Reply to Reviewer #1:

The manuscript "The SPRINTARS version 3.80/4D-Var data assimilation system: development and inversion experiments based on the observing system simulation experiment Framework" presents an assimilation system based on 4D-Var assimilation method and the SPRINTARS global aerosol model. The system assimilates aerosol optical depth and seeks to find the emissions and initial concentration that represent the best compromise between the background information and the observations. To reduce the computational costs the authors have divided the system in outer and inner loop with the on-line version of SPRINTARS in the outer-loop and an off-line model version in the inner loop. The off-line model version was examined in its performance to reproduce the on-line equivalent observations. Finally, the system was applied in a series of experiments to study the feasibility and capability for aerosol inverse modelling. This is a very interesting paper, well written and the methodology as well as the results are clearly presented. However, I consider that important aspects should be addressed before its publication.

We thank a referee for careful reading our manuscript and for giving useful comments. We have deliberately confirmed and considered your comments. We believe that we have made adequate answers to your comments. In revised manuscript, the changes are highlighted by yellow makers.

General comments:

1) The cost function in equation 3 includes one term corresponding to the initial concentration and one to the emissions. Therefore in the inversion process the system will use both, initial concentration and emissions, to find the minimum of the cost function. However, after introducing the cost function and the state vector in section 2, no further mention about the initial concentration is given in the manuscript. The authors should explain why the initial concentration and its role in the inversion have been omitted after section 2.

In order to simplify the equations, as defined in the equation at Line 14 on Page 3431, we used the unified vector ***x***, which consists of initial concentrations and emissions, in equations 6, 7, 8, and 11. As mentioned in Section 2, the SPRINTARS/4D-Var data assimilation (DA) system is capable of assimilations for both initial conditions and emissions. In OSSEs in section 5, only aerosol emissions were included in the control parameters (the state vector) to evaluate the performance of the DA system for inverse modeling. As explained at Lines 23-24 on Page 3442, the NR, CR, and AR used identical aerosol fields as initial conditions. We added more detailed description about the experiment setting of inverse modeling in section 5. Also see the reply to the general comment #2.

2) Although the authors present the main parts of the inversion system, additional information not provided to the reader should be included. What is the assimilation window, is it the full 10 days period? The adjoint of SPRINTARS has been derived, but what is its accuracy? The state vector should be better presented. Is just the total dust emission estimated or each one of the their bins separately? How is the initial concentration defined in the state vector? What about the sea salt? Since a scaling factor is applied, I assume the system does not create sources, just increases/ decreases the existing ones, this should be put explicitly.

The assimilation window is 10 days based on average lifetime of aerosols in atmosphere (about a week). We revised a state at Line 25 on Page 3442 as follows:

"The experimental period was 10 days (21–31 May 2007) based on average lifetime of aerosols in atmosphere. The assimilation window was the full 10 days period."

As mentioned at Line 11 on Page 3436, the adjoint version of SPRINTARS was derived directly from the discrete model source codes of OFS. By using the following equation R1, we had confirmed that the degrees of precisions of adjoints of each individual process (e.g., advection, reaction, depositions and emission) for each aerosol are on the order of the round-off error.

$$\left(\mathbf{Mx}\right)^T \mathbf{Mx} = \mathbf{x}^T \left(\mathbf{M}^T \mathbf{Mx}\right) \qquad \text{(R1)}$$

We inversely estimated total dust and sea salt emissions. As mentioned in the reply to the general comment #1, in the OSSEs, optimization of the initial concentrations were not included. The scaling factor allowed increases or decreases of the existing emissions, and cannot detect missing sources.

We added a new paragraph in section 5 to describe the setting of the inverse modeling more carefully.

"We assigned scaling factors of aerosol emissions to control parameters. The scaling factor allowed increases or decreases of the existing emissions, and could not detect missing sources. Because the CR was generated by emissions perturbed by the scaling factor (as shown by Equation 15), detection of missing aerosol sources is beyond the scope of this experiment. Emissions of dust and sea salt aerosols, which have several particle bins, were adjusted as total emissions (not each emission of their bins). The initial aerosol conditions were not included in the control parameters, and the CR, NR and ARs were initialized with identical aerosol fields. The background errors were based on the setting of the CR. We assigned 200% for SO2 and carbon emissions. For the natural aerosol emissions (dust and sea salt), 300% of uncertainty was assigned. Temporal and special correlations were not considered in this study. The experimental period was 10 days (21–31 May 2007) based on average lifetime of aerosols in atmosphere. The assimilation window was the full 10 days period."

3) The definition of the error covariance matrix in equations 4 to 6 is crucial in the minimization of the cost function since it will determine the weight of the different pieces of information in the minimization. The authors present briefly the observation error used but do not present neither of the background error covariance

matrices. The authors should not only present how they define these matrices but also explain their choice.

The background errors were defined based on the setting of the CR. The variations of the perturbing scaling factors, which deviated the CR from the NR, were assigned to the background errors. The background errors were 200 and 300% for SO2 and carbon emissions, and dust and sea salt emissions, respectively. We added a more detailed description of the setting including a definition of observation and background errors in section 5. Also see the replies to the general comment #2 and the comment # 3 by reviewer #2.

4) As part of the Monitoring Atmospheric Composition and Climate (MACC) project, the assimilation of aeorosl optical depth from MODIS was included in the Integrated Fore- cast System from the ECWMF. This work and the results have been presented in Benedetti et al. (2009). Zhang et al. (2008) also assimilate aerosol optical depth in an operational forecast system. The authors should at least include these works in the introduction. Furthermore, additional studies estimating aerosol emissions from inverse methods are Zhang et al. (2005), Huneeus et al. (2012, 2013) and Fu et al. (2012).

Thank you for your suggestion. We added these references in introduction.

Specific comments:

1) The abstract should include which kind of observation is assimilated and what the state vector is?

We can find the description about the observation at Line 22 on Page 3428. We added an explanation about the state vector (the control parameter) in the abstract.

"… and coverage (land and ocean), and assigned aerosol emissions to control parameters. "

2) Section 2, provide a reference with the theoretical background of variational assimilation

We added (Talagrand and Courtier, 1987).

3) Page 3438, line 20, reformulate "does the dust emission begin to emit". Replace wit something like "dust is emitted".

We replaced that with "dust is emitted".

4) Page 3438, line 23, STD is used but not defined. Define acronym of STD.

It should be ONS, not STD. There were some mistakes (e.g., Tables 1-3). We corrected them.

5) Page 3434, lines 24-27, the aerosol species included in the SPRINTAR models are included. Even though the authors provide the reference for further information of the model, the size distribution of the dust and sea salt aerosols (presented in page 3441) should be given here.

The SPRINTARS has 6 bins for dust and 4 bins for sea salt. We added description about the size distribution of the dust and sea salt aerosols in section 3.1.

"SPRINTARS has 6 dust particle bins whose effective radii are 0.13, 0.33, 0.82, 1.27, 3.20 and 8.02 µm, and 4 sea salt particle bins whose effective radii are 0.18, 0.56, 1.78, and 5.62 µm."

6) Page 3439, lines 26-29, in line 26 lower layer is defined as sigma level <0.8 but then in line 29 the lower layer is defined as sigma level>0.85. Please correct. Furthermore, why use different sigma values in both, why not just use 0.8?

"5" was dropped in the text. We corrected to 0.85. Sigma level < 0.85 means the lower 4 layers.

7) Page 3439 and 3440, What about the overestimation of dust in the upper layers, is it also due to the reasons presented in the last sentence of section 4? Please clarify.

Yes. As explained in the txt, the underestimation of wet removal is responsible to the positive biases (overestimation). We edited the manuscript to clarify that.

8) Page 3441-3442, section 5.1, in experiments E4 and E5 the authors use fine and coarse mode AOT over land emulating Terra and Aqua satellite. The authors should be aware that even thought a fine mode product exist over land it has not been validated up to this day and is therefore not recommended to be used (Huneeus et al., 2012). A total AOT is provided over land that has been validated and that can be used in the inversion. To assess the real capability of the inversion system in inverse modelling applications realistic synthetic observations need to be used.

We do know the problems of AOT over land, and mentioned the notice of the land product in the manuscript (Lines 21-25 on Page 3441) referring to Remer et al. (2008). In addition to the larger uncertainty in the land product, the MODIS product cannot provide AOT over bright surface regions (such as desert) except for the Deep Blue product. This leads the much less coverage of the land product than the ocean product. In this study, as mentioned in the manuscript (Line 25 on Page 3441), Experiments 1-3 (E1-3), which assimilated fine- and coarse-mode AOTs over only ocean, were performed to assess the real capability of the system. Moreover, we also conducted two additional "sensitivity" experiments (E4 and E5) as mentioned at Lines 25-28 on Page 3441. In the sensitivity experiments (E4 and E5), we assumed the case if the MODIS product could provide fine- and coarse-mode AOTs over land with the same coverage and accuracy as over ocean, and evaluated how much that land product has impacts on the inversion (because the real land product would have much less impact comparing with the ocean product due to the combined AOT, the lager uncertainty and the less coverage). This is also because we use the same error over land than over ocean. We added more description about the sensitivity experiments in the revised manuscript.

"In the sensitivity experiments (E4 and E5), we assume the case if we could obtain fine- and coarse-mode AOTs over the land with the same frequency and accuracy as those over the ocean."

9) Page 3442, line 5, An observational error of 0.05 was used for the experiments. Why use the same error over land than over ocean. The errors characterizing the MODIS product over ocean and over land are defined differently reflecting the higher accuracy over ocean than over land. Zhang et al. (2008), Benedetti et al. (2009) and Huneeus et al. (2012, 2013) define the errors differently over ocean than over land. The reference provided by the authors is a study conducted over ocean and therefore does not apply to justify applying the same error over land. I strongly

recommend repeating the experiments with larger errors over land than over ocean or otherwise justify why you decide not to.

Please see the reply to the specific comment #08.

10) Page 3445, lines 2-5, which "AOTs for sulphate and carbonaceous aerosols" were directly assimilated? Please elaborate and explain or remove the sentence.

This sentence means that we performed two additional inversion tests. One used sulfate AOT, not total AOT. The other test assimilated AOT of carbonaceous aerosol. We revised the sentences as follows:

"We perform two additional inversion tests. One uses sulfate AOT to optimize SO2 emission. The other assimilates AOT of carbonaceous aerosol for carbon emission. The two tests achieve …"

11) Page 3445 lines 28-29 and page 3446 lines 1-2, the authors conclude from the difficulties to obtain larger improvements in cases where different aerosol species coincide, the inadequacy of fine and coarse mode AOT to identify major tropospheric aerosol species. The problem is not the classification of aerosols as presented by the authors but the amount of information that is provided to differentiate between species. In regions where sulphate aerosols coincide with carbonaceous and dust ones, as can be the case in Easter Asia, the system is provided with one piece of information to constrain all three species, clearly not enough. Additional information is needed, such as the Angström exponent as suggested by the authors. I suggest to reformulate the sentence and do so accordingly in the conclusions and in the abstract.

We wanted to present that, in ultimate case, the observations, which can identify differentiate between aerosol species, can have a significant impact on the inversion. However, in real case, such observations are available only at limited in-situ sites, and less useful to the aerosol inversion in the global scale. As the reviewer mentioned, our suggestion was not enough. In the revised manuscript, we suggested that fine- and coarse-mode AOT has not enough information for the inversion in the heavily polluted regions, and additional information (e.g., Ångström exponent, depolarization and color ratio) will has a significant impact. We edited the section 5.2, abstract and conclusion in the revised manuscript as follows:

[Section 5.2]

"The second implication obtained from the inversion experiment is the importance of information that differentiates between aerosol species."

"The Angstrom exponent, depolarization ratio, and color ratio provide characteristics of the aerosol layer (e.g., dominant aerosol species), and are also useful for aerosol inverse modeling."

[Abstract]

"The experimental results also indicate that information that provides differentiations between aerosol species is crucial to inverse modelling over regions where various aerosol species co-exist."

[Conclusion]

"Another implication obtained from the inversion experiments is that information that differentiates between aerosol species is crucial over regions where various aerosols are brought together and coexist."

"Measurement of aerosol characteristics (e.g. the Angstrom exponent, depolarization ratio and color ratio) would be useful."

12) Page 3470, Figure 10, Figure caption should include the units of the emission in each sub-figure. It is difficult to read.

Thank you for your useful comment. We inserted the units into the revised caption.

[Reference]

Remer, L. a., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanré, D., Mattoo, S., Martins, J. V., Ichoku, C., Koren, I., Yu, H., and Holben, B. N.: Global aerosol climatology from the MODIS satellite sensors, Journal of Geophysical Research, 113, D14S07, 10.1029/2007JD009661, 2008.

Talagrand, O., and Courtier, P.: Variational assimilation of meteorological observations with the adjoint vorticity equation. I: Theory, Quarterly Journal of the Royal Meteorological Society, 113, 1311-1328, 10.1256/smsqj.47811, 1987.