

Interactive
Comment

Interactive comment on “Evaluation of a regional air quality model using satellite column NO₂: treatment of observation errors and model boundary conditions and emissions” by R. Pope et al.

Anonymous Referee #1

Received and published: 22 September 2014

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.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



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 NO_x 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 NO_x losses). The tropospheric chemistry can, sometimes significantly, influence the NO_x 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 NO_x 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 NO_x 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

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



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 NO_x is less than 1 day (say, several hours) during summer (Schaub et al., 2007; Lam-sal 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 hygroscopic 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 (γ N₂O₅): 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

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



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 2×3 panel

2. P. 21763, line 23 and P. 21764, line 4 (i.e., “around $0 - 3 \times 10^{15}$ ” and “between $0 - 6 \times 10^{15}$ ”)

The minimum background columnar NO_2 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 N_2O_5 heterogeneous chemistry, these positive biases were “significantly” reduced even during the summer episode, indicating that columnar NO_2 are significantly decreased. N_2O_5 is thermally unstable and decomposes to NO_3 and NO_2 at high temperatures. In other words, during summer,

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

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.

REFERENCES

Huijnen, V., Eskes, H. J., Poupkou, A., Elbern, H., Boersma, K. F., Foret, G., Sofiev, M., Valdebenito, A., Flemming, J., Stein, O., Gross, A., Robertson, L., D'Isidoro, M., Kioutsioukis, I., Friese, E., Amstrup, B., Bergstrom, R., Strunk, A., Vira, J., Zyryanov, D., Maurizi, A., Melas, D., Peuch, V.-H., and Zerefos, C.: Comparison of OMI NO₂ tropospheric columns with an ensemble of global and European regional air quality models, *Atmos. Chem. Phys.*, 10, 3273-3296, doi:10.5194/acp-10-3273-2010, 2010.

Brown, S. S., Ryerson, T. B., Wollny, A. G., Brock, C. A., Peltier, R., Sullivan, A. P., Weber, R. J., Dube, W. P., Trainer, M., Meagher, J. F., Fehsenfeld, F. C., and Ravishankara, A. R.: Variability in nocturnal nitrogen oxide processing and its role in regional air quality, *Science*, 311, 67–70, 2006.

Lamsal, L. N., Martin, R. V., van Donkelaar, A., Celarier, E. A., Bucsela, E. J., Boersma, K. F., Dirksen, R., Luo, C., and Wang, Y.: Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, *J. Geophys. Res.*, 115, D05302, doi:10.1029/2009JD013351, 2010.

Riemer, N., Vogel, H., Vogel, B., Schell, B., Ackermann, I., Kessler, C., and Hass, H.: Impact of the heterogeneous hydrolysis of N₂O₅ on chemistry and nitrate aerosol formation in the lower troposphere under photochemical conditions, *J. Geophys. Res.*, 108(D4), 4144, doi:10.1029/2002JD002436, 2003.

Schaub, D., Brunner, D., Boersma, K. F., Keller, J., Folini, D., Buchmann, B., Berresheim, H., and Staehelin, J.: SCIAMACHY tropospheric NO₂ over Switzerland: estimates of NO_x lifetimes and impact of the complex Alpine topography on the re-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



trieval, Atmos. Chem. Phys., 7, 5971-5987, doi:10.5194/acp-7-5971-2007, 2007.

van Noije, T. P. C., Eskes, H. J., Dentener, F. J., Stevenson, D. S., Ellingsen, K., Schultz, M. G., Wild, O., Amann, M., Atherton, C. S., Bergmann, D. J., Bey, I., Boersma, K. F., Butler, T., Cofala, J., Drevet, J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J.-F., Lawrence, M. G., Martin, R. V., Montanaro, V., Müller, J.-F., Pitari, G., Prather, M. J., Pyle, J. A., Richter, A., Rodriguez, J. M., Savage, N. H., Strahan, S. E., Sudo, K., Szopa, S., and van Roozendaal, M.: Multi-model ensemble simulations of tropospheric NO₂ compared with GOME retrievals for the year 2000, Atmos. Chem. Phys., 6, 2943-2979, doi:10.5194/acp-6-2943-2006, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 21749, 2014.

ACPD

14, C7310–C7315, 2014

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

C7315

