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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.

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Reply to anonymous referee 3

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:

(A) As pointed out by other reviewers, the authors should expand their analyses considering various uncertainties associated with model simulation. Several questions remain unanswered. For example, how do CMAQ simulations with various emissions and loss processes compare with satellite retrievals? Does CMAQ include all NOx emissions (e.g. lightning and aircraft NOx emissions)? You have changed emissions by 20%. Are there any justifications for selecting this value? Do you change emissions by 20% for all sources or just for anthropogenic sources? What are the resolutions of emissions? How reliable is mixing height depth in CMAQ and how does that affect the diurnal variation? Are there any issues in CMAQ regarding loss processes?

Reply: According to the reviewer's comments, we have made revision to clearly state that comparisons are mainly made with data from the CMAQ base case simulations at a 80-km horizontal resolution. In our CMAQ simulations, aircraft emissions below an altitude of 1 km are included but lighting NOx emissions are not included. This is now mentioned in the revised manuscript. A 20% change in emissions has been tested as an increase in the emission strength by 20% leads to an increase in NO2 VCDs by an amount comparable to that found when the spatial resolution is improved from 80 to 10 km for CEC, as stated in section 2. We simply change the emission strength for all sources of NOx by $\pm$20% over the whole of the model domain. We have revised the section 2 to clearly mention the spatial resolution of emissions (0.25 deg x 0.25 deg). Discussion of potential issues in CMAQ regarding mixing height depth and loss processes is now added and the need of their evaluation is suggested.

(B) Comparison of model simulation with satellite retrievals is affected by a-priori profiles used in the retrievals. Satellite algorithms require a-priori NO2 vertical profiles which are taken from a global simulation at resolution much coarser than offered by

regional models such as CMAQ. Besides, the profiles used in retrievals may be based on old emissions. Asian emissions are undergoing rapid changes which likely affect NO2 vertical profile shapes, to which satellite NO2 retrievals are sensitive. This may be a large effect, which needs to be accounted for while comparing model simulations with satellite retrievals. Moreover, the effect could be seasonally varying. Therefore, I suggest authors to recalculate tropospheric NO2 from satellite using CMAQ profiles (using vertical sensitivity such as averaging kernel information), and then repeat the analyses.

Reply: We agree with the reviewer that the tropospheric NO2 from satellite observations depends on a priori profiles used in the retrieval. In the present study, however, we have considered potential systematic biases based on the work by Irie et al. (2012) and satellite data corrected for such potential biases have been used to compare with CMAQ. Since possible effects by a priori should have been accounted for in this bias correction, we use the same satellite data sets as in the original manuscript.

(C) From the discussion, methodology is not clear enough. There might be spatial and temporal differences in sampling between model and satellite observations. The three satellite instruments have different foot-print size. For OMI, the pixel size changes with scan position. Different pixel sizes lead to different levels of spatial smearing. Model spatial resolution is different from satellite spatial resolution and they hardly match in spatial coverage. How did you account for those effects? Local time and local equator crossing time may differ by as much as 1 hour. You indicated that model simulations were interpolated to local times and from table 3, it appears that the temporal interpolations are for local equator crossing times, not actual local times. If the interpolation was performed for actual time of measurements which differ each day within a repeat cycle of the satellite, it should be clarified. For winter time, satellite retrievals may be affected by ice and snow. Cloud parameters and therefore satellite retrievals are likely erroneous in those conditions. Excluding such observations may lead to limited number of satellite observations to compare with model. Same cloud fraction threshold applied to

different satellite may affect the sampling size. While creating monthly means, did you use the same selection criteria for model as for satellite observations?

Reply: We also think that the spatial and temporal differences in sampling between model and satellite observations are important. We have selected diagnostic regions to be wider than any horizontal resolutions of satellite observations and CMAQ simulations, in order to discuss satellite vs. CMAQ comparisons under conditions with a similar spatial representativeness over the area of interest. Also, to increase the representativeness over time as well, monthly-mean tropospheric NO2 VCDs from satellite observations and CMAQ simulations are compared for each of the diagnostic regions. Moreover, different cloud fraction thresholds and different areas of regions (BEI, NCP, and CEC) have been tested. These are stated in the revised manuscript. Regarding the time for interpolation, the revised manuscript now clearly states "For all grids, the model values are interpolated over time to estimate tropospheric NO2 VCDs at local times of 9:30, 10:00, and 13:45 LT for comparisons with satellite data." The reviewer's concern for satellite data in winter is not an issue now, since the revised manuscript concentrates on summer only. We use the same selection criteria for three satellite data sets but this is not an issue according to the result of the test for various cloud fraction thresholds and different areas of regions (BEI, NCP, and CEC). For each diagnostic region, monthly mean VCDs were calculated using 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%. Thus, we have done consistent comparisons within uncertainty less than 20%.

SPECIFIC COMMENTS:

1. Section 2 (CMAQ): Does CMAQ include emissions from lightning and aircraft? How do these emissions affect the observed discrepancies between satellite retrievals and

model simulation and the discrepancies in diurnal changes?

Reply: Aircraft emissions below an altitude of 1 km are included but lighting NOx emissions are not included. This is stated in the revised manuscript. On monthly basis, emission from lightning and aircrafts above 1 km are likely minor compared to anthropogenic emissions over our diagnostic regions that cover major cities of China, Korea, and Japan, as seen in Fig 3 of the revised manuscript. However, they may be important in interpreting the daytime variation of NO2 VCD, particularly over oceans. This possibility is now stated in the revised manuscript.

2. What is the spatial resolution of emissions? What is the temporal resolution of emissions and where does this information come from?

Reply: We have used monthly-gridded REAS version 2 data with a 0.25 deg x 0.25 deg resolution. This information come from the paper of Kurokawa et al. (2013). This is now clearly stated in the manuscript.

3. What is the year of anthropogenic emissions (REAS)? How does it compare with other emission inventories (e.g. from Streets et al and Zhang et al)?

Reply: The revised manuscript now states that we use REAS version 2 data for 2007. Also, we have added the sentence "REAS Version 2 NOx emissions from China are similar to those of Zhang et al. (2007, 2009) for 2000-2004 and 2006 but larger than those of EDGAR 4.2 (EC-JRC/PBL, 2011) especially after 2005 (Kurokawa et al., 2013)."

4. Page 14043, line 2: Please, provide average equator crossing time, not the range as the information could be misleading.

Reply: The sentence has been revised.

5. Page 14043, lines 11-12: Wasn't 1x10^{15} molec cm^{-2} the error in slant column, not the vertical column?

Reply: This sentence has been deleted.

6.Page 14044, lines 7-8: You brought the issue of satellite instrument degradation. Are there any instrument degradation effects in satellite retrievals of NO2?

Reply: For example, the so-called row anomaly has occurred for the OMI instrument and showed a change with time. Its effect can be significant and unreliable data affected by this problem have been screened out.

7. Page 14044, line 10: Why are these regions selected? Was the selection motivated by any issues related to emissions or retrievals?

Reply: Within a relatively small domain adopted for our CMAQ simulation, we have attempted to cover major cities, where strong emissions are expected, and oceans where influences of background/ship-emissions are expected.

8. Page 14044, lines 24:26: Are there any reasons for the increase in tropospheric NO2 retrievals with cloud fraction? Why was 0.2 cloud fraction threshold was selected?

Reply: At a larger cloud fraction threshold, more data are used. At the same time, a larger influence by the a priori used in the satellite retrieval is expected. This is now mentioned in the revised manuscript. A cloud fraction threshold of 20% was selected to ensure moderate quality and a sufficient number of data points, as stated in the text.

9. Page 14049, lines 14-17: This statement may be incorrect. Should not stratospheric NO2 be larger in summer than in winter?

Reply: In the revised manuscript, this part has been deleted.

10. Figure 1: What is the sampling time of CMAQ simulation? Which day, month, year?

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

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