was one of the first studies to assimilate aerosol optical properties into a CTM.

In the introduction we give a brief summary of previous works on the estimation of emission intensities through variational assimilation. The work of Collins et al. (2001) does not concern the estimation of aerosol emissions but the improvement of aerosol mass mixing ratio. Further they do not use variational data assimilation but optimal interpolation. Therefore, including Collins et al. (2001) would change the scope of the paragraph. Other papers would then need to be added; for instance Zhang et al. (2008) on the operational assimilation of AOD over global oceans and Benedetti et al. (2009) on the operational assimilation of AOD over oceans and continents. We therefore choose not to add the work of Collins et al. (2001).

641.15: Examples of earlier gas-phase variational assimilation works other than those cited in the present manuscript are Marchuk (1974), Robertson and Langner (1998), and Pudykiewicz (1998).

The sentence “Robertson and Langner (1998) successfully applied this method to estimate the source intensity of an inert gas.” was included. However, Pudykiewicz and Marchuk were not integrated in the text for the same reasons as given above. It is not within the intentions to make a review including all methods used to estimate source intensities and all assimilation methods. We choose to concentrate our introduction on previous works on variational data assimilation for the estimation of source intensity.

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641.18: In discussing the history of variational data assimilation work for aerosol, it should be mentioned that the first adjoint of a detailed, coupled gas and aerosol simulation (GEOS-Chem) was developed by Henze et al. (2007) and applied (Henze et al., 2009) using 4D-Var to estimate emissions of aerosols precursors: NO_x, NH₃ and SO₂.

The reference to Henze et al. (2009) was included.

641.6 : “Sandu et al. (2005) developed an aerosol model for inverse modeling of aerosol dynamics that focuses only on the physical particle dynamics excluding the chemical and thermodynamic transformations.” → “Henze et al. (2004) and Sandu et al. (2005) developed inverse box models of aerosol dynamics that focus on the physical particle dynamics with limited chemical and thermodynamic transformations.”

Changed accordingly

641 or 642 : I see Viskari’s paper in the bibliography, but it is not referenced anywhere. It should probably be included somewhere around here.

Viskari’s paper was removed from the bibliography.

641 or 642 : DDM aerosol models (e.g., Koo et al., 2007; Napelenok et al., 2008) are essentially used for variational data assimilation, address many of the modeling issues (linearity, model reduction) touched upon in the present article, and are hence probably worth mentioning.

As described above we have only included works concerning the estimation of source intensity through variational data assimilation.

General: Why use a simplified model? Is it because of the computation expense of the detailed aerosol models may become prohibitive for the scope of your intended data assimilation applications, or because there are aspects of the full model not amenable to automatic differentiation with TAPENADE?

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We did not find any limitation with TAPENADE software within this study. The simplifications were introduced for a numerically efficient variational assimilation of AOD.

650.10: “Fine mode AOD computations are only conducted over ocean since MODIS retrievals for the fine mode AOD are not reliable over land.” Discussion about linearity in the fine mode simulations is limited to only over the oceans, but otherwise the rest of the manuscript discusses fine mode simulations throughout the globe. As it should. I don’t recommend limiting the applicability of a work to a single satellite product. There are other MODIS products with improved performance over land (eg., Drury et al, 2008) in addition to several other satellites (OMI, Parasol, CALIOP, someday even GLORY) for which the model presented here is of value.

Necessary changes were made in the text and figures in order not to restrict the article to a single satellite product.

651.3: I find that people often struggle to understand whether the spatial distribution of values in a plot of sensitivities ($\partial y/\partial x$) represents the spatial distribution of the numerator or the denominator. It may help your discussion to rephrase the following sentence “namely maximum sensitivity in AOD to perturbations in the emissions of desert dust particularly over Central Asia, large sensitivities to emissions of sea salt and fine mode aerosols (species 2)” to “namely maximum sensitivity in AOD over Central Asia to the perturbations.” It seems a bit premature at this point to specify which aspect of the perturbations the sensitivities reflect as they reflect the sensitivity with respect to all of the perturbations for all species emissions in all locations as well as the chemical lifetime of the gaseous precursors. While such information can be speculated, that type of analysis is more appropriate for the adjoint sensitivities discussed later. The same potential for confusion arises when discussing model sensitivities on page 653. Peaks in the TLM sensitivities are attributed to sources. Yet as shown in the adjoint modeling section, the influence of sources can be quite distant. Further over short time periods the peaks may reflect locations of secondary aerosol production from SO₂ owing to an area of heavy cloud processing rather than a location of a specific source.

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We do our sensitivity analysis based on the results of calculating the derivatives of AOD with respect to the emission with the Tangent Linear Model. These derivatives give the impact on the total and fine mode AOD for perturbations of each simulated aerosol species.

651.13: “Sensitivity tests (not shown) . . .” I find it odd that this is the only result from your sensitivity analysis that is mentioned in the abstract, yet the actual analysis is not presented. Basically, I would like to see this analysis if it’s going to be treated as a primary finding of the paper.

Analysis included.

Section 3.2.2: Some of the analysis in this section might be misleading; a factor of 3 difference between the AOD response to a 10% BB perturbation compared to a 10% FF perturbation might just mean that BB emissions were three times larger than FF emissions during selected analysis period. To avoid this issue, normalize the sensitivities with respect to the magnitude of the emissions. You could also compare to sensitivities of AOD at other wavelengths if you normalized the results, i.e. $(\partial y/\partial x)(x/y)$.

We study the sensitivities of AOD to the emissions through the different terms in the Jacobian matrix (eq. 8). Each term in the matrix corresponds to the partial derivative of the AOD with respect to the emissions, i.e. the increment in AOD due to each new aerosol emitted. The results presented are therefore independent from the relative emissions intensity between species. Changes were made in the text to avoid confusions.

654.24: “Differences in magnitude. . .” This isn’t the clearest sentence, so perhaps I misunderstood, but do you mean to say that the sensitivity with respect to the emissions increases as one considers emission on days that are increasingly prior to the observation? You are showing only the sensitivity with respect to emission on specific days, not the total emissions sensitivities integrated from the observation point back to those days, correct? If so, then wouldn’t the influence of emission on a specific day

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eventually decrease as one considers days further in the back in time than the average lifetime of the aerosols? In other words, if you look at the sensitivities 20 days back, it is probably smaller than the sensitivity 5 days back, which would contradict the sentence in question.

The figures present the necessary emissions to create two (Fig 10a) and five (Fig 10b) days after a given perturbation of fine mode AOD in a particular location. It is premature to make this type of analysis at this stage since it is not only a matter of emission flux but also a matter of circulation. We therefore eliminate the sentence from the text.

660.4: “As a consequence, several modifications had to be introduced”. This strikes me as a perfect opportunity to test the 4D-Var capabilities of your model in a novel way. Why not generate some pseudo observations with the full model and then assimilate these with the simplified model, letting the system adjust parameters of the simplified model in an optimal fashion such that the simplified model best matches the estimate of the full model?

We appreciate the comment. We will keep this suggestion for the article presenting the full assimilation system and the results from applying this system to estimate the aerosol and aerosol precursor emissions. At this point we prefer to keep this article strictly to presenting the model with its tangent linear and adjoint model versions. The inclusion of an assimilation experiment would oblige us to present the assimilation system and a series of concepts that would both extend considerably the length of the article and change its scope.

General: comparison of full and simplified models: One frequently mentioned drawback to most 4D-Var studies, compared to filtering approaches, is the assumption that the forward model is perfect. Here, as usual, the simplified forward model is not perfect. However, the authors are in the special position of having just quantified the areas in which the model is imperfect, at least with respect to the LMDz model. How will the information gleaned of the SPLA and LMDz models be used to guide a real

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data assimilation study or to interpret the results of such study? Could the differences noted here be used to construct model error covariance matrices to be used in a weak-constraint 4D-Var framework (e.g., Trémolet, 2006)?

We thank the reviewer for this comment. We will use SPLA and its adjoint in a first step within a strong-constraint 4D-VAR framework, i.e. assuming that the model is perfect. We agree with the reviewer that the combination of simplified model and original model gives a unique opportunity to study the impact of model-error on assimilation and will be certainly explored in the future. However, we would like to point out that the simplification error, i.e. the differences between LMDz and SPLA, give only a small part of the model-error characteristics, the remaining and most important ones, come from the discrepancies against the observations (see Fig 6 and 7).

2. Minor comments and clarifications

Abstract : Suggest the first sentence be rearranged to read "... simplified aerosol optical depth model together with its tangent linear and adjoint versions for the ultimate aim of optimizing global aerosol and aerosol precursor emission using variational data assimilation".

Sentence was change to "... simplified aerosol model together with its tangent linear and adjoint versions for the ultimate aim of optimizing global aerosol and aerosol precursor emission using variational data assimilation"

641.4: "small" → "smaller"

Changed accordingly

641.11: "optimal initial state" → "optimal state"

Changed accordingly

641.18: "aerosol emission" → "aerosol and/or aerosol precursor emissions"

Changed accordingly

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644.23: "The conversión from terpenes to OC varies from 0.1 to 15%". Actually, the SOA mass yields for some terpenes can be greater than 100% (eg Ng et al., 2007), so it might be prudent to revise this statement.

Sentence was changed to "The conversion from terpenes to OC can vary largely depending on factors such as the initial concentration of terpenes,..."

647.6: Do you really mean POM (an acronym yet defined but likely meaning primary organic matter)? If so, what happened to the secondary organic aerosol from terpene oxidation? Please check usage of POM on page 656 as well.

POM was changed to OM in both pages. The emissions of OM already include both primary and secondary organic aerosols.

650.16: "This can be obtained. . ." Can this sentence be expanded and clarified?

The sentence was changed to "The minimization of these non-quadratic functions can be obtained with an increase of the computational load (eg. higher number of iterations) compared to the linear case"

650.21: "compare it with the difference. . ." This is a bit vague. I think it would be clearer to write out in equations the comparison that is being made here: $H(x)-H(x+dx)=H'(x)dx + \text{nonlinear terms}$. Also, any error in construction of H might accidentally get lumped into the nonlinear terms. So it would be nice to mention, as you do for the adjoint model, that H has been numerically validated.

The equation terms have been included in the text and the tangent linear model version was numerically validated with the Taylor formula.

Throughout: should differentiate between scalars and vectors using bold

Corrections were made and the sentence "We follow here and elsewhere the notation of Ide et al. (1997)" was added.

651.17: "July 2002 are shown." -> "July, 2002."

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Changed accordingly.

651.21: I don't think the BB and FF acronyms have been defined

Definitions have been added to the text.

654.4: "130 times" excellent! Any comment on the computation expense relative to the forward model, or the size of the checkpoint files relative to the model grid size and time step?

The computational cost of the adjoint model is between 5 to 6 times that of the forward model. The model grid size and time step are the ones from LMDz and the size of the checkpoint files is approximately 2 Go.

642.20: similar to the flow presented in the conclusions, I think it would be better to place the contents of section 4 directly after the introduction of the reduced model (3.1) and before the sections on sensitivity analysis. So make section 3 all about the definition and validation of the reduced model, and then make what are currently sections 3.2 and 3.4 to section 4.

Changed accordingly

Conclusions: "Variational data assimilation technique have been developed for individual aerosol species that determine the emissions field that represents the best compromise between a given set of observations and the a priori information." While this is the ultimate goal, only some of the tools necessary for performing variational data assimilation have been presented thus far. Other components, such as the optimization algorithm and construction of the a priori and observation error covariance matrices, have yet to be addressed.

The sentence referred to other studies. To avoid confusion we changed the sentence to: "Previous works have used variational data assimilation techniques to estimate the emission field for single aerosol species that represents the best compromise between a given set of observations and the a priori information."

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