

Interactive comment on “Which processes drive observed variations of HCHO columns over India?” by Luke Surl et al.

Anonymous Referee #2

Received and published: 13 December 2017

This manuscript provides an in-depth and comprehensive analysis of OMI HCHO columns over India. To answer the question posed by the manuscript title, HCHO columns over India are driven largely by biogenic isoprene, with smaller enhancements from anthropogenic VOCs and pyrogenic activity. The authors use a box model analysis to analyze the potential contribution of various VOCs to observed HCHO columns. The spatial and temporal patterns of HCHO columns were compared against results from GEOS-Chem. Isoprene emissions derived from the linear approach suggest the effects of monsoons may not be well captured.

Below, I suggest a few adjustments which would improve the analysis, mostly relating to the isoprene-HCHO relationship as discussed in the manuscript. In general, the manuscript is well written and will be of high interest to readers, and I recommend

C1

publication after minor revisions.

Major comments:

1.) Verison 10-01 of the GEOS-Chem model is used in this analysis. The mechanism in v11-01 (released Feb 2017) includes large revisions on the isoprene oxidation mechanism effecting the HCHO yield. Specifically, the prompt yield of HCHO from the reaction of the isoprene peroxy radical with NO ($\text{RIO}_2 + \text{NO}$) is increased by 24% (from 0.66 to 0.82). The revised mechanism is in good agreement with fully explicit mechanisms and generally reproduces observations (Marvin et al., 2017). Because we have more confidence in the updated mechanism, and because the mechanism is likely to affect the modeled HCHO columns shown here, the updated isoprene oxidation mechanism should be used in this analysis.

2.) It is unclear here if the chemical mechanism used in the CABBA/MECA box model is the same as used in the GEOS-Chem simulation. Ideally, the same mechanisms would be used.

3.) Recently, Zhu et. al (2016) found that the OMI SAO HCHO product used here was biased low by 37% compared to observations over the Southeastern United States. Do we have any reason to suspect that this bias may persist in other regions of the globe? I understand that in-situ HCHO observations are likely limited for this region; however, acknowledgement of the potential for bias and the need for validation is warranted.

Minor comments:

Page 3, line 79: It is unclear what is meant by “this approach”.

Page 3, sentence starting at line 88: Wolfe et al. (2016) is cited as a reference for the uncertainty in HCHO production in the low NO_x regime. In contrast, I think this paper suggests that the NO_x dependency of HCHO production is well captured by updated chemical mechanisms. In any case, it is not clear to me why uncertainty in VOC emissions and in the low NO_x yield means “both approaches provide useful

C2

insights". Perhaps it is more useful to point out that while the inverse model approach gives a more rigorous result, it comes at a higher computational expensive, and the local linear approach provides a useful approximation where there is little smearing.

Page 7, line 201: List which version of MEGAN.

Page 7, line 213: GEOS-Chem HCHO columns are averaged between 1300 and 1500 to provide comparison with OMI observations. Is this a hold-over from GEOS-Chem simulations run at coarse resolution with longer (60 min) timesteps? At the 10 minute chemistry timestep often used for $0.25^\circ \times 0.3125^\circ$ simulations, I would have expected the output to be averaged over a smaller time frame (1300 to 1400). Perhaps the effect is small, but Is there a reason for the 1300 – 1500 averaging window? What are the chemistry and transport time steps used in this simulation?

Page 8, lines 240-226. The box model analysis of isoprene oxidation as a function of NOx seems redundant with other recent publications, which also have the benefit of comparison to in-situ observations (Wolfe et al., 2016; Marvin et al., 2017). For example, figure 3 is reminiscent of Figure S3 in Marvin et al. 2017. At the very least, the box model results here should be compared with those in previous literature.

Page 11, line 370: add a section reference (i.e. "as described in section 2.2")

Page 15, lines 491: The procedure of deriving a posteriori emissions using the linear approach should be briefly described for readers unfamiliar with the process.

Page 38, Table 3: Consider adding a "% change" column

Typographical errors:

In general, several subsections are titled but lack numbers (e.g., "Box modeling" in section 2.1). Sub-sub sections may be useful to the reader.

Page 8, line 253: replace "isoprene getting oxidized" with "isoprene oxidation"

Page 13, line 429: should read "collected"

C3

Page 14 line 471: should read "16-25°N and"

Page 14 line 472: should read "°E (". Also, replace "We remove scenes that correspond to" with "We remove scenes with"

Page 16 line 522: should read "emissions"

Page 23, Figure 1 caption, line 6: should read "where we infer"

Page 23, Figure 1 caption, line 7: should read "area denotes"

References: 11 Marvin, M. R., Wolfe, G. M., Salawitch, R. J., Canty, T. P., Roberts, S. J., Travis, K. R., Aikin, K. C., de Gouw, J. A., Graus, M., Hanisco, T. F., Holloway, J. S., Hübner, G., Kaiser, J., Keutsch, F. N., Peischl, J., Pollack, I. B., Roberts, J. M., Ryerson, T. B., Veres, P. R., and Warneke, C.: Impact of evolving isoprene mechanisms on simulated formaldehyde: An inter-comparison supported by in situ observations from SENEX, *Atmos. Environ.*, 164, 325-336, <http://dx.doi.org/10.1016/j.atmosenv.2017.05.049>, 2017.

Wolfe, G. M., Kaiser, J., Hanisco, T. F., Keutsch, F. N., de Gouw, J. A., Gilman, J. B., Graus, M., Hatch, C. D., Holloway, J., Horowitz, L. W., Lee, B. H., Lerner, B. M., Lopez-Hilfiker, F., Mao, J., Marvin, M. R., Peischl, J., Pollack, I. B., Roberts, J. M., Ryerson, T. B., Thornton, J. A., Veres, P. R., and Warneke, C.: Formaldehyde production from isoprene oxidation across NOx regimes, *Atmos. Chem. Phys.*, 16, 2597-2610, doi:10.5194/acp-16-2597-2016, 2016.

Zhu, L., Jacob, D. J., Kim, P. S., Fisher, J. A., Yu, K., Travis, K. R., Mickley, L. J., Yantosca, R. M., Sulprizio, M. P., De Smedt, I., Abad, G. G., Chance, K., Li, C., Ferrare, R., Fried, A., Hair, J. W., Hanisco, T. F., Richter, D., Scarino, A. J., Walega, J., Weibring, P., and Wolfe, G. M.: Observing atmospheric formaldehyde (HCHO) from space: validation and intercomparison of six retrievals from four satellites (OMI, GOME2A, GOME2B, OMPS) with SEAC(4)RS aircraft observations over the southeast US, *Atmos. Chem. Phys.*, 16, 13477-13490, doi:10.5194/acp-16-13477-2016, 2016.

C4

