Atmos. Chem. Phys. Discuss., 13, C6489–C6494, 2013
www.atmos-chem-phys-discuss.net/13/C6489/2013/
© Author(s) 2013. This work is distributed under
the Creative Commons Attribute 3.0 License.

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.

H. Irie et al.

hitoshi.irie@chiba-u.jp

Received and published: 4 September 2013

Reply to anonymous referee 1

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Considering comments from all three reviewers, we have decided to revise the manuscript to concentrate on June 2007, since validation results obtained by Irie et al. (2012) are based mainly on comparisons around summer (mainly June). To adequately interpret the wintertime satellite vs. CMAQ comparisons along the reviewers' comments, robust validation results for winter would be critical as suggested

in the original manuscript. Detailed responses to the comments, including statements on this major revision, are given below.

General comments:

1. 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.

Reply: As mentioned above, we have revised the manuscript to concentrate on summer (June 2007) upon comments from all three reviewers. This reviewer's comment will be helpful, when we extend our analysis in the future.

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.

Reply: We agree with the reviewer that uncertainty in these chemical NOx sinks is important. In the original manuscript, complicated results seen in December 2007 due to the lack of robust validation results prevented us from discussing in detail potential causes, including these chemical NOx sinks. We have revised the manuscript to concentrate on June 2007 and included a discussion on the uncertainty in chemical NOx sinks. We now mention the importance of evaluating chemical NOx sinks, in support of studies by Lin et al. (2012) and Stavrakou et al. (2013).

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.

Reply: We agree with the reviewer. However, our results on the diurnal variation pattern between AM and PM is unlikely sensitive to uncertainty in emissions, as discussed in the revised manuscript. Then, we argue that our study supports the need of evaluating planetary boundary layer mixing and chemical processes in a model. To identify the exact causes as a next step, we also think that additional comparisons with data other than satellite data will be useful as the reviewer suggests.

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 of consistent comparison?

Reply: For each diagnostic region, we have used monthly mean VCDs calculated from all CMAQ outputs without any cloud filtering in June 2007. For an adequate cloud filtering, we are afraid that accurate modeled cloud fields may be needed. On the other hand, we tested the dependence of satellite-derived monthly-mean VCDs on the cloud fraction (CF) threshold and found that differences between monthly-means from all data (CF<=100%) and data obtained under cloud-free conditions (CF < 20%) were as small as less than 20%. So, we have done consistent comparisons within uncertainty

less than 20% for this effect.

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?

Reply: According to this comment, we have added emission maps (Fig. 7) showing that the strong emission area in YTD is not similar but larger than those of BEI, PRD, and JPN. This can explain the features seen in YTD.

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.

Reply: We believe that it is very difficult to explain the specific reasons at this stage due to the lack of robust validation results in winter as mentioned in the original manuscript and above. Since the revised manuscript now concentrates on June 2007, we do not state differences between June and December.

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?

Reply: We believe that difficulty in explaining this has come from the lack of robust validation results in winter. We do not discuss the differences between June and December, since the revised manuscript now concentrates on June 2007, as mentioned above.

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 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.

Reply: We are also interested in such unexpected features seen for some areas in winter. As a result of the revision excluding results for winter, however, all modeling results now show an expected decrease from the morning to the afternoon. However, information of OH is important and is now given in the revised manuscript.

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.

Reply: Following this comment, we have added the sentence "For CEC in June 2007, for example, soil NOx contributes to ∼11% of the total NOx emission fluxes." in the revised manuscript.

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

Reply: Ideally, the true tropospheric NO2 VCD should be used to validate a model. While such a true VCD was unavailable, satellite data after correcting potential systematic biases is regarded as the true VCD. Since a modeled VCD degraded with AKs can depart from the true VCD, we simply perform comparisons without applying AKs.

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").

Reply: We agree with the reviewer. In the revised manuscript, this figure has been replaced with a new figure, in which no CMAQ NO2 data are shown.

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

Reply: We have revised the manuscript to clarify this point. The revised manuscript now states "For the sensitivity to emissions, we simply change the emission strength for all sources of NOx by ±20% over the whole of the model domain."

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