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Interactive comment

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 study of Lu et al. analyzes the processes influencing lower tropospheric ozone over the Indian subcontinent with a focus on the influence of the South Asian monsoon but also analyzing interannual variability and trends. It mainly relies on a set of 20-year (1990-2010) long GEOS-CHEM simulations and analyzing the lower tropospheric ozone budget with respect to contributions from photochemistry, transport and deposition. The model-derived results are backed up with lower tropospheric ozone data derived from OMI satellite observations of the years 2006 - 2014 which, overall, show very good agreement with the model in terms of spatial distribution and seasonal behavior.

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Discussion paper

The publication is very well written (with small grammatical issues that will require some copy-editing), clearly structured, and the analyses are comprehensive and convincing. In particular, the authors present a thorough analysis of the factors driving ozone variability over India including meteorological variability (seasonal, interannual), variations in anthropogenic and biomass burning emissions, and trends in methane. The paper has a good chance to become an important reference for future studies on the lower tropospheric ozone budget over India.

I support publication of this manuscript and have only a few small comments that may require minor revisions:
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: - As pointed out by Lelieveld et al. (The Indian Ocean Experiment: Widespread Air Pollution from South and Southeast Asia, Science, 2001), the ratio of emissions of NOx to those of CO and VOCs is much smaller over India than e.g. over Europe or the United States. As a consequence ozone production over India is likely strongly NOx-limited. It would have been nice if the authors had paid somewhat more attention to this aspect, notably in the analysis of the factors driving the long-term increase in ozone.
Response: Thanks for the suggestion. We now discuss the ratio of H2O2/HNO3 as an indicator of ozone chemical production regime as presented in Figure S2. We state in Section 3.1 (Variations of meteorology and emissions): "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)”.

We also state in Section 5 (Long term trend), “Increases in anthropogenic NO emissions (about 3% year-1, Supplemental Figure S5) likely dominate the ozone increases due to the NOx-limited ozone production condition over this region as we discussed above”.

Added references:

Kumar, R., Naja, M., Pfister, G. G., Barth, M. C., Wiedinmyer, C., and Brasseur, G. P.: Simulations over South Asia using the Weather Research and Forecasting model with Chemistry (WRF-Chem): chemistry evaluation and initial results, Geoscientific Model Development, 5, 619-648, 10.5194/gmd-5-619-2012, 2012.
Sharma, S., Chatani, S., Mahtta, R., Goel, A., and Kumar, A.: Sensitivity analysis of ground level ozone in India using WRF-CMAQ models, Atmos. Environ., 131, 29-40, 10.1016/j.atmosenv.2016.01.036, 2016.
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: - Figure 2c shows the seasonal cycle of lower tropospheric ozone together with the different monthly production and loss terms. Why do these terms not sum up to positive values when the total burden of ozone increases? Take e.g. the budget for the month of March: The losses sum up to about -12 Tg, the gains only to about +11 Tg. Despite this negative budget, the ozone concentrations increase. from

March to April. Please clarify.
Response: Thanks for pointing it out. We find an error in calculation of vertical ozone fluxes for January to March. It is now corrected in the figure, and does not affect our conclusions.

Comment: Page 4, line 86: decreases in ozone associated with the summer monsoon had also been reported based on MOZAIC aircraft observations, e.g. Srivastava et al. (Atmos. Env., 2015) or Bhattacharjee et al. (Meteorol. and Atmos. Phys., 2015).
Response: These studies are now cited in the manuscript. We state here "Decreases of tropospheric ozone with the summer monsoon in South and East Asia have been reported from . . . aircrafts measurements (Bhattacharjee et al., 2015; Srivastava et al., 2015; Ojha et al., 2016) . . .".

Added references:
Bhattacharjee, P. S., Singh, R. P., and Nédélec, P.: Vertical profiles of carbon monoxide and ozone from MOZAIC aircraft over Delhi, India during 2003–2005, Meteorology and Atmospheric Physics, 127, 229-240, 10.1007/s00703-014-0349-x, 2014.
Srivastava, S., Naja, M., and Thouret, V.: Influences of regional pollution and long range transport over Hyderabad using ozone data from MOZAIC, Atmos. Environ., 117, 135-146, 10.1016/j.atmosenv.2015.06.037, 2015.
Ojha, N., Pozzer, A., Rauthe-Schöch, A., Baker, A. K., Yoon, J., Brenninkmeijer, C. A. M., and Lelieveld, J.: Ozone and carbon monoxide over India during the summer monsoon: regional emissions and transport, Atmos. Chem. Phys., 16, 3013-3032, 10.5194/acp-16-3013-2016, 2016.

Comment: - It was not always entirely clear, which regions the individual budgets and concentrations were representing, i.e., over which domain the values were averaged. "Lower troposphere over India" is not an exact term. Is this only over land,

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Discussion paper

or over a rectangular domain covering India? I suggest adding such information to the figure captions.
Response: We now state in the text “In this study, we present an integrated analysis of the processes controlling lower tropospheric (surface to 600 hPa) ozone concentrations over the terrestrial land of India and their linkage to the South Asian monsoon.” We have also clarified it in the figure captions.

Comment: - The same problem holds for some of the stated correlations, e.g. the correlation of r = 0.81-0.97 mentioned on page 10, line 223. What exactly was correlated here?
Response: Thanks for pointing it out. We now state here “they generally capture the seasonal and spatial variations of OMI observed lower tropospheric ozone concentrations (r = 0.81–0.97 for the spatial variations of OMI observations vs. model results).” We have also clarified some other correlations in the text and figure captions.

Comment: - Page 15, line 343: The statement "the mean correlation over India is still dominated by the temperature impact on ozone chemical production" is not quite correct. I would argue that high temperatures and high ozone levels are both a result of intense solar radiation, rather than temperature being directly the main driver for high ozone production.
Response: We now clarify in the text: “the mean correlation over India still shows a positive effect of temperature on ozone chemical production, which can be driven by solar radiation affecting both temperature and ozone production rates, as well as the sensitivities of natural sources to temperature as discussed above.”

Comment: - Page 16, Eq. 1: The grid indices i,j should probably be discarded, since equation 2 suggests that the norm is calculated by integration over a domain rather than a single grid cell.
Response: We rewrite some of the sentences here to avoid confusion. The monsoon

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Discussion paper

index is first calculated for each grid and then averaged over the South Asian region. We now state here “The monsoon index is first calculated for each model grid (δ(i, j)) in the Northern Hemisphere in the month m and year y” and “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: Probably the manuscript would also benefit from a schematic figure of the ozone budget in the pre-monsoon and monsoon seasons, i.e., a box representing the volume of the lower troposphere over India and the individual budget terms shown as arrows (in case of transport and deposition) or just numbers (in case of chemical production and loss within the box).
Response: Thanks for the suggestion. We now present such a schematic figure in the main manuscript as Figure 4. We state in Section 3.3 (Variations in the summer monsoon season): “Figure 4 summarizes changes in the lower tropospheric ozone budgets over India in the pre-summer monsoon season (March-April) and the summer monsoon season (June-July-August). We can see that decreases in the Indian lower tropospheric ozone in the summer monsoon season are mainly associated with the reduction in ozone net chemical production and strengthening upward transport.”

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