Response to Referee #1 for the manuscript: "Assimilation of POLDER observations to estimate aerosol emissions"

Dear Referee #1,

Thank you for reviewing our manuscript. Your comments help us to improve and define better some aspects of our work. Below you can find our point-by-point responses to all of your comments.

Best regards,
On behalf of all co-authors
Athanasios Tsikerdekis

Format

Questions
Responses
"Quotes from the manuscript and revised or added text."

Comments

What I missed is some information for the meteorological set up of the simulations with ECHAM-HAM climate model (e.g. if the simulations are nudged, spin-up time of the simulations, if is there is a specific reason for the selection of the year 2006).

Thank you for your suggestion, indeed this information was missing from the document. We added a supplement TableS 1 (shown below), along with all the references in the main manuscript. The TableS 1 is referred in the subsection 3.2 Experimental Setup: "A list of selected meteorological and aerosol options used for the experiments is presented in TableS 1."

In addition, the year 2006 was selected based on the availability of POLDER SRON observations. A sentence was added in subsection 2.1: "In the present study aggregated (1° × 1°) POLDER data are used in the assimilation for the year 2006. The year was selected based on the availability of POLDER aerosol products from the SRON retrieval algorithm."

TableS 1. List of selected meteorological and aerosol options of ECHAM-HAM used for the experiments.

Description (Reference)	Model Option
Horizontal resolution of 1.875°, corresponding to 192 x 96 grid cells. For RES_{LOW} only 3.75°	hres = T63
Vertical resolution of 31 hybrid sigma pressure levels up to 10hPa	vres = L31
Cumulus cloud convection scheme (Nordeng et al., 1994)	iconv = 1
Sub-grid-scale stratiform clouds scheme (Sundqvist et al., 1989)	icover = 1
Rapid Radiation Transfer Model for General circulation models (RRTM-G; Iacono et al., 2008)	-
Land surface model JSBACH (Reick et al., 2013)	-
Boundary layer parameterization (Stevens et al., 2013 and reference therenin)	-
Nudge vorticity, divergence, temperature and surface pressure to ERA5 reanalysis	-
Dust emission scheme (Stier et al., 2005) with updated East Asia soil properties	ndust = 4

Sea salt emission scheme (Long et al., 2011)	nseasalt = 7
Air-sea exchange parameterization for DMS emissions (Nightingale, 2000)	npist = 3
Kappa-Koehler theory for aerosol water growth (Petters and Kreidenweis, 2007)	nwater = 1
Size depended in-cloud and below-cloud scavenging (Tegen et al., 2019 and reference therein)	nwetdep = 3
Enable interactive dry deposition scheme (Tegen et al., 2019 and reference therein)	ndrydep = 1
Enable radiatively active aerosol	naerorad = 1

Maybe they authors could think of revising the title inserting also the term of assimilation.

The title has been changed to: "Assimilation of POLDER observations to estimate aerosol emissions"

Caption of Figure S1: I think it is wrongly written (f) sulphur dioxide (SO2). It should rather be WAT. Furthermore, the acronym WAT should be also defined in the caption and could be introduced in the text of the manuscript when the water uptake of soluble species (resulting to high AOD values) is discussed (e.g. lines 575-584).

The caption and the figure has been updated and corrected: "FigureS 1. Optical depth at 550nm of CTL_{ECHAM} for (a) dust (DU), (b) sea salt (SS), (c) organic carbon (OC), (d) black carbon (BC), (e) sulphates (SO_4) and (f) water condensed on the surface of aerosol particles (WAT). The global contribution of each species to the total aerosol optical depth at 550nm is depicted at the right bottom corner. Third and fourth row depicts the contribution of each species to total aerosol optical depth at 550nm in each pixel."

I addition, we explain the abbreviation "WAT" in the first reference of FigureS 1 on section 4.1: "The CTL_{ECHAM} AOD_{550} per species along with the optical depth due to condensed water on the surface of aerosol particles (WAT) is depicted in FigureS 1."

Caption of Figure S8: For consistency with the text, it should be noted as "CTLERA5" than simply "ERA5".

In FigureS 8a the ERA relative humidity is depicted, which is used to compute aerosol water growth in the experiments CTL_{ERA5}. The caption has been corrected to make that clear: "FigureS 8. The relative humidity of (a) ERA5 used for aerosol water growth in CTL_{ERA5}, (b) CTL_{ECHAM} and the difference (c) CTL_{ECHAM} – ERA5 for 2006 at 800hPa. The global mean, the global mean error (ME) and the global mean absolute error (MAE) is depicted at the right bottom corner of each plot."

line 20: I would suggest " over isolated island sites at the ocean" instead of "over isolated island sites over the ocean".

Corrected as suggested.

line 24: Define at some place the acronyms such as GFAS.

The full name along with the abbreviation has been added to the first reference of GFAS in the abstract: “The biomass burning changes (based on POLDER) can be used as alternative biomass burning scaling factors for the Global Fire Assimilation System (GFAS) inventory distinctively estimated for organic carbon (2.93) and black carbon (1.90), instead of the recommended scaling of 3.4 (Kaiser et al. 2012).”

line 48 and line 50: You may delete "note that" in both sentences.

Corrected as suggested.

line 50: It should read "(from 1m to several km)" instead of (about 1m to several km).

Corrected as suggested.

line 52: "Emissions from biomass burning" instead of "Emissions from biomass burning emissions"

Corrected as suggested.

Lines 54-56: Please define at some place the acronyms such as GFED4, FINN1.5, QFED2.4, FEER1.0 and GFAS.

The full name along with the abbreviation was added in the introduction where this datasets are mentioned: “Emissions from biomass burning are based on satellite measurements that are related to burned area and use emission factors to convert the burned dry matter into emissions of aerosol and gas species (Global Fire Emissions Database v4 (GFED4); Van Der Werf et al., 2017), active fire count (Fire INventory from NCAR v1.5 (FINN1.5); Wiedinmyer et al., 2011) or fire radiative power (Quick Fire Emissions Dataset v2.4 (QFED2.4); Darmenov & da Silva, 2015, Fire Energetics and Emissions Research version v1.0 (FEER1.0); Ichoku & Ellison, 2014 and Global Fire Assimilation System (GFAS); Kaiser et al., 2012).”

Line 62: Is the term "diversity" used throughout the manuscript, the proper or the common word to express the ratio of the standard deviation to the mean. In many studies the word "range" is commonly used.

Thank you for your comment. The range can be misinterpreted as the difference between the maximum and the minimum value in a distribution. In this study we defined relative diversity of a distribution as the ratio of standard deviation to the mean, as in the study Schutgens et al., (2020).

Schutgens, N., Sayer, A. M., Heckel, A., Hsu, C., Jethva, H., de Leeuw, G., Leonard, P. J. T., Levy, R. C., Lipponen, A., Lyapustin, A., North, P., Popp, T., Poulsen, C., Sawyer, V., Sogacheva, L., Thomas, G., Torres, O., Wang, Y., Kinne, S., Schulz, M., and Stier, P.: An AeroCom–AeroSat study: intercomparison of satellite AOD datasets for aerosol model evaluation, Atmos. Chem. Phys., 20, 12431–12457, https://doi.org/10.5194/acp-20-12431-2020, 2020.

line 70: I would suggest "at least" instead of "at best".

Corrected as suggested.

line 72: I would suggest the plural "precursors" instead of "precursor".

Corrected as suggested where applicable throughout the document.

line 74: Maybe "assessed" or "found" instead of "used".

Corrected “used” to “found”

line 79: I suggest " ...similar information and methods which they are not ..." instead of "... similar information and methods and are not ..".

Corrected as suggested.

lines 119-121: The sentence is not clear and it needs some rephrasing.

Thank you for noting that, the sentence was indeed unclear. It was rephrased as:

“Although this study was very insightful, the discretization of scattering enhancement factor based on RH could correspond to a diverse aerosol load for each model. The low and high RH conditions may have occurred in different times and dates for every model, as well as for the observations.”

line 273: It is not clear what is the setup of the DAS_{ERA5} experiment? Is it the data assimilation experiment in ECHAM-HAM using relative humidity for ERA5? Please clarify in the text.

Thank you for bringing this up. A short description was added in the subsection "3.2 Experimental Setup" as depicted below. In addition, the title of subsection was corrected from "3.2 The Local Ensemble Transform Kalman Smoother" to "3.2 Experimental Setup"

"CTL_{ERA5} quantifies the effect of the underestimated relative humidity in ECHAM compared to ERA5 on aerosol optical properties. CTL_{ERA5} uses the relative humidity of ERA5 for aerosol water uptake. Note that this modification affects only the simulated aerosol optical properties in ECHAM-HAM, while the simulated water cycle (precipitation and evaporation) of the model remains unaltered. A data assimilation experiments based on this new CTL_{ERA5} setup was conducted named DAS_{ERA5} in order to quantify the effect of overestimated relative humidity profile to the aerosol emission estimation."

line 298: "along with the MAE" instead of "along the MAE".

Corrected as suggested.

line 298: " 3-hourly differences between the Experiments – POLDER" . It is better to be more specific. e.g. 3-h differences of CTL_{ECHAM}-POLDER and DAS_{ECHAM}-POLDER.

Corrected as suggested.

lines 531-533: Considering the complexity of the loss and production processes that control the SO2 and SO_4 fate in the atmosphere mentioned in this sentence, it could be nice to have a link to section 4.4.2 that you discuss these processes (e.g. as discussed in Section 4.4.2).

Thank you for your suggestion, a sentence was added that refers to the subsection 4.4.2 at that point.

"Thus, highlighting that inter-model differences in SO_4, may be caused primarily by differences in gain and loss processes rather than differences in the primary SO_2 emissions. The production and loss processes of SO_4 are discussed in more detail in subsection 4.4.2."

line 544: It should be "Figure 10" instead of "Figure 9".

Corrected.

line 579: " ...matches the underestimation of RH by ECHAM-HAM (Figure 10c) while ..." I am rather confused here with the underestimation. Do you mean the small underestimation over ocean (Figure 10 c) below 500 m? Above this level there is clear overestimation of RH.

Thank you for noting this. I was referring to the clear overestimation, the sentence has been corrected.

"Consequently, over ocean aerosol extinction profile differences (Figure 11c) matches the overestimation of RH by ECHAM-HAM (Figure 10c) while over land this is not the case (Figure 11b and Figure 10b)."