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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of a regional air quality model using satellite column NO_2: treatment of observation errors and model boundary conditions and emissions" *by* R. Pope et al.

Anonymous Referee #1

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In this manuscript, authors tried to evaluate the performance of a regional air quality model of AQUM using tropospheric column NO2 from satellite observations over north-west Europe, especially over UK. Also, authors suggested an algorithm to reduce the retrieval errors via averaging satellite data and using AKs. The manuscript looks solid and the description is straightforward. However, several points should be addressed properly in the manuscript to be accepted for the final publication in ACP. Here are some general and specific comments for further considerations.

General comments

1. Emissions

There are three different types of uncertainties originated from three major components: (i) the atmospheric physical/chemical processes in the CTM; (ii) satellite retrieval; and (iii) emission fluxes. In this study, the manuscript covered two aspects of the uncertainties (i.e., satellite and CTM-originated uncertainties). However, how about emission? It is required to describe how the emission fluxes used are accurate (or uncertain).

1-1. Biogenic emission: Biogenic species (i.e., isoprene, mono-terpene, etc) play an important role even in NOx chemistry by controlling OH radical concentrations. For example, if isoprene emissions are overestimated, the estimated levels of NO2 can be higher than the actual levels of NO2 (because of slow NOx losses). The tropospheric chemistry can, sometimes significantly, influence the NOx analyses for summer episode. Thus, authors should clarify which biogenic inventory (e.g., MEGAN, GEIA, etc) was used in your study and explain how the biogenic inventory is uncertain (or reliable).

1-2. Biomass burning emission: I wonder whether biomass burning emission was considered in the CTM simulations. If it was considered, authors need to mention/describe it.

1-3. Variations of NOx emissions: This issue can be an important factor in the analysis of seasonal trends of columnar NO2. Authors should provide the seasonal (or monthly) information of NOx emission. Also, this information can be useful to analyze the seasonal trends of columnar NO2 over the London and northern England regions in Fig. 3. Higher columnar NO2 over London and northern England from OMI observations are well captured by 3D-CTM simulation. However, usually, both NO2 columns from the CTM and satellite observations during winter are higher than those during summer (e.g., van Noije et al., 2006; Huijnen et al., 2010). Authors should explain the unusual

trends of the NO2 columns (higher NO2 columns during summer) over the regions shown in Fig. 3.

2. Sensitivity experiments

2-1. Chemical LBCs: In the manuscript, authors mentioned that using chemical GEMS gives better result and it is consistent with the findings of Savage et al. (2013). However, authors do not mention what makes it better. What are the main differences between the GEMS and MACC LBCs? If the MACC is an improved version of the GEMS, what has been improved? Give some more detailed information on both the GEMS and MACC.

2-2. E2 (idealized point source tracer): In the E2 sensitivity test, authors tried to examine the spatial patterns of the tracer. The experiment remains unclear in the manuscript. i) How to idealize tracer from the point sources? ii) Which species are used as a tracer? It appears to be a "reactive species" having one-day lifetime. iii) What are the reasons to determine a tracer having one-day lifetime? The lifetime of NOx is less than 1 day (say, several hours) during summer (Schaub et al., 2007; Lamsal et al., 2010). iv) In Fig. 7-c, the columnar NO2 over other regions seems to be "zero". Did you consider only tracer's emission for this sensitivity simulation? If yes, what is the point of considering one trace species specially having one day lifetime? If no, was the tracer tagged for identification and how to tag the tracer in your model?

2-3. Aerosol surface area (A) in the Schwartz formula (Eq. 11): As mentioned in your manuscript, aerosol surface areas can be changed by hydroscopic growth of aerosols, depending on aerosol types and relative humidity. Also, there are other atmospheric processes such as coagulation and condensation/evaporation, related to the aerosol surface areas. How are those processes treated in the AQUM model?

2-4. Reaction probability of N2O5 (γN2O5): As mentioned in the manuscript, usually, the reaction probability of N2O5 is known to range from 0.02 to 0.001 (Riemer et al., 2003; Brown et al., 2006). It may exist somewhere between the two values. What are

the reasons to choose only two extreme cases in the sensitivity simulations?

3. Satellite error

When the black polygonal regions are determined in Fig. 4, authors used the two values of MB and satellite error. Here, the satellite error is the criteria (i.e., the magnitude of the MB greater than the satellite error). In that case, how did you consider the satellite error (i.e., averaged value over domain or each pixel value)? For better understanding this analysis, authors should provide some ranges of satellite errors over specific regions (e.g. northern England + London, Benelux, Po valley, North Sea, etc).

4. Geographical information

Where is Dartmoor located in England? For the sake of reader's convenience, you would better provide some geographical information (e.g., Dartmoor, Irish Sea, North Sea, Po Valley, and many regions mentioned in the manuscript), possibly in Fig. 2.

Specific comments

1. Figs. 3 and 4

Merging two figures 3 and 4 in a 2$\times$3 panel

2. P. 21763, line 23 and P. 21764, line 4 (i.e., "around 0 - 3$\times$1015" and "between 0 - 6$\times$1015")

The minimum background columnar NO2 over background could not be "zero". Authors should provide approximate values.

3. P. 21767, line 26-27

It is a quite interesting that by introducing N2O5 heterogeneous chemistry, these positive biases were "significantly" reduced even during the summer episode, indicating that columnar NO2 are significantly decreased. N2O5 is thermally unstable and decomposes to NO3 and NO2 at high temperatures. In other words, during summer,

the decomposition of N2O5 is more active than the formation of N2O5. It would not have a significant impact on the columnar NO2 during summer. Thus, you would better provide other reasons in this part.

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