Atmos. Chem. Phys. Discuss., 13, C4876–C4881, 2013
www.atmos-chem-phys-discuss.net/13/C4876/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "An evaluation of the CMAQ reproducibility of satellite tropospheric NO_2 column observations at different local times over East Asia" *by* H. Irie et al.

Anonymous Referee #1

Received and published: 17 July 2013

This manuscript by Irie et al. provides information on the reproducibility of multi-satellite observed NO2 columns from several sensitivity simulations. Also, authors discussed the diurnal pattern of NO2 columns and suggested some possible reasons for discrepancies between the model-calculated and satellite-observed data over several regions.

Here are some questions and comments for consideration possibly leading to modification.

First of all, for the study, authors chose two different seasonal episodes (i.e. summer and winter) to investigate the diurnal patterns of NO2 columns for the time span cov-

ered within those two episodes. Unlike the analysis for summer, the analysis for winter seems to remain incomplete. Therefore, this manuscript should clearly finalize this part and discuss features differing between seasonal episodes. Correspondingly, authors should provide possible reasons for the discrepancies of the R values (shown in Table 3) between the model-calculated and satellite-observed data for winter.

Secondly, there are some important issues influencing the levels of NO2 and/or chemical NOx sink in the atmosphere (Lin et al., 2012; Stavrakou et al., 2013). These include: i) NO2+OH reaction (Mollner et al., 2010; Sander et al., 2011; Henderseon et al., 2012); ii) NO+HO2 reaction (Butkovskaya et al., 2005, 2009); iii) the uptake rate of N2O5 (Riemer et al., 2003; Evan and Jacob, 2005; Brown et al., 2006; Davis et al., 2008); v) the uptake rate of HO2; and vi) OH recycling (Lelieveld et al., 2008; Butler et al., 2008). In addition to the uncertain NOx emissions, uncertainty in these chemical NOx sink could cause large discrepancies between the model and satellite-derived NO2 columns. Without the additional analysis for the sensitivity simulations, authors cannot say that the disagreement between two NO2 columns during winter cannot be explained by your sensitivity simulations.

Thirdly, anthropogenic, biogenic and pyrogenic emissions used in your CMAQ simulations could be uncertain in East Asia. How then did authors evaluate the model performances? Validation of the model results is required in your manuscripts using the in-situ measurements (EANET data may be available during episodes, http://www.eanet.asia/). Regarding the first comment, the NO2 diurnal pattern (and/or the ratio of NO2, afternoon to NO2, morning) can also be obtained from the in-situ measurement of NO2 at the EANET monitoring sites. I wonder if the authors have tried to compare/utilize EANET data.

Specific comments:

1. All satellite-derived data under cloud-free conditions (cloud fraction < 20%) were used in your study. Have you treated the model data in the same manner for the sake

ACPD
13, C4876–C4881, 2013

of consistent comparison?

2. In Figures 3 and 4, reducing the horizontal resolution to 10 km (or 20 km for PRD) displays larger NO2 columns than those by enhancing the strength of NOx emissions by +20% for the BEI, PRD, JPN and KOR regions (i.e., NO2 column for Run 1 > NO2 column for Run 5) which are characterized by strong emissions occurring in a limited space. The same pattern could also be expected for the YTD region where the NOx species is also highly emitted and the analysis area in size is similar with those for BEI, PRD, JPN and KOR. But, it shows a different trend (i.e., NO2 column for Run 5 > NO2 column for Run 1). Also, as shown in Table 3, R values for YTD are relatively small compared with the values for other regions in June. Does any special feature exist for the YTD region?

3. In Table 3, unlike the consistent results for BEI and other regions (except marine areas), the R values in June and December show large inconsistencies, particularly for JPN and KOR. Authors should explain the specific reasons in the manuscript.

4. Regarding the trend (i.e., NO2 column for Run 1 > NO2 column for Run 5 for BEI, PRD, JPN and KOR) in Figures 3 and 4, as commented in the "specific comment 2", the reverse trend occurs for PRD, JPN and KOR during the winter episode (i.e., NO2 column for Run 5 > NO2 column for Run 1) as shown in Figure 7. What possible reasons exist for the different seasonal trend?

5. Normally, the NO2 columns tend to decrease from the morning (for GOME2 and SCIAMACHY) to the afternoon (OMI) because of the high photolysis rate (JNO2) of NO2 due to strong solar radiation in the afternoon. However, the unexpected features from the modeling results are found in many regions except PRD and some marine regions as shown in Figures 6 and 7. Interestingly, the trend in these exceptional areas (particularly PRD) located in the lower latitude is consistent with those from the satellite observations. There is a possibility of high levels of OH radicals being present in the lower latitudes. These high levels of OH radicals enhance the NOx loss rate through

ACPD
13, C4876–C4881, 2013

the reaction, NO2 + OH + M -> HNO3. Therefore, I would like to suggest that the authors analyze the levels of OH radicals, NOx loss rates, and NO2/ NOx ratios for the diagnostic regions in order to establish ant other unexpected features.

6. In your sensitivity simulations, the effects of soil NOx emission fluxes on the tropospheric NO2 columns are significant in China, particularly over CEC, NCP, and SCN for June (Figure 3). It seems to be a non-negligible factor for the summer episode. I wonder how much soil NOx contribute to the total NOx emission fluxes over the areas.

7. The averaging kernels (AKs) allow for a direct comparison between model data and observations. When the AKs are applied, the comparison is no longer complicated by systematic biases caused by unrealistic a priori assumptions (Eskes and Boersma, 2003). I wonder whether authors tried to apply AKs to this study.

Technical correction:

1. Seasonal information of the CMAQ NO2 columns should be included in Figure 1's caption for the sake of readers' convenience. It seems to be the NO2 columns for December. (i.e., "Fig. 1. Twelve selected diagnostic rectangular regions superimposed on a map of CMAQ tropospheric NO2 columns for December at 80, 40, 20, and 10 km horizontal resolutions").

2. I understand that the emission strength in your sensitivity simulations (i.e. run 5 and 6) means the emission strength by $\pm$ 20% for only NOx species (it does not include other species). If it is, authors need to clarify this in your manuscript.

References

Brown, S. S., Ryerson, T. B., Wollny, A. G., Brock, A. C., Pletier, R., Sullivan, A. P., Weber, R. J., Bubé, W. P., Trainer, M., Meagher, J. E., Fehsenfeld, F. C., Ravishankara, A. R.: Variablility in nocturnal nitrogen oxide processing and its role in regional air quality, Science, 311, 67, DOI: 10.1126/science.1120120, 2006.

Butkovskaya, N. I., Kukui, A., Pouvesle, N., and Le Bras, G.: Formation of nitric acid

ACPD

13, C4876–C4881, 2013

in the gasphase HO2 +NO reaction: effects of temperature and water vapor, J. Phys. Chem. A, 109, 6509–6520, doi:10.1021/jp051534v, 2005.

Butkovskaya, N., Rayez, M.-T., Rayez, J.-C., Kukui, A., and Le Bras, G.: Water vapor effect on the HNO3 yield 5 in the HO2 +NO reaction: experimental and theoretical evidence, J. Phys. Chem. A, 113, 11327–11342, 2009.

Butler, T. M., Taraborrelli, D., Brühl, C., Fischer, H., Harder, H., Martinez, M., Williams, J., Lawrence, M. G., and Lelieveld, J.: Improved simulation of isoprene oxidation chemistry with the ECHAM5/MESSy chemistry-climate model: lessons from the GABRIEL airborne field campaign, Atmos. Chem. Phys., 8, 4529-4546, doi:10.5194/acp-8-4529-2008, 2008.

Davis, J. M., Bhave, P. V., and Foley, K. M.: Parameterization of N2O5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, Atmos. Chem. Phys., 8, 5295–5311, 2008.

EANET (Acid Deposition Monitoring Network in East Asia, http://www.eanet.asia/)

Eskes, H. J. and Boersma, K. F.: Averaging kernels for DOAS total-column satellite retrievals, Atmos. Chem. Phys., 3, 1285-1291, doi:10.5194/acp-3-1285-2003, 2003.

Evans, M. J. and Jacob, D. J.: Impact of new laboratory studies of N2O5 hydrolysis on global model budget of tropospheric nitrogen oxides, ozone, and OH, Journal of Geophys. Res., VOL. 32, L09813, doi:10.1029/2005GL022469, 2005.

Henderson, B. H., Pinder, R. W., Crooks, J., Cohen, R. C., Carlton, A. G., Pye, H. O. T., and Vizuete, W.: Combining Bayesian methods and aircraft observations to constrain the HOâĂć+NO2 reaction rate, Atmos. Chem. Phys., 12, 653–667, doi:10.5194/acp-12-653-2012, 2012

Lelieveld, J., Butler, T. M., Crowley, J. N., Dillon, T. J., Fischer, H., Ganzeveld, L., Harder, H., Lawrence, M. G., Martinez, M., Taraborrelli, D., and Williams, J.: Atmospheric oxidation capacity sustained by a tropical forest, Nature, 452, 737–740, 2008.

Lin, J. T., Liu, Z., Zhang, Q., Liu, H., Mao, J., and Zhuang, G.: Modeling uncertainties for tropospheric nitrogen dioxide columns affecting satellite-based inverse modeling of nitrogen oxides emissions, Atmos. Chem. Phys., 12, 12255-12275, 2012.

Mollner, A. K., Valluvadasan, 5 S., Feng, L., Sprague, M. K., Okumura, M., Milligan, D. B., Bloss, W. J., Sander, S. P., Martien, P. T., Harley, R. A., McCoy, A. B., and Carter, W. P. L.: Rate of gas phase association of hydroxyl radical and nitrogen dioxide, Science, 330, 646–649, doi:10.1126/science.1193030, 2010

Riemer, N., Vogel, H., and Vogel, B.: Impact of the heterogeneous hydrolysis of N2O5 on chemistry and nitrate aerosol formation in the lower troposphere under photosmog conditions, Journal of Geophys. Res., VOL. 108, NO. D4, 4144, doi:10.1029/2002JD002436, 2003.

Saunders, S. M., Jenkin, M. E., Derwent, R. G., and Pilling, M. J.: Protocol for the development of the Master Chemical Mechanism, MCM v3 (Part A): tropospheric degradation of nonaromatic volatile organic compounds, Atmos. Chem. Phys., 3, 161–180, doi:10.5194/acp-3-161-2003, 2003.

Stavrakou, T., Müller, J. –F., Boersma, K. F., van der A, R. J., Kurokawa, J., Ohara, T., and Zhang, Q.: Key chemical NOx sink uncertainties and how they influence top-down emissions of nitrogen oxides, Atmos. Chem. Phys. Discuss., 13, 7871-7929, 2013.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 14037, 2013.