

Interactive comment on “An evaluation of the CMAQ reproducibility of satellite tropospheric NO₂ column observations at different local times over East Asia” by H. Irie et al.

Anonymous Referee #1

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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-

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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 NO_x sink in the atmosphere (Lin et al., 2012; Stavrou 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 NO_x emissions, uncertainty in these chemical NO_x 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

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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 NO_x 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 NO_x 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 NO_x loss rate through

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

6. In your sensitivity simulations, the effects of soil NO_x 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 NO_x contribute to the total NO_x 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 ± 20% for only NO_x species (it does not include other species). If it is, authors need to clarify this in your manuscript.

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