

## ***Interactive comment on “Estimating aerosol emissions by assimilating observed aerosol optical depth in a global aerosol model” by N. Huneus et al.***

**N. Huneus et al.**

nicolas.huneus@lsce.ipsl.fr

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Response to Reviewer #2

We greatly appreciate the reviewer comments that helped to improve the quality of the paper. We have addressed each one of the comments below.

General comments

I agree with the comments made by the first reviewer, hence, I will try not to repeat things already mentioned here.

R: The introduction includes a fairly thorough account of previous work in the area

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of aerosol emissions estimation using data assimilation, however, the purpose of this study is only mentioned briefly in the final paragraph. I would suggest that the authors expand the discussion of what their study contributes, for instance, is it that there have been no previous top-down estimates of SS, POM and SO<sub>2</sub> (previous studies on BC and DD are mentioned)? Also, that this is the first multiple aerosol species inversion.

A: This information is given in the last two paragraphs of the introduction. However we have reformulated the last paragraph to highlight the contribution of this study.

R: The writing is sometimes unclear and could be made more succinct.

A: In places pointed out by the reviewer the text has been corrected to make it clearer to the reader to understand.

Specific comments

R: p3079, l15: for clarity “the analysis vector” or “state vector” (and hereafter)

A: Although the reviewer is correct and the analysis should strictly speaking be referred to as “analysis vector” in our case, in the literature it is often simply called “analysis”. We therefore prefer to keep this terminology and omit the term “vector” when talking about the analysis.

R: p3079, l16-17: this sentence is confusing and should be rewritten

A: The sentence has been changed trying to make it clearer to the reader.

R: p3079, l23: “e.g.” Rodgers et al 2000 (there are many texts about this)

A: Changed as suggested.

R: p3080, l18: for clarity “simulated (Hxb) and observed (y) values”

A: Changed as suggested.

R: p3080, l23-24: the variational approach becomes advantageous only when the H cannot be defined (either it is too large or it’s terms are not known explicitly).

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A: Indeed, and we state the case when the size is too large. We have kept the explanation limited to our case in order to avoid going into details that would not contribute to the understanding of the article and complicate the reader.

R: p3083, l2: should outline what these considerations were and add some statement about how these emission inventories i.e. from 1 decade ago are different from recent ones

A: Recent emission inventories such as the one from Lamarque et al. (2010) compared to old ones (as the one used) include more up-to-date information in their estimation, e.g. reduction in sulfur emission over Europe (Figure 10) possibly due to influence of mitigation policies. One of the reasons to use the old emission inventory was to see whether the estimations were approaching the more recent inventories. We do not consider it necessary to highlight this in the text given that it does not contribute much to final result of the paper. The corresponding paragraph in section 4 has been modified to include a statement on the differences between the old and more recent inventories.

R: p3083, l5-11: this should go in the introduction (see general comments).

A: The information in the above-suggested lines is already included in paragraph 5 of the introduction, it is just formulated differently. The last paragraph starts with a sentence stating that this is the first study to simultaneously estimate different aerosol species. We have changed this first sentence to make the statement stronger. The sentence reads now “This is the first study to estimate simultaneously the global emissions for multiple aerosol species and one gaseous precursor (namely DD, SS, BC, POM and SO<sub>2</sub>).”

R: p3085, l16: the authors say the data were “thinned” do they in fact mean that they were averaged to the lower model resolution. Please clarify.

A: The thinning is motivated by the need to remove correlated observation errors as

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much as possible. We have considered different ways to do it, based on random sampling or averaging. For this study, we sample one satellite pixel each day for each model grid box, among the ones contained within this grid-box.

R: p3086, I3-4: perhaps the covariance between errors in e.g. OC and BC emissions are difficult to determine, however, one could imagine that such a covariance could be large since they are both emitted by e.g. biomass burning. What is the potential impact of ignoring possibly large covariances such as these?

A: Indeed, as mentioned in lines 2 and 3 of page 3086, emission errors between two species in one region might be correlated. Ignoring these covariances, on the one hand allows to assume a diagonal B matrix, which simplifies the computation of the solution. On the other hand positive correlations in the B matrix reduce the number of degrees of freedom of the state vector. If we could identify and quantify some of them, the observational constraint would then increase.

R: p3087, I16-17: why are there two error numbers listed directly one after the other?

A: The MODIS errors on AOD are defined as a linear function of the AOD and the two errors correspond to the two constants defining this linear function. The first error corresponds to the y-intercept while the second corresponds to the slope.

R: p3090, I15: approximately how many AERONET data points went into the monthly mean at each site?

A: The data points going into the monthly mean vary not only from station to station but also from month to month. As an example, in Capo Verde the monthly mean for May was computed using 27 days whereas the one for March uses only 16 days. The monthly mean of May at Banizoumbou was computed with only 7 days. In spite of these large differences we highlight that the model monthly mean is computed considering the same days used in the AERONET monthly mean.

R: p3099, I13: “assess the impact of the assimilation on the errors” here it is not clear

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which errors are meant. Diagonal elements of A are the posterior uncertainties of the state variables (as is mentioned l12) so what is the error being referred? It appears as the terms “error” and “uncertainty” are being used here interchangeably, which makes this whole paragraph confusing.

A: The sentence has been extended to “to assess the impact of the assimilation on the errors of the estimated emission fluxes” and the term “uncertainty” has been replaced with “error”.

R: p3100, l13-17: these three sentences repeat information and should be made clearer and more succinct

A: The sentences have been reformulated and read now “In spite of the additional constraint from the assimilated fine mode AOD over ocean, the total AOD presents a larger improvement (with respect to MODIS) after assimilation in terms of RMS error and R over land than over ocean. This is due to the larger departures of the simulated AOD to the observed one over land than over ocean.”

R: p3096, l22: Could the authors offer an explanation as to why the MODIS and AERONET data so different at Mauna Loa and for other stations with large differences in general?

A: No in-depth analysis has been conducted to determine the cause of the difference between AERONET and MODIS. However, as stated in lines 2-6 of page 3101, this mismatch could be the result of the thinning applied to the MODIS data in order to convert them to the coarser model resolution.

R: Fig 8: MODIS data are missing at Solar Village.

A: This is correct. Solar Village is located in the Saudi Arabian desert and MODIS does not retrieve the AOD over bright surfaces such as deserts. This can be seen in Figures 3 and 5, white surfaces (corresponding to no retrieval) can be seen over northern Africa and the Middle East.

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## Technical comments

R: p3080, l20: “the sensitivities of the observation operator (H) and the relative weights of the R and B matrix” (remove “to”).

A: Changed as suggested.

R: p3080, l22: need to use consistent terminology either “state” or “analysis” vector.

A: The state vector is the vector composed by the elements one intends to estimate and the analysis corresponds to the state vector at the solution. The analysis contains therefore the final estimates that represent the best compromise with respect to the observations and the a priori information. We have used the term analysis only when referring to the estimated fluxes whereas state vector was used elsewhere. The use of state vector was limited to sections describing the assimilation method (sect. 2.1), the state vector itself (sect. 2.3) and the experimental setup (2.6). We believe that we have been consistent in the use of this terminology and have therefore kept it as it was.

R: p3081, l28: “caused by”.

A: Changed as suggested.

R: p3084, l10: “provided” not “delivered”

A: Changed as suggested.

R: p3084, l11: “chose” not “choose”

A: Changed as suggested.

R: p3084, l12: “onboard the Terra satellite”

A: Changed as suggested.

R: p3085, l15: “south of 40\_S” not “over”

A: Changed as suggested.

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R: p3093, l20: “associated with”

A: Changed as suggested.

R: p3102, l21: “corresponding to” or “correspondent with”

A: Changed as suggested.

R: p3102, l21: “than at present”

A: Changed as suggested.

R: p3103, l6: “large uncertainties in the aerosol impact on climate”

A: Changed as suggested.

R: p3103, l23: “one year’s worth”

A: Changed as suggested.

R: Fig 8 - 10: figures are difficult to read – the axis labels, titles and legends are too small. The legends could perhaps be removed as it is the same in every sub-plot and simply given in the captions.

A: The size of the axis labels and titles was increased and the legend removed and given in the captions.

R: Please also note the supplement to this comment: <http://www.atmos-chem-phys-discuss.net/12/C344/2012/acpd-12-C344-2012-supplement.pdf>

A: No additional comment could be found in the supplement document.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 3075, 2012.

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