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Atmospheric Chemistry and Physics Discussions

Interactive comment on "How consistent are top-down hydrocarbon emissions based on formaldehyde observations from GOME-2 and OMI?" *by* T. Stavrakou et al.

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The authors would like to thank the reviewer for the positive evaluation of the manuscript, the careful reading and the useful comments and suggestions. Below we address the raised concerns. The reviewer's comments are *italicized*.

This is an interesting paper that describes the inversion of HCHO columns of GOME-2 (morning orbit) and OMI (afternoon orbit) in the IMAGES model, with the aim to optimise emissions of isoprene, from biomass burning and anthropogenic HCHO sources. Two consistent source of satellite data provide the interesting possibility to study the

diurnal behavior, which is done in this paper. The results are interesting, but unfortunately, the paper is rather lengthy with many figures, which does not stimulate (full) reading. The paper provides an interesting contribution, after the following issues have been addressed.

Major issues :

1. *The structure of the paper is somewhat messy. An example is section 2.2. Here the reader gets very detailed information about anthropogenic VOC emissions and their chemistry without knowing the HCHO budget. This budget should certainly be included in the introduction. I also suggest to move section 2.2 to an Appendix, be- cause it distracts from the main aim of the paper. In the introduction, the authors partly describe their method (e.g. page 12012, l24 and further). Also the paper is a bit short in referencing work of others, and how this study fits in existing knowledge. So, the introduction should be improved in this respect. Further, in section 3.1 reference is made to sensitivity simulations before they have been introduced. It would therefore be good to first do a complete method section, before the discussion of the results. Also, on page 12024, section 5, part of the method is introduced in a section entitled: "Overview of the results". A proper method section would certainly improve the paper. This also gives the opportunity to introduce terms like "ccost function", terms that now pop up without any reference.*

   All points suggested by the reviewer have been adopted. More specifically,

   — the Subsection 2.2 is moved in the Supplement. Its content is briefly presented in Section 2;

   — a new table is added (Table 1 of the revised version) summarizing all forward sensitivity simulations used to investigate the diurnal variation of HCHO columns. The table is introduced in the first paragraph of Section 3.1;

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— a new section “Inversion methodology" (Section 5) has been added with information on the technique and material from the section “Overview of the results" of the first version of the manuscript;

— the global budget of HCHO is discussed in the last paragraph of Section 2;

— more references have been added in the introduction of the revised version.

2. *The description of the model is slightly misleading. As far as I know, IMAGES uses monthly mean meteorological fields to transport and mix the tracers. This important issue is not clearly mentioned. It would be good to add this, and also add a discussion of its potential impact on the inversion. I expect some impact on the inversion, because of difficulties of co-sampling the model with the observations and potential clear sky biases. Also, in comparing with aircraft observations on page 12027, line 11, there might be issues with monthly-averaged winds, and some words of caution are required.*

The referee is correct that a description of the model transport was lacking. IMAGES uses monthly winds but daily or subdaily fields for other meteorological fields (convection, PBL mixing, temperature, water vapor, rain and cloud fields) and for biogenic isoprene emissions. The model description now includes the following text: “Meteorological fields are obtained from ERA-Interim analyses of the European Centre of Medium-Range Weather Forecasts (ECMWF). Advection is driven by monthly averaged winds, while the effect of wind temporal variability at time scales shorter than one month is represented as horizontal diffusion (Müller and Brasseur, 1995). Convection is parameterized based on daily ERA-Interim updraft mass fluxes. Turbulent mixing in the planetary boundary layer uses daily diffusivities also obtained from ERA-Interim. Rain and cloud fields (and therefore also the photolysis and wet scavenging rates) are also based on daily ERA-Interim fields. The effect of diurnal variations are considered through correction factors on the photolysis and kinetic rates obtained from model simu-

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lations accounting for the diurnal cycle of photorates, emissions, convection and boundary layer mixing".

Regarding the co-sampling of model and observations, the Section 4 now clarifies that “the simulated monthly averaged columns are calculated from daily values weighted by the number of satellite (OMI or GOME-2) measurements for each day at each model grid cell." Any clear sky bias in the satellite data is therefore taken into account in the model averages.

Finally, the use of monthly winds is not a serious issue in the comparisons of the model with formaldehyde data. Due to the short lifetime of formaldehyde and its precursors (except CH_4 which is well-mixed), long-range transport does not play a significant role, as illustrated by the fact that several recent inverse modelling studies (Palmer et al., 2003; Millet et al., 2008; Barkley et al., 2013) derived isoprene emissions from formaldehyde columns without considering the effects of horizontal transport. To substantiate the weak sensitivity of formaldehyde to the wind fields, Figure 1 illustrates the impact of 1) halving the wind components in the model (left panel), and 2) using the winds of 2005 instead of 2004 (right panel) on the calculated CH_2O mixing ratios during INTEX-A. The differences are minor at most locations. The correlation coefficient between the observed and modelled values is decreased from 0.811 in the standard run to 0.806 and 0.809 when using alternative wind sets.

3. *In the discussion I also would expect some reflection of the separation of biomass burning sources, anthropogenic sources, and isoprene sources. In general, the inversion should give error reductions, and also the posterior co-variance terms that would reflect the ability to separate the different sources. I understand that an error estimate is more difficult for a non-linear system, but the sensitivity experiments give some room for error discussion. But statements on page 12030, line 15: “Chinese isoprene emissions are decreased from 7 Tg year-1 to 6.5 Tg (OMI) and 5.9 Tg (GOME-2)" need to be accompanied by error estimates. I can-*

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not imagine that you can properly separate isoprene HCHO sources from other sources.

In response to a comment of Reviewer 1, we have now included a discussion of the possible confusion between biomass burning and biogenic emissions over Russia. Over China, biogenic emissions have a distinct seasonal variation and are mostly located in the South, which reduces the possibility of confusion with other categories. We agree however that the separation of posterior emissions between different categories is tentative and subject to errors. However, providing reliable error reductions and posterior co-variances is complex and somewhat out of scope since the main focus of the present article is a comparison of the inversion results obtained from two sensors. The sensitivity simulations provide indeed some hint regarding the possible uncertainties, and we tried to incorporate their results in the discussion to better reflect the uncertainties.

4. *Units: please check all the units in the paper. They are often missing or incorrect (e.g. TG instead of Tg/year). Also check and add legends to figures. e.g. figure 12: does this show TG/month?*

   Units are added where incomplete or missing.

Minor issues :

1. *12009, l 23: units are missing*

   Units are TgVOC/yr and have been added.

2. *12009, l 25: add per year in the unit*

   Done.

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3. *12011, l 1: CO and H_2 (add H_2)*

   Added.

4. *12012, l23: The inversion framework is assumed known to the reader. I think it would be could to describe this a bit better in the introduction, i.e. also by referring to earlier studies in this field by other groups.*

   The new Section 5 “Inversion methodology" provides some detail about the inversion methodology used to derive top-down VOC emissions. References to earlier studies is provided in the introduction of the revised version.

5. *12013, l2: Here method and introduction are mixed. I would prefer in the introduction references to studies that show the need for these sensitivity studies (e.g. associated with diurnal cycle of emissions). Referring to “inversion design" is a bit too short and methodological.*

   – The new Table 1 summarizes the sensitivity studies undertaken to investigate the impact of different parameters on the diurnal variation of the HCHO columns. This Table is introduced in the first paragraph of Section 3.

   – The sentence now reads : “Sensitivity studies are carried out to assess the robustness of the findings to different assumptions, e.g. to changes of the prescribed a priori errors on the emission fluxes in the inversion."

6. *12014: l7: add unit kg/kmol (or g/mol)*

   Units added.

7. *12014: l14: “The African. . ..worldwide". Maybe good to add some cautious remarks here. Over peat fires (e.g. Russia in section 6.2) this assumption is certainly not valid, and maybe also not for boreal fires.*

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The sentence now reads : “This profile is in fairly good agreement with the averaged diurnal cycle of active fire observations constructed from the GOES geostationary satellite encompassing North, Central and South America (Mu et al. 2011), and therefore it is applied to all fires worldwide. Note, however, that this specific temporal profile might not be appropriate for some locations, e.g. peat fires over Russia."

8. *12015, l9: I miss somehow some recent references, e.g. Fuchs, H. et al. Experimental evidence for efficient hydroxyl radical regeneration in isoprene oxidation. Nature Geosci. 6, 102ž20131026 (2013).*

   Reference added.

9. *12017, l3: I would use something like (g CHCO/g OAHC) as unit here.*

   Replaced, cf. Supplementary material.

10. *12018, l10 (and further on): on Fig. xx → in Fig. xx*

    Corrected.

11. *12020, l14: please repeat that you evaluate the diurnal cycle in the column, and not in the near surface concentration.*

    The sentence now reads : “To evaluate the diurnal cycle of the modelled HCHO column..."

12. *12022, l3: here I wonder why the modeled HCHO concentrations in the boundary layer are not compared to observations. I agree that boundary layer mixing complicates issues here, but the authors should al least argue why they did not evaluate the model with other HCHO measurements. Also, by comparing only diurnal profiles they might hide deficiencies in the model.*

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We now provide in the Introduction a more clear argument for using ground-based column measurements instead of in situ concentration measurements: “Field campaign measurements show that the diurnal patterns of surface HCHO concentrations are mostly influenced by the magnitude and diurnal variability of precursor emissions and the development of the boundary layer. (...) Long-term diurnal measurements of HCHO columns are limited, but are less influenced by variations in boundary layer mixing and are directly comparable with the satellite observations."

13. *12023, l29: on this figure → in this figure*

    Corrected.

14. *12024, l16: Table 2 lists other sensitivity studies than described earlier in the discussion of the diurnal profiles. I suggest to include one table with all simulations performed.*

    A new Table is added summarizing all forward tests performed to investigate the diurnal cycle of HCHO columns. Section 3 and the caption of Figure 2 make reference to this Table.

15. *12031, l8, acronym IASI is introduced, but was used before*

    The acronym is now mentioned in the introduction.

16. *12033, l3: contrasted → contrasting*

    Corrected.

17. *12033, l14: Tropical Asia emissions have been studied using IASI: (Basu, S. et al. The seasonal variation of the CO_2 flux over Tropical Asia estimated from GOSAT, CONTRAIL, and IASI. Geophys Res Lett 41, 1809–1815 (2014))*

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Agreed. We modified the discussion of the IASI CO results over Indochina (Section 8.3) as follows: “Indeed, as seen on the lower panel of Fig. 14, modelled CO simulations using biomass burning fluxes optimized using OMI data (i.e. reduced by ca. 26% in March relative to GFED3) display a better agreement with the observed CO columns, despite an underestimation by ∼10% over most of the peninsula. This result is consistent with the moderate reduction (ca. 20% in March) of biomass burning emissions of CO over Tropical Asia inferred by Basu et al. (2014) in an inversion based on IASI CO columns utilizing the TM5 atmospheric model with GFED3 as a priori inventory. "

18. *12035, l27: and (to a lesser extent) meteorological parameters. It is unclear what is meant with this statement.*

    The influence of the diurnal cycle of meteorological parameters on the diurnal cycle of HCHO columns is expected to be minor, as confirmed by sensitivity simulations (with diurnally constant convection and PBL mixing). Photochemistry and the emissions are the main players. The influence of meteorological parameters is mentioned here for completeness.

19. *Figure 2: Please use a common y-ax metric.*

    Done.

20. *Figure 9: the order of the panels does not make sense. Jan-mar-aug-oct? why not Jan, Apr, Jul, Oct?*

    March and August coincide with the fire peak season in Indochina and Amazonia, respectively. Because these regions are discussed in detail in our manuscript, we believe that including these two months (instead of April and July) in Figure 9 makes more sense and facilitates the discussion.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 12007, 2015.

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Interactive Comment

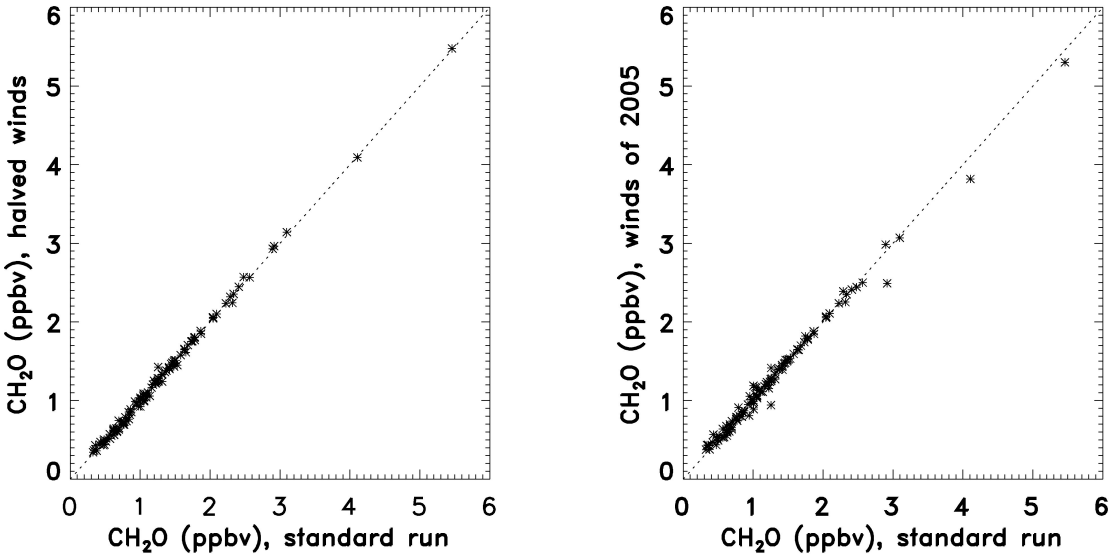

Fig. 1. Comparison of the simulated HCHO concentrations during INTEX-A in the standard model run and in a simulation using halved winds (left) and a simulation using 2005 winds (right).

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