Atmos. Chem. Phys. Discuss., 13, C2507–C2513, 2013
www.atmos-chem-phys-discuss.net/13/C2507/2013/

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Atmospheric inversion of SO_2 and primary aerosol emissions for the year 2010" *by* N. Huneeus et al.

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Received and published: 19 May 2013

We greatly appreciate the reviewer comments that helped to improve the quality of the paper. We have addressed each one of the comments below. In addition, an error in the computation of the annual uncertainties was corrected. However, this correction has no consequences on the general conclusions of our work, which is based on uncertainties at the monthly scale. Only the magnitude of the global annual mean uncertainty is affected.

Major comments:

Referee: My only concern with the overall approach is that there is no mention of the potential role that secondary-organic aerosols could play in the overall distribution of

aerosols, and how that would affect the results of the inversion. It is clear that modeling SOA is still very much an open question, but it is nevertheless worth asking the question how much of the biases that lead to changes in emissions could be attributed to missing aerosols.

Answer: It should be noted that the model does include a crude representation of secondary organic aerosols (SOA) of biogenic origin. Our simplified aerosol model (SPLA) follows the approach of the original model (Reddy et al., 2005) in that it considers that 11% of the terpene emissions are converted to SOA. A more detailed description of the different emissions sources included in the aerosol model can be found in Huneeus et al. (2009). Although included in the model, this source of aerosols is not considered active and therefore is not optimised. However, we agree that not including the SOA in the state vector could lead to biases in the estimate of other aerosol species. Moreover our scheme does not include SOA from anthropogenic emissions of volatile organic compounds. This missing aerosol type may affect the inverted fluxes of anthropogenic SO2, BC and POM. A more complex aerosol model would be needed to test the biases this may introduce. We have introduced the following text in the conclusions to address this issue: "The general validity of the resulting emission fluxes depends also on the aerosol model used in the inversion. The representation on processes such as the formation of secondary aerosols and the aging of particles could lead to biases in the estimated fluxes. To explore the impact of these representations on the resulting emissions, they need to be used in models with higher complexity. The posterior validation of the simulated AOD could reveal weaknesses in the simplification of the aerosol model that will require improvement in the future."

R: My other comment relates to the sole use of optical depth as verification. It seems that this could be considerably improved by using comparisons against surface measurements of aerosol concentrations.

A: In this work we force the model to fit the observations by adjusting the total aerosol content within the atmospheric column through the increase or decrease of emissions.

The use of surface concentration for verification would not just evaluate the quality of the inversion but also the model's performance to reproduce the aerosol vertical distribution and the boundary layer height. Therefore, an improvement or deterioration to reproduce the surface observations with a new emission estimate would not necessarily allow us to conclude on the performance of the inversion.

Minor comments:

R: Page 6172, line 10: duplicate (2)

A: The word document submitted to ACPD had only one equation number. Special attention will be given at the proof reading step to avoid this kind of repetition.

R: Page 6173, line 24: 80% still seem like a rather large threshold for potential cloud interference. How was that number selected?

A: The threshold is taken from the study of Zhang and Reid (2006). The authors in that study made a thorough analysis of the MODIS aerosol optical depth (AOD) over ocean and developed a quality assurance procedure and empirical corrections to further decrease the biases and error variances of the AOD. The aim was to prepare the data for near-real time data assimilation. The reference in the text was corrected since Zhang et al. (2008) is cited.

R: Page 6173, line 27: the statement "thinned" is somewhat puzzling. Should it be "averaged"? What is the procedure to identify the importance of and take into account the variability within the 3.75x2.5 grid box?

A: "Thinning" is the term usually employed to refer to the reduction of data density by sampling. In the case of our work, the satellite data, with a resolution of 1°x1° are brought to the model resolution of 3.25° x 2.5°. This is motivated by the need to remove correlated observational errors as 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 those contained

within this grid-box.

R: Page 6174, line 16: while the physical process is the same everywhere, the biases in wind speed are probably not.

A: This is true and is one of the reasons to increase the number of SS regions in the future. However, we note that the impact of this choice of regions on the emission estimate of SO2, BC and POM is limited. This is because on one hand the main constrain on these emissions is coming from AODs over the continents (where AOD is also larger than over ocean) and on the other hand SS has little or no impact on the AOD over continent.

R: Page 6176, line 5: there has been a significant number of small volcanoes that lead to the observed increase in stratospheric aerosols. Any indication of how that would affect the results (for 2011)?

A: Background volcanic emissions of SO2 in the troposphere are included in the model but not in the state vector and are therefore not optimised. However the reviewer is right that there has been a series of small volcanic eruptions in the stratosphere that has increased slightly the optical depth of stratospheric aerosols, which is not represented in the model. However we concentrate here on the year 2010 for which the stratospheric AOD remains fairly small ($\sim$0.003, Vernier et al., 2011) and unlikely to affect significantly our inversion and the conclusions which are drawn. The following text was added to Section 2.3: "The contribution of a number of small volcanoes to stratospheric AOD (not represented in the model) remains fairly small ($\sim$0.003) for the period of interest in this study and is therefore unlikely to affect significantly our inversion and the conclusions which are drawn (Vernier et al., 2011)."

R: Page 6177, lines 23-24: It is hard to see the connection between the CO2 inversion (and the adjustments on error) and this research. Please expand.

A: Both applications, the inversion of CO2 and aerosol fluxes, share the same theoretical framework. Each of them estimates the emissions through the minimization of a cost function as presented in Eq. 1. As for the case of CO2, we assume the error covariance matrix of the observations to be diagonal. Yet correlated errors exist between adjacent pixels that are not represented in a diagonal error covariance matrix. To our knowledge no published study on the inversion of aerosol source has presented a method on how to deal with these error correlations. We have therefore benefited from the experience gained in the inversion of CO2 fluxes and applied their method to represent error correlations in the R matrix. We chose not to change the text in this aspect since we consider it would not improve the general understanding of the results.

R: Page 6178, lines 1-3: isn't it obvious that increasing the model error leads to improvement against observations?

A: The estimated fluxes correspond to the best compromise between the a priori information and the observations based on the weights assigned to each one of these pieces of information. This is expressed in Eq. 1 and the weights correspond to the error statistics of the a priori information and of the observations as defined in the matrices B and R, respectively. Therefore if the errors of the observations are increased (while keeping the a priori ones constant) their relative weight is reduced and the solution will be closer to the a priori. The model error was introduced in the R matrix thus impacting the weight of the observations. An increase of the model error as defined in our approach decreases the weight of the observations with respect to the a priori and therefore does not necessarily lead to an improvement against the observations.

R: Page 6181, line 4: add reference to ACCMIP

A: The reference to Granier et al. (2011) was added.

R: Page 6182, line 25: it is probably important to comment on the degree of independence between MODIS and AeroNET? Was AeroNET used by MODIS for ground-truthing?

A: The reviewer is right: there is a certain degree of dependence of MODIS data to AERONET. The latter has been used as "ground truth" in the validation of MODIS products and the aerosol models used in the MODIS algorithm combine information from sun/sky photometers (not only AERONET) and from analysis errors in the products from previous versions of the MODIS algorithm. In spite of this dependence, these datasets can be largely considered as independent (and they are considered as such in the literature) in view of the differences not only in their algorithms, but also their viewing geometry, representativity (AERONET being a point measurement whereas MODIS represents a satellite pixel), sampling, etc. Furthermore, in the mentioned lines we try to explain the bias degradation after assimilation of the fine mode AOD with respect to MODIS and AERONET even though the rms error and the correlation coefficients are improved with respect to both datasets. The analysis with respect to MODIS fine mode AOD is limited over the ocean since no data are assimilated over land whereas the analysis with respect to AERONET corresponds mostly to continental stations. Therefore the interpretation of this bias degradation is different for both datasets and is not related to their possible dependence but rather to the reasons indicated in the manuscript.

R: Page 6185, lines 25-27: it could also be that the observations over those areas are so biased that they bring no information to the assimilation.

A: We consider the assimilated observations to be the truth and do not apply any bias correction to the observed AOD. In that sense the observations always bring some information to the assimilation system, but that information may be incorrect and contradict the first guess. However we do not have any objective way to know if the retrieved AOD is biased over a particular region and assuming the MODIS AOD is not biased is a legitimate assumption at this stage. More sophisticated methods exist to perform bias corrections for the assimilated data but are beyond the scope of this study. The sentence was added to the lines following the ones in questions, "We assume at this stage that the MODIS AOD are not biased. Sophisticated methods exist to perform bias corrections for the assimilated data but are beyond the scope of this study."

R: Page 6187, line 22: change "coincide" to "agree"

A: Modified as suggested.

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 6165, 2013.