

Interactive comment on “Adjoint inversion of Chinese non-methane volatile organic compound emissions using space-based observations of formaldehyde and glyoxal” by Hansen Cao et al.

Anonymous Referee #1

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This study reports top-down estimates of non-methane volatile organic compound emissions over China based on formaldehyde and glyoxal column observations from two sounders, OMI and GOME-2 for 2007. Based on model simulations with the adjoint of the GEOS-Chem model, Cao et al. analyze the impacts of the different satellite datasets on the top-down emission estimates. They find that the annual total top-down VOC emission amounts to 30 Tg C, by 10% higher than the a priori inventory. In addition, using glyoxal retrievals from OMI, the authors estimate the annual aromatics Chinese source from 5 to 7.3 Tg C, also higher than in the bottom-up inventory. This study addresses an interesting subject for Atmospheric Chemistry and Physics journal. However, there are several weaknesses in the current work. For example, the figures

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are not informative enough and cannot properly feed the discussion, the tables appear in an illogical order, some key statements appear without citation, references are missing. In addition, I see contradictions in the top-down emission estimates mentioned in the abstract and not enough details (and possibly errors) in the chemical scheme. Therefore, I have doubts regarding the validity of the conclusions and think that the manuscript will need a major revision before it becomes suitable for publication.

General comments :

- The chemical mechanism described very briefly in Section 2.1 is the core ingredient of the top-down VOC studies.
 - In l.164-165, several NMVOC precursors of formaldehyde are mentioned, but key precursors like methanol, acetaldehyde, ethanol, acetone, etc. do not show in the list. Why are these compounds omitted? Provide also more details on C4 alkanes (l.165).
 - In l.166 propane and (higher) alkanes are mentioned as glyoxal precursors. I have serious doubts on this. Please elaborate on the degradation scheme leading to glyoxal in your model.
 - l.170-172 : provide more details on how glyoxal is formed at both high- and low-NO_x levels.
 - l.172-176 : I don't get this. Li et al. (2016) discusses the AM3 mechanism, not the GEOS-Chem mechanism. Furthermore, the statement that the updated scheme matches the MCM yields is not correct, the NO_x-dependence of the yield is completely different in the two schemes.
 - Provide a table with formation yields at high and low NO_x conditions for formaldehyde and glyoxal from their respective precursors.
- The comparisons between emission estimates shown in Table 1 relies heavily on conversion factors of 0.84, 0.57 and 0.85 for anthropogenic, biomass burning

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and biogenic VOC, respectively. There is no reference on how these numbers are calculated. In particular, for isoprene and monoterpenes the factor of 0.85 is wrong. For methanol the real factor is also much lower.

- Table 1 misses emission estimates from widely used recent bottom-up and top-down inventories, e.g. GFED4 (van der Werf et al. 2017) on biomass burning emissions, HTAPV2 (Janssens-Maenhout et al. 2015) and EDGARv4.3.2 (Huang et al. 2017) on anthropogenic emissions, MEGAN-MACC (Sindelarova et al. 2014) and MEGAN-MOHYCAN (Stavrakou et al. 2014) on biogenic VOC, MACCity (Granier et al. 2011) on global anthropogenic and fire inventories. Especially for China, top-down estimates from Fu et al. (2008), Bauwens et al. (2016), Stavrakou et al. (2017), Granier et al. (2017) are missing.
- The Table ordering is illogical. Table 3 should rather become Table 1 or move to the supplement. Table 2 describes the simulations, so it should come first. Table 3 shows results and comparisons to previous studies so it should be called in the result section.
- In the abstract you mention that the annual total NMVOC emissions ranges from 23.5 to 35.4 Tg C (mean of 30.8). This does not match the sum of individual categories given in lines 27-29 of the abstract (23.5-36 Tg C). This brings confusion to the reader already from the first lines. Which one is correct? Change accordingly throughout the paper and the Tables. In l.29-30 provide a name for the "most widely used bottom-up inventory".
- l.231 : Do you mean 19.8 Tg C from Table or am I missing something? I have several doubts about the reported numbers. Check again before you resubmit.
- In l. 239, the uncertainty of a priori emissions is given, $\pm 200\%$. Is this what is really meant here? It would correspond to a range of -20 to 60 Tg C. This makes

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no sense given the reported numbers for the anthropogenic flux from different inventories. Same for l. 224, 267.

- l.249 : Liu et al. (2015) is based fire radiative power, not burnt area.
- l. 265 : GFED4 (van der Werf et al. 2017) accounts for agricultural fire burning, which was not the case in GFED3. You should compare with GFED4 for this emission category.
- There are many language errors in the manuscript. This decreases its readability. I strongly recommend that the manuscript is corrected by a native speaker among the co-authors and thoroughly re-read.
- The discussion in Sections 3, 4 is not quantitative. The reader does not get enough information about absolute differences. This should be improved in the revised version.
- l. 466 : Can you specify what are the differences between the retrievals algorithms? I wonder why you didn't use retrievals from GOME-2 and OMI based on the same retrieval algorithm. These products are available. This should remove undesirable biases due to the different retrieval methodologies.
- All figures are based on model/data comparisons only for January, April, June, October. By doing that, we miss important information for other months, especially for July and August (maximum of biogenic emissions). The figures are also hard to read. More synthetic figures should be added, for instance showing the monthly variation of the satellite/model columns over large regions.
- Detailed comparisons with ground-based measurements are missing. The ground-based measurements shown in Figures 4-7 leave a lot to be desired. No concrete conclusion can be drawn from these plots with regards to the observed monthly variation and how well the model can reproduce it.

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Bibliography

Fu, T.-M., D. J. Jacob, P. I. Palmer, K. Chance, Y. X. Wang, B. Barletta, D. R. Blake, J. C. Stanton, and M. J. Pilling, Space-based formaldehyde measurements as constraints on volatile organic compound emissions in East and South Asia and implications for ozone, *J. Geophys. Res.* 112 (D6), doi:10.1029/2006jd007853, 2008.

Granier, C., B. Bessagnet, T. Bond et al. Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980-2010 period, *Climatic Change*, 109, 163–190, 2011.

Granier, C., T. Doumbia, L. Granier, K. Sindelarova, G. Frost, I. Bouarar, C. Lioussé, S. Darras and J. Stavrakou: Anthropogenic emissions in Asia, *Air Pollution in Eastern Asia : an integrated perspective*, eds. Bouarar, I., Wang, X., Brasseur, G., Springer international Publishing, doi:10.1007/978-3-319-59489-7-6, pp. 107-133, 2017.

Huang, G., Brook, R., Crippa, M., Janssens-Maenhout, G., Schieberle, C., Dore, C., Guizzardi, D., Muntean, M., Schaaf, E., and Friedrich, R.: Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970-2012, *Atmos. Chem. Phys.*, 17, 7683-7701, <https://doi.org/10.5194/acp-17-7683-2017>, 2017.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAPv2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, *Atmos. Chem. Phys.*, 15, 11411-11432, <https://doi.org/10.5194/acp-15-11411-2015>, 2015.

Sindelarova, K., C. Granier, I. Bouarar, A. Guenther, S. Tilmes, T. Stavrakou, J.-F. Muller, U. Kuhn, P. Stefani, and W. Knorr, Global dataset of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, *Atmos. Chem. Phys.*, 14,

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9317-9341, 2014.

Stavrakou, T., J.-F. Muller, M. Bauwens, I. De Smedt, M. Van Roozendaal, A. Guenther, M. Wild, and X. Xia, Isoprene emissions over Asia 1979-2012 : impact of climate and land use changes, *Atmos. Chem. Phys.*, 14, 4587-4605, 2014.

Stavrakou, T., J.-F. Muller, M. Bauwens, I. De Smedt, Sources and long-term trends of ozone precursors to Asian Pollution, *Air Pollution in Eastern Asia : an integrated perspective*, eds. Bouarar, I., Wang, X., Brasseur, G., Springer international Publishing, doi:10.1007/978-3-319-59489-7-8, pp. 167-189, 2017.

van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y., Rogers, B. M., Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J., Yokelson, R. J., and Kasibhatla, P. S.: Global fire emissions estimates during 1997-2016, *Earth Syst. Sci. Data*, 9, 697-720, <https://doi.org/10.5194/essd-9-697-2017>, 2017.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-1136>, 2018.

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