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Comment

## ***Interactive comment on “Simultaneous assimilation of satellite NO<sub>2</sub>, O<sub>3</sub>, CO, and HNO<sub>3</sub> data for the analysis of tropospheric chemical composition and emissions” by K. Miyazaki et al.***

### **Anonymous Referee #2**

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Review of

Simultaneous assimilation of satellite NO<sub>2</sub>, O<sub>3</sub>, CO, and HNO<sub>3</sub> data for the analysis of tropospheric chemical composition and emissions by K. Miyazaki, H. J. Eskes, K. Sudo, M. Takigawa, M. van Weele, and K. F. Boersma

General comments:

The paper presents a comprehensive overview of a state-of-the-art chemical data assimilation system (DAS) for the global CTM CHASER based on a Kalman-Filter technique. CHASER-DAS assimilates satellite retrievals of NO<sub>2</sub>, O<sub>3</sub>, CO, and HNO<sub>3</sub> from different instruments to optimise the concentrations fields as well as emissions. The

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system is applied to assimilate each observation type separately and all simultaneously for three 1- month periods in 2006 and 2007.

The authors can be congratulated on their technical and scientific achievement because CHASER-DAS shows a convincing performance. With CHASER-DAS, the authors tackle important problems of chemical DA such as the instantaneous optimisation of concentrations fields and emissions as well as the definition of inter-species and species-emissions covariances of the background error. The performance of CHASER-DAS is carefully evaluated not only with independent observations but also with OSEs and self-consistency test using chi-square diagnostics.

It is an regretful omission of the paper, that authors did not discuss the benefit (or drawback) of including the emissions in the state vector for the realism of the assimilated concentration fields itself. It would have been very interesting to present a comparison of an assimilation run without emissions in the state vector (not included in the paper), with the full DA run and the emissions inversion run (both included in the paper). In particular the concentration profiles in the PBL and lower troposphere should be different, if optimised emissions are used.

Improving emission data sets by inverse methods (top-down) is a fascinating research topic. One of the important question is whether the optimised emissions compensate for other model errors, e.g. in the chemical mechanism. The authors should have discussed this point in more detail. Do they believe that the optimised emissions from the inversion run or the one from the full DA provide a better estimate of the emissions. The evaluation of an independent model run using the optimised emissions could have clarified this. Further, is the DAS, which is built on the assumption of unbiased model and observations, able to correct emission biases or does it only minimise the variance of the emission error (i.e. by introducing an improved variability)

The paper is very long and could be shortened or re-arranged for better readability. On occasions, one has to go through a lot of text until one reaches the truly interesting

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pieces of information. The authors should consider if the general description of the considered species in the introduction can be omitted or shortened. The presentation of the observations used for assimilation (2.1) or evaluation (2.2) contains a lot of detail, which is important in general, but can be found elsewhere and is not so much of interest for the focus of the paper. Moving this section into an appendix might improve readability. The presentation of the evaluation of the data assimilation experiments is very long. The reader is easily convinced that CHASER-DAS overall improves the match with independent or assimilated observation. Providing more and more numbers is not needed. The evaluation statistics are already provided in tables.

Specific comments:

p 16133 | 17: be more specific about “large” uncertainties, provide references and numbers

p 16133 | 25: provide references

p 16134 | 16: SAGE II and OSIRIS provided early NO<sub>2</sub> observations

p 16135 | 15: provide more detail on the differences of CHASER-DAS with other chemical data assimilation systems presented in the literature. This includes the multi-species vs. uni-variate approaches.

p 16135 | 18: “stiffness” is a numerical property of a system of ODS representing the chemical mechanism. Stiffness is caused by the different time scales and causes numerical instability. It can not be argued that this stiffness “damps out” perturbation or that it the reason that CTM simulations depend not only on initial conditions. The initial conditions are less important because of the boundary conditions (i.e. the fluxes) and because of the chemical equilibrium.

p 16135 | 25: please provide reference for the claim that uncertainty of emissions is main source of model error

p 16135 | 25: The theory of the KF assumes a bias free model (BLUE). The emissions

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inventories are not only uncertain in terms of variability but they are “biased”, .i.e. the most probably have a constant off-set. Please explain better how a KF system based on BLUE can fix the emission bias.

p 16136 | 15: Please mention other DA systems, which optimise concentrations and model parameters such as Elbern et al. (2007 et al) and Hanea et al. (2004)

p 16137 | 7: is yo-yb the satellite - analysis difference (?) yb is defined as the model field in observations space

p 16137 | 13: “for a large part” - please explain or omit

p 16137 | 17: consider moving section 2.1. and 2.2 into an appendix

p 16138 | 18: (=2.8 degree ), please provide resolution in km, omit “=”

p 16140 | 6: MOPITT also measures in the 2.2-2.4 microm interval

p 16143 | 25: “both peak” is unclear - does this mean they have a maximum ?

p 16146 | 15: be precise, convective cloud top height is the parameter used in this parameterisation

p 16147 | 3: Please summarise briefly differences between LETKF and the more common EnKF.

p 16147 | 8: Please explain how the ensemble X is obtained (variation of which model parameter)

p 16147 | 16: Please explain how Pa is projected in time (this is the key feature of any Kalman Filter), Please explain how the model error is taken into account, and if not how deflation of Pa is avoided.

p 16148 | 11: explain choice of period and its length

p 16148 | 22: see comment on stiffness above

p 16149 | 11: Does cib include all species including the very short-lived like OH?

p 16149 | 19: It is not clear how the localisation, i.e. the suppression of certain covariance values, was derived from the sensitivity experiment. It seems more a practical but ad-hoc choice. Please clarify, and provide more detail. The choice of this localisation might be useful for other chemical DAS.

p 16150 | 10 ff: this paragraph would be better placed in section 3.3

p 16150 | 27: please mention that the covariance deflate because no model error is assumed.

p 16151 | 5: Was the error specification of the emissions constrained in any other way apart from the initial error value? Were large day-to-day changes damped (by using a “red noise” error model) Were points with low emissions (say over the oceans) restricted to maintain low emissions by the DA system.

p 16152 | 9: The term “localisation length” is not clear - is it the horizontal correlation length ? Is there an assumption about the vertical correlation length?

p. 16153 | 9: Figure 3 is a very interesting figure. It is not clear how the “global mean” was constructed. Is it the spatial means of the covariance for each grid point or is the covariance of the global mean state vector. Is there a distinction between day and night grid points?

p 161f3 | 9: Why does Figure 3 not contain OH - this would have been very interesting as well.

p 16153 | 10 ff: This discussion is interesting but should not only focus on the positive correlations. Because of the chemical nature there are also many negative correlations between the species (as many as positive) These should also be discussed. For instance the strong negative correlation between Ox and ethylene should be explained.

p 16154 | 9 - 15: Where is this statement backed up - is there a figure available?

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p 16156 I 25: chi-square tests are a large class of methods. The chi-square diagnostics by Menard and Chang (2000) is motivated by chi-square testing.

p 16164 I 15 - 25: I do find this reasoning a bit far fetched. It remains speculative and assumes a perfect representation of the chemistry by the model. The assimilation of MLS HNO<sub>3</sub> is clearly not efficient enough to improve HNO<sub>3</sub> overestimation in the lower and mid troposphere (Figure 13). I do not see that adding more NO<sub>x</sub> (more lightning or more convection?) will help to reduce the positive OH bias.

p 16166 I 1-5: The emissions are part of the state vector. If I understand the DA method correctly, the optimised emissions are available for each analysis time step and each surface grid point. It would therefore be interesting not only to discuss the bias but also the temporal and spatial variability of the optimized emissions. Is this variability, w.r.t. to time and space, large compared to the bias? Is there a temporal trend of the emissions within the time frame? By including the emissions in the state vector, they can simple vary much more than the emissions from the static inventories.

p 16168 I3: provide reference

p 16168 I15: Table 8 shows much lower numbers for new inventories despite increased emission inversion results for Europe. Please discuss this further.

p 16170 I 15: The low CO bias might also be caused by an overestimation of OH. Increasing emissions alone seems not the entire solution.

p 16173 I25: please also mention the deterioration of some of the fields by DA in the lower troposphere (see Figure 13)

p 16174 I 16: Please discuss the benefit and drawbacks of (i) emission inversions, (ii) concentration field DA and (iii) combined concentration field emission DA

p 16174 I 20: “somewhat different” - please be more specific

p 16175 I12: What are the conclusion of the paper w.r.t to model improvements - just

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changing emissions? This assumes that emissions are the only source of the model error, which is perhaps not true. The optimised emission from the full assimilation run and the emission inversion run differ - why. Is a model run with the optimised emissions as good as the full DA run ?

p 16204 figure 2: clarify 260 hPa / 220 hPa

p 16205 , figure 3: Mention averaging period, please clarify Ox, why is OH not included.

p 16197: table 4: explain localisation length

p 16201, Table 8: are the lower values for newer emissions for Europe and E-USA correct?

p 16216, figure 14: Why are there no lightning emissions over the oceans?

Reference:

Elbern, H., Strunk, A., Schmidt, H., and Talagrand, O.: Emission rate and chemical state estimation by 4-dimensional variational inversion, *Atmos. Chem. Phys.*, 7, 3749-3769, doi:10.5194/acp-7-3749-2007.

Hanea, R. G., G. J. M. Velders and A. Heemink, Data assimilation of groundlevel ozone in Europe with a Kalman filter and chemistry transport, *J. Geophys. Res.*, 109, (D10302), 2004.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 16131, 2012.

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