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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Lower tropospheric ozone over India and its linkage to the South Asian monsoon" *by* Xiao Lu et al.

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Comment: The manuscript by Lu et al. investigated the lower tropospheric ozone over India and its linkage to the south Asian Monsoon by using satellite observations and GEOS-Chem model. Spatial and temporal characteristics of lower tropospheric ozone over India were analyzed in terms of seasonal cycle and inter-annual variability (for 2006-2010) and also long term-trends (for 1990-2010). The contribution/roles of different processes, including precursor emissions (anthropogenic NOx, biomass burning etc), meteorology (horizontal and vertical transport), chemical production, and dry deposition were discussed. The linkage of lower tropospheric ozone concentrations over India to the south Asian Monsoon were also quantified.

This comprehensive analysis focus on the ozone over Indian region where the precursor emissions are still rising in recent years. This study is thorough and clear, which would help to understand the local and global environment effects of India ozone. Overall, this manuscript is well organized, states the problem, outlines the model experiments, and describes the model results. This study fits the scope of ACP. I recommend publication.

Below are several comments that I think the authors may address to improve the manuscript.
Response: We thank the reviewer for the valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment: General comments:
1. Line 162-163. The authors reduced the 1990-1996 ACCMIP emissions by 30% to correct the gap between GFED3 and ACCMIP, and also to get a full set of data for 1990-2010. It's quite understandable and straightforward. While the readers might get the impression that GFED3 is more accurate than ACCMIP. I'm just curious is there any reference that evaluate the two biomass burning emission inventories for this region? Any discussions of the sensitivity of the biomass burning emissions would be helpful.
Response: Thanks for pointing it out; we do not have enough evidence from the literature to conclude which biomass burning emission inventory is more accurate over India. We now state in the text "Comparison of GFED3 and ACCMIP biomass burning CO emissions for their overlapping years (1997–2000) suggests ACCMIP is 30% higher. Here we reduce the 1990–1996 ACCMIP emissions by 30% to reconcile the two inventories, although this may lead to underestimates of biomass burning emission contributions for the period. We find that biomass burning emissions of CO over India (2.6 Tg a-1 (per annum) for 2006–2010) are relatively small compared with anthropogenic emissions (61.9 Tg a-1)".

Comment: 2. Line 177. The contributions of different processes are analyzed, including chemical production and loss, horizontal and vertical transport and dry deposition. I'm wondering if the cloud-chemistry/wet scavenging are considered as the two processes might involve the O3 chemistry. Or are they already considered but just ignored as the contribution are too small? Some discussion would be helpful.
Response: Cloud chemistry and wet deposition are included in the GEOS-Chem model, but they do not affect ozone concentrations directly. We now state here in the Section 2.3: "The GEOS-Chem model also include cloud chemistry (e.g., formation of sulfate aerosol via aqueous-phase reactions with ozone and H2O2) and wet deposition of soluble gases. The two processes have small effects on ozone directly due to its low solubility and thus are not diagnosed here".

Comment: 3. Line 181. For the horizontal transport (E-W, N-S) in the text and also figure 2, it would be better to give the directions for positive/negative values.
Response: We now state here in the text "Horizontal transport for each grid is calculated by horizontal fluxes from or to adjacent grids. Here we define transport from west to east or from south to north as positive values", and in the caption of Figure 2 "Horizontal transport from west (W) to east (E) and from south (S) to north (N), and downward (D) vertical transport at 600 hPa are defined as positive values".

Comment: 4. Section 3.1. The anthropogenic NOx emissions and biomass burning emissions are shown in this section while no discussions of NMVOC emissions are provided. While anthropogenic and biogenic NMVOC are also important ozone precursors. Especially for biogenic NMVOC, it might have strong seasonal cycle which might change the NOx/VOC regime in different seasons. Some discussions would be necessary here.
Response: We now show in Figure S1 the seasonal variations of anthropogenic NMVOC emissions, biogenic isoprene emissions, and soil NO emissions. This is also for addressing Comment 6 below. We now state in the text "Anthropogenic CO and

NMVOC emissions over India are 61.89 Tg a-1 and 15.5 Tg a-1, respectively, with similar seasonal variations as anthropogenic NO emissions (Figure S1)".

We also discuss in this section biogenic isoprene emissions and NOx/VOC regimes, "Model calculated biogenic isoprene emissions in India are 39.8 Tg C a-1, with a strong seasonality peaking in May and June (Figure S1). Previous studies have shown that the ratio of NOx emissions to CO and NMVOCs emissions over India is relatively small compared to other regions at northern mid-latitudes (Lelieveld et al., 2001; Li et al., 2017). Here we also examine the model simulated H2O2/HNO3 concentration ratios, which have been used as an indicator of ozone production chemical regime (Sillman 1997; Zhang et al., 2016). We find that the H2O2/HNO3 ratios in the Indian lower troposphere range from 1.0 to 5.0 for all four seasons, higher than those in eastern China and eastern US (Figure S2). This indicates strong NOx-limited conditions for ozone chemical production over India, consistent with previous studies (Kumar et al., 2012; Sharma et al., 2016)".

Added references:
Lelieveld, J., Crutzen, P. J., Ramanathan, V., Andreae, M. O., Brenninkmeijer, C. M., Campos, T., Cass, G. R., Dickerson, R. R., Fischer, H., de Gouw, J. A., Hansel, A., Jefferson, A., Kley, D., de Laat, A. T., Lal, S., Lawrence, M. G., Lobert, J. M., Mayol-Bracero, O. L., Mitra, A. P., Novakov, T., Oltmans, S. J., Prather, K. A., Reiner, T., Rodhe, H., Scheeren, H. A., Sikka, D., and Williams, J.: The Indian Ocean experiment: widespread air pollution from South and Southeast Asia, Science, 291, 1031-1036, 10.1126/science.1057103, 2001.
Li, M., Zhang, Q., Kurokawa, J.-I., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 935-963, 10.5194/acp-17-935-2017, 2017.

Sillman, S., He, D., Cardelino, C., and Imhoff, R. E.: The Use of Photochemical Indicators to Evaluate Ozone-NOx-Hydrocarbon Sensitivity: Case Studies from Atlanta, New York, and Los Angeles, J Air Waste Manag Assoc, 47, 1030-1040, 10.1080/10962247.1997.11877500, 1997.

Zhang, Y., Cooper, O. R., Gaudel, A., Thompson, A. M., Nédélec, P., Ogino, S.-Y., and West, J. J.: Tropospheric ozone change from 1980 to 2010 dominated by equatorward redistribution of emissions, Nature Geosci., 9, 875-879, 10.1038/ngeo2827, 2016.

Comment: 5. Line 213. When retrieving the OMI observed tropospheric ozone, some discussions of the sensitivity of the priori profile and average kernel matrices would be helpful. Are there any system biases or different biases in different seasons? Any uncertainties?

Response: Thanks for the suggestion. We now state in Section 2.1 (OMI satellite observations) “The degrees of freedom for signals (sum of the diagonal elements of averaging kernel matrices) for OMI ozone retrievals are typically 0.3–0.5 in the lower troposphere over India. Previous evaluations of the OMI retrievals with ozonesonde measurements have shown a clear improvement over the a priori in the lower troposphere of the tropics (30°S-30°N), and the mean retrieval biases in the tropics are less than 6% with little seasonality (Huang et al., 2017a)”.

Comment: 6. Line 242. It's quite important to state that biogenic isoprene and soil NOx emissions are higher in the pre-summer monsoon than wintertime which help the reader to understand the results. As suggested in Q4, it will be great to give a table that lists the seasonal/annual emissions from different sources (biogenic VOC, anthropogenic VOC, soil NOx, biomass burning CO etc.)

Response: Thanks for the suggestion. We now add Figure S1 in the Supplement to show seasonal variations of anthropogenic NMVOCs, biogenic isoprene, and soil NO emissions, as described in Comment 4 above. We state in the text: “Biogenic isoprene emissions over India increases from 1.8 Tg month-1 in winter to 5.2 Tg C month-1 in

the pre-summer monsoon season. The soil NO emissions also increase from 0.08 Tg month-1 to 0.21 Tg month-1 (Figure S1).”

Comment: 7. Line 285. “Strong vertical convection”. Just curious, is there any index to show the strength the convection?

Response: We now state in the text “Strong vertical convection with the 600 hPa upward velocity greater than 5 mm s-1 in July–August effectively uplifts ozone pollution from the lower troposphere to the upper troposphere”.

Comment: 8. Line 340-342. In addition of NOx emission reduction, how about the changes of reaction rates and biogenic VOC emission caused by T reduction during presummer monsoon? And their roles in the O3 production?

Response: We have discussed in Section 3.2 (Variations in the pre-summer monsoon season) the influences of changes in temperature on ozone concentration through changing natural emissions and chemical production in the pre-summer monsoon. This sentence here was intended to emphasize the benefits of controlling NOx emissions. We now remove this sentence to avoid confusion.

Comment: 9. Line 375. Can you show the selected region in one of your figures?

Response: We now show the selected region for calculating the South Asian summer monsoon index in Figure S2. We state in the text “we then average δ(i, j) over the region of 35°E–90°E, 5°N–35°N (Figure S2) at 850 hPa and over May–August to represent the South Asian summer monsoon index (SASMI).”

Comment: 10. Line 384. How the statistics are conducted? How many samples are compared? Are the correlations for grid-to-grid? Or just the regional averages?

Response: We now state here “Interannual variations of Indian regional mean lower tropospheric ozone concentrations are significantly negative correlated with the SASMI, as can be seen for both OMI observations (r = −0.46, 2006–2014, n = 9) and GEOS-Chem BASE results (r = −0.52, 1990–2010, n = 21).”

Comment: 11. Figure 7. The caption is not clear. Do you mean ” Differences in May-August mean between the lowest and highest SASMI conditions ”?
Response: Yes, we now clarify in the caption “Differences in May–August monthly mean (a) lower tropospheric ozone concentration, (b) . . . between the lowest and highest SASMI conditions. Values are calculated using averages of the five lowest SASMI years minus averages of the five highest SASMI years”.

Comment: 12. Figure 3. Dry deposition is not shown here. As there is no obvious season changes? A little bit explanation would be helpful.
Response: We now present in Figure S1-d the spatial distribution and seasonal variation of ozone dry deposition to India. We also state in the text “Dry deposition of ozone to India shows a weak seasonal variation (1.5 $\pm$ 0.15 Tg month-1; Figure 2c and Figure S1-d).”

Comment: 13. Figure 4. Can you give the number pairs of data?
Response: We now state in the figure caption “Interannual correlation coefficients (r) between surface temperature and ozone (number of regional averages n = 9 for observations and 21 for model results) are shown inset”.

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