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Interactive comment

Interactive comment on "Evaluation of nitrogen oxides sources and sinks and ozone production in Colombia and surrounding areas" *by* Johannes G. M. Barten et al.

Anonymous Referee #2

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In their paper, Barten et al. provide an evaluation of atmospheric chemistry over Colombia using WRF-Chem modelling evaluated with satellite and surface observations. The paper is very well written, contains a good and balanced set of references, has an appropriate length and amount of detail and I could not discover any obvious flaws. Colombia is a complex and interesting country with isolated regions and various climate zones where the dominant emissions may vary from anthropogenic, biomass burning to lightning. The approach is of wider interest, because similar studies may be conducted for other countries with limited air-quality monitoring networks. Figure 4-b is a central figure in the paper and starting point to discuss the different regions in more detail.

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Discussion paper

I am in favour of publishing this work, but I have four major general comments, provided below, which will require a major revision.

General comments:

1. The authors present only one month of simulations (January 2014) but the seasonal dependence of NOx and ozone is not discussed. There are good reasons to focus on January because it is the dry season, but the country experiences wet and dry seasons where the relative importance of sources of NO may change. The diurnal variability as well as the multi-annual variability in the satellite data are discussed, but the seasonality is missing. Would it be possible to extend the simulations to a couple of months to sample the yearly cycle in emissions? It would be interesting to present also the seasonality of the satellite (and surface) observations.

2. The authors show that the lightning source is the dominant source for 63% of the grid cells and is also the largest source in terms of total amount. Therefore lightning is a key aspect for Colombia, and, given also the major uncertainty in the modelling of this process, deserves special attention. The uncertainty in the lightning source is e.g. demonstrated by the adjustments made to the default settings of WRFchem, which scaled down lightning by a factor 20. However, the authors (if I understood correctly) have used only the clear-sky observations of OMI. It has been shown in several publications that lightning source estimates may be derived using the observations over high clouds. Because the resolution of WRF is comparable to OMI, it would be interesting to include a comparison between these cloud covered observations and WRF-Chem, to test the capability of the model to describe major thunderstorms. See for instance: Beirle, S., Huntrieser, H., and Wagner, T.: Direct satellite observation of lightning-produced NOx, Atmospheric Chemistry and Physics, 10, 10 965–10 986, https://doi.org/10.5194/acp-10-10965-2010, 2010. Pickering, K. E., Bucsela, E., Allen, D., Ring, A., Holzworth, R., and Krotkov, N.: Estimates of lightning NOx production based on OMI NO2 observations over the Gulf of Mexico, Journal of Geophysical Research: Atmospheres, 121, 8668–8691, https://doi.org/10.1002/2015JD024179, 2016.

3. The discussion of soil biogenic emissions is very limited. The regions where these emissions are dominant are identified. Therefore the comparison with OMI could be extended: are there indications that soil emissions are under (over) estimated? Where and by what amount?

4. The differences between the single-column model and WRF-Chem should be more clearly described. Why does WRF-Chem produce so much lower concentrations in the city?

Detailed comments:

Abstract, l15: “averaged difference of 0.02Âů10ˆ15". How significant is this number?

l19: "WRF-Chem was unable to capture NOx and CO urban". For which cities?

l80: "air quality in Colombia concerns are generally" Replace by "concerns about air quality in Colombia are generally"

l101: The spin-up time is very short? How is the model initialised?

l103: "in Appendix A we show how the selected study period can be deemed being representative for the baseline state of air quality in Colombia. " This claim is not very clear to me. Figure A1 shows the year-to-year mean variation in January. Why is it representative? I would like to see also a seasonal variation, e.g. linked to the wet and dry seasons. Only showing results for January is a weak point of the paper that should be better motivated. Also, a seasonal mean could be more representative because of the limited number of OMI pixels in a month, see Fig.2.

l107: " ... (ECMWF) .. meteorological boundary conditions." Is this the operational dataset or reanalysis?

l158: "data filtering recommendations by the QA4ECV": What is the filtering criterion for clouds? I assume cloud-covered pixels are not used?

l161: "limiting the quality of and which increases the uncertainty". Please reformulate.

l184: "mostly decreases in AMF". Please replace by "increases in the VCD" because not all readers will be familiar with the AMF concept. This remark applies to the whole section: please refer to VCD only.

l190: "This reflects a higher abundance of NO2 higher up in the troposphere". It is not fully clear to me how this can be concluded. The whole profile will be important. Are lightning emissions modelled higher in the atmosphere in WRF compared to the TM5-MP? Was this checked? Also, cloud-covered observations are removed from the OMI dataset by the filtering?!

l196: "In this research, we focus on tropospheric NO2 columns." This line can be removed.

Sec. 3.2 in-situ data: please provide information about the instruments used. Is the data publicly available?

Fig. 4a: Unit? Is this per grid cell, per square km, or something else?

Fig. 4b, showing a map of the dominant source, is a very useful plot!

l232-237: The numbers in the table are repeated in the text. This paragraph may be shortened therefore. I would suggest to add the % of land where the source is dominant (as given in the text) as extra column in the table.

l241: "very low VCD over Carribbean". What is the influence of the (free troposphere) boundary conditions?

l248: "northeastern part". Please provide a more detailed explanation where these high values come from.

l257: "Even though the overestimation is small in absolute terms". Could a possible bias in the satellite observations contribute to the difference observed?

l261: "This further confirms the finding that lightning NOx emissions are overestimated". Maybe this conclusion should be weakened. As explained by the authors in l262-265, the comparison with OMI is only performed in OMI clear-sky conditions. Therefore the comparison may be biased.

l290: "These results confirm the application of the recalculated OMI data." It should be noted that the differences in mean, median, 90% confidence interval is not very large. So it is questionable if the confirmation is significant.

p14: I was wondering if the authors have analysed the precise location of the surface stations in Bogota? Closeness to major sources/roads could perhaps explain the difference with WRF-Chem?

l341: "nudging the concentrations of long-lived tracers such as O3, NOx and CO above the boundary layer using the CAMS data". Why not use the WRF-Chem data for this?

l342: "used the same emissions, including diurnal cycle, as in the WRF-Chem simulation." I find it conceptually difficult to understand why the SCM works better than WRF-Chem with such similar inputs. From the previous section I understood that the local (traffic) emissions at the surface stations are underestimated. But the SCM apparently uses the same emissions as WRF-Chem. Why are concentrations of NOx/CO so much higher in the SCM than in WRF-Chem? Which aspect of the SCM is responsible for this difference?

l367: "VCD analysis for January 2014 is representative for the NOx emissions for the larger study domain". I find this not well justified. The A1 plot only shows January. In particular the seasonal variability would be of interest while the study is limited to one year and January only.

l389: "still overestimates NOx emissions". Please explain why? The domain studied is a very active lightning region. What fraction of the global lightning total is expected to come from this area? E.g. the comparison with Miyazaki is quite close.

l400: "urband"

l415: "which explains part of the overestimation by WRF-Chem". It should be mentioned that there is a strong variability from year to year, most likely related to other reasons than changes in emissions which are expected to result in more gradual changes. See next line.

l423: "we found that WRF-Chem does not systematically underestimate urban VCDs". What is this statement based on? Do the authors refer to studies over other regions with the same setup?

l434-436:"overestimations of VCDs ... in contrast with ... there is an overestimation " ? Please reformulate.

l445; l457 : "indicating that all surface monitoring stations are located at or near busy roads", "EDGAR emissions integrated in a relatively simple Single Column Model, can represent the averaged diurnal cycles of O3, CO and NOx reasonably well.". This is confusing and should be explained more clearly. The SCM concentrations also match quite well in absolute amount the surface observations, which seems to suggest that EDGAR is OK and that the surface network is representative for larger areas?!

There is a repetition of concluding remarks when comparing the discussion and conclusion sections. This repetition should be removed. Maybe the two sections can be merged (and thereby shortened a bit)?

The close agreement of the mean/median is a bit over-emphasised to my taste. In the different source sectors there are major differences with compensating effects on the mean or total.

l489: "showed that the selected simulation period is representative for the baseline state of air quality in Colombia". Is this a statement for January only?

l497: "evaluate the impact of a modified representation of emissions based on the observed to WRF-Chem simulated CO mixing ratio". The description of the SCM, section 5, says: "and used the same emissions, including diurnal cycle, as in the WRF-Chem simulation". This is confusing. Please explain more clearly the setup of the

SCM and explain why e.g. the CO/NOx concentrations have increased substantially compared to WRF-Chem.

l507: "It may provide as a base for more local studies or the application towards future predictions". Please re-write.

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