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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluating the performance of pyrogenic and biogenic emission inventories against one decade of space-based formaldehyde columns" *by* T. Stavrakou et al.

T. Stavrakou et al.

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The authors would like to thank the referee for his/her constructive criticism and comments which contributed to substantially improve the manuscript.

Please note that the main changes of the revised manuscript can be summarized in the following points.

- To reply to Referee's#1 comment, a new section has been included (Subsect. 4.2), providing a tentative assessment of model errors.
- The section on the description of the HCHO dataset has now been shortened, as suggested by Referee#4.

- To comply with the Referee#2 and #4 request to shorten the manuscript, the descriptions of the emission databases have now moved to the Supplemental material (Part A).
- The model results presented in the revised manuscript are obtained with a model time step of 3 hours (instead of one day). This change does not affect much the results and the conclusions.
- The error bars in Fig. 6 and Figs. 8-12 now represent the retrieval error estimated by De Smedt et al.,2008.
- To reply to the Referee's#4 comment, we have now added two tables (Table 5 and 6) with the average biases and the spatiotemporal correlation coefficients over large regions for the burning season and for the rest of the year.
- The abstract, the Section 5 and the conclusions are reformulated to reflect the existence of uncertainties in the HCHO retrieval, especially over fire scenes, as requested by all referees.

A point-to-point reply to the referee's#1 comments (in italics) follows.

The work presented in this article shows how satellite observations can be used to constrain biogenic and pyrogenic VOC emissions. The strengths of this work is to use long term (a decade) and state of the art global scale data sets of tropospheric formaldehyde columns (from GOME and SCIAMACHY) and of biogenic (MEGAN-ECMWF) and pyrogenic emissions (GEFD). A weakness is that conclusions drawn from the comparison between observed and simulated formaldehyde columns on the validity of emission data bases are overstressed, because they do not take into account uncertainties in observations and in simulations. A second weakness is that the modelling approach with the IMAGE model remains somewhat unclear because monthly mean and daily/hourly

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input data are mixed. All in all, this work is an interesting contribution to the field, which should be published in ACP after correcting the two weaknesses pointed out.

The authors state that uncertainty in observed formaldehyde columns is about 20 to 40%, depending on cloudiness conditions. In several parts of the globe, strong differences (several tenths of %) with formaldehyde columns derived from GOME by other groups (using other spectral windows and different set-up of retrieval methods) are also noticed by the authors. Last uncertainties in simulations with the IMAGE model are not assessed, but could be also significant. In the context, biases of a several tenths of % between simulated and observed HCHO columns cannot be related to the accuracy of VOC emission data bases, but are simply within the error bars of observations. This should be made clear, and it should be stated which discrepancies are larger than combined observation and simulation uncertainties. Still the phase (timing) of the seasonal and interannual variability can be analysed from this data-set.

We fully agree that caution is necessary when presenting comparisons between observed and simulated formaldehyde columns. Conclusions drawn from these comparisons are now formulated more carefully to reflect the existence of uncertainties, when appropriate. The model uncertainties are now discussed in a new subsection (Subsec. 4.2). Sensitivity calculations have been performed to evaluate the impact of possible errors related to the transport scheme, the OH and photolysis fields, the chemical mechanism, the numerical implementation of the model, etc. (see Table 3, also the Supplemental information file, Part C). A crude evaluation of the associated errors is provided (Table 3). The largest errors (of the order of 10%, but as much as 20% in several areas) are those related to the isoprene degradation mechanism.

Implications of mixing monthly and diurnal/ hourly input data on the accuracy of simulations should be made clear. What is the impact of using monthly averaged wind fields (and of diffusion coefficients to mimic the synoptic variability) on the accuracy of simulations (section 3.1)? Have there been comparisons between IMAGE simulations and CTM simulations using wind fields more resolved in time (several per day)?

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Interactive Comment

The most prominent NMVOCs responsible for the strong HCHO enhancements over source regions (isoprene, ethene, propene, 2,3-butanedione, etc.) have sufficiently short lifetimes (several hours) that their long-range horizontal transport is very limited. The inverse modeling studies of the Harvard group using the GEOS-Chem model relied on the assumption of a local relationship between the NMVOC emissions and the HCHO column, implying that horizontal transport is believed to play at most a minimal role in the HCHO distribution over source regions.

Given the model time step of one day, a special correction (by off-line diurnal modelling) is applied to obtain a diurnal variation in concentration fields (necessary for example to compare to observations during satellite overpasses). What are the details of this method? Is it applied at a given day, for given 0D boxes ? Can the model uncertainty due to these and other issues be estimated (i.e. accuracy of OH fields? This would be important in order to correctly assign uncertainty to emission data.

Note that the model time step is now 3 hours, instead of one day, in the simulations presented in this manuscript. As explained above, the model errors are explored through a series of sensitivity simulations. In particular, the model has been run with a fully explicit diurnal cycle, a time step of 20 minutes, and the Rosenbrock chemical solver embedded in the KPP package. The results show that the numerical implementation adopted (long time step, fast chemical solver and use of correction factors determined by off-line diurnal modeling) induce only small errors on the calculated HCHO columns (of the order of 3all model grid cells in the off-line simulation, as is now added in Subsection 2.2.

Specific remarks :

Page 16989: How big is uncertainty and bias (with known sign) in HCHO columns introduced by neglecting the aerosol correction, especially for strong biomass burning events ? This again affects the utility of observations to constrain emissions in section 5.

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As already stated in the manuscript, absorbing aerosols can lead to reduction of the air mass factor of up to 40% over hot spot areas. Radiative transfer calculations by Fu et al. (2007) indicated that the magnitude and even the sign of the aerosol effect are influenced by the aerosol properties and by the vertical distribution of both the aerosol layer and the HCHO concentration. However, calculations with the GEOS-Chem model suggest that, on average, absorbing aerosols tend to decrease the HCHO air mass factors (AMFs). Therefore, the omission of the aerosol correction may lead to a significant underestimation of the derived HCHO column over fire scenes, by up to about 40%. This is now mentioned in the manuscript (Subsec. 2.1). As a consequence, the overestimations of the modeled columns over several biomass burning areas (e.g. over Amazonia for GFEDv1, over Indochina and Indonesia for GFEDv2) might be partly or entirely due to the absence of aerosol correction in the retrievals. Conversely, underestimated columns by the model (e.g. over Amazonia for GFEDv2, over Indonesia for GFEDv1, etc.) probably imply a large underestimation of the NMVOC emissions in the model.

Page 16993: It should be stated that biogenic emission inventories are monthly means (but with superimposed average diurnal variation). That is what I understood given the monthly average met. input data. The sentence "In addition to this, the emissions of the MEGAN-based inventory account for the diurnal, daily, seasonal and year-to-year variability" at page 16995 is then misleading.

The model uses daily isoprene emission fields, as already stated in the text. Besides the winds, all meteorological fields are daily averages (with superimposed diurnal variation in the off-line diurnal cycle calculations).

Page 16998, section 4.3: The text suggests that HCHO production in the current study is calculated from emissions and yields given in Table 2. Please make clear that values in Table 2 are given only for a comparison purpose, and that HCHO formation is explicitly treated within IMAGE.

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Corrected, see Subsection 3.3.

Page 16999 : Use monthly OH, HO2, NO, NO2 and NO3 fields for sensitivity tests. Wouldn't it be more coherent to include O3 in the list of fixed species ? It is an oxidant as NO3, and governs the NO/NO2 ratio.

Ozone is only a minor oxidant for the NMVOCs considered in this study. Both NO and NO2 are kept fixed, therefore the NO/NO2 ratio is not influenced by ozone in these simulations.

Page 17002 : discussion of differences over North America. Here conclusions are drawn on the relative validity of the GEIA and MEGAN-ECMWF data base from biases with HCHO columns. But at the same time, strong differences to other satellite datasets of HCHO columns are noted, which tend in my view to make the previous statement invalid.

Conclusions are not drawn in the manuscript on the relative validity of the different biogenic emission databases over Northern America. The strong differences to other satellite data sets have been mentioned in this section and elsewhere in the manuscript.

Does the calculated correlation coefficient refer to temporal or spatio-temporal correlation? Please make this clear. Later on, the text suggests spatio-temporal correlation (page 17003): "The high correlation coefficient values (0.9) calculated over the extended Amazonian region, Guatemala and Santarem yield strong confidence to the spatiotemporal distribution of the implemented emission inventories."

The calculated spatio-temporal correlation coefficients and biases are now summarized in Table 5 (for the burning season) and Table 6 (non-burning season).

Page 17003 : "Note finally ? in Palmer et al. (2006)." This paragraph is difficult to understand. Please clarify it.

This paragraph has been removed, being not much relevant to the discussion.

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Interactive Comment

Page 17005 : Comparison over Africa during biomass burning events. Please indicate if the fact not to take into account biomass burning aerosols in the retrieval could explain the differences.

The overestimation by up to 100<cannot be explained by biomass burning aerosols and most probably reflects an overestimation of emissions. The focus of the discussion over Southern Africa is on the timing, not the amplitude, of the emissions.

Page 17007, line 21 : typo by ouR comparisons

Corrected.

Page 17009, Conclusion : "The high correlation coefficients" Please note that for the extreme case of a seasonal variation given by a function (i.e. sinusoidal), correlation is only sensitive to the phase and not to the amplitude. Some of the seasonal variations (of monthly means) are not far from sinusoidal functions, so the interpretation of a good correlation should not be overstressed.

We agree that temporal correlation coefficients are sensitive to the timing of the emissions more than to their amplitude. However, it is worthwhile to mention if and when the model and the data show a similar seasonal behavior. Note also that the correlation coefficients given in the revised manuscript are spatio-temporal correlation coefficients.

Page 17010 , line 18 : typo puts

Corrected.

Page 17011 "It is understood that our conclusions depend vitally on the quality of the retrieved columns. However, discrepancies among the retrievals, inherent to differences in the retrieval methods, exist between HCHO datasets. For instance, our GOME slant columns are by 5 about 30-40% lower than the Chance et al. (2000) dataset over North America (Palmer et al., 2006) and desert regions, whereas over central/Southern Africa, HCHO columns used in Meyer-Arnek et al. (2005); Wittrock et al. (2006) are by 40% lower than in the TEMIS dataset." The authors should be made clear which of the

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conclusions still hold given these discrepancies.

We feel that this request is a bit unfair. The reader is warned about the existence of these discrepancies. Previous model/data comparisons provided useful information on the differences between a specific model (GEOS-Chem) and a specific retrieval (the Chance et al. retrieval). We provide here an extensive comparison between another model and another retrieval, and we point out important differences regarding the implications for NMVOC emissions. Our conclusions clearly contradict previous findings e.g. over the U.S. A systematic intercomparison of the HCHO retrievals is clearly needed in order to provide a clear picture of the differences. More importantly, the reasons for these differences need to be elucidated.

Figure 1 : What is the unity of the represented value ?

Units are μg/m2/h. This is now indicated in the figure caption.

Figure 8 : What exactly represent the error bars ?

They represent the HCHO retrieval error as determined by De Smedt et al. (2008). It is now mentioned in the caption of Figs. 6 and 8.

Figure 10 and following : Lines colors like in Fig. 8 (not 10)

The lines codes are now given in all figure captions, when applicable.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16981, 2008.