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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.

H. Irie et al.

hitoshi.irie@chiba-u.jp

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Reply to anonymous referee 2

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:

P14042, l8-10: Your selection criterion for the sensitivity experiments seems ad-hoc and limited. It would be good to put this study better into perspective of other sensitivity studies using retrievals, such as Lin et al. (2012) and others.

Reply: According to this comment, we have revised the manuscript to clearly state that our sensitivity simulations have been carried out from aspects of the spatial resolution and emission. This is different from but complementary with Lin et al. (2012), who made sensitivity experiments from aspects of meteorological and chemical parameters/processes. Then, the revised manuscript now mentions the need of evaluating planetary boundary layer mixing and chemical NOx sinks, in support of studies by Lin et al. (2012) and Stavrakov et al. (2013).

P14044, l26: "The largest difference of NO2 VCDs with respect to the value at a cloud fraction of 20% is found to be < 30%, which is much smaller than the quoted uncertainty in the satellite retrievals". I understood from P14043, l12 that uncertainty in satellite retrievals is actually ∼ 30%, so in the order of magnitude of the satellite retrieval uncertainty. Please explain. Furthermore, here you provide statistics for the largest region (CEC), where one may expect some smoothing from the regional averaging. I wonder to what extent numbers change if smaller regions with large pollution sources, like 'BEI', are selected. Such sites may additionally contain a considerably less amount of individual observations due to combined aerosol / cloud shielding. The question is whether these (small) regions contain sufficient pixels with observations to be able to provide quantitative statements.

Reply: As pointed out by the reviewer, the general sentence about a satellite retrieval error of about 30% does not match quoted uncertainties in the satellite retrievals. So, we have deleted this general sentence from the revised manuscript. Also, statics (the

dependence of monthly-mean NO2 VCD on the cloud fraction threshold) for BEI has been added in the revised manuscript to indicate that sufficient pixels are contained even in BEI.

P14045,l10: "The associated error bars represent simple averages of quoted uncertainties in the satellite swath data used for monthly-mean calculations.": It appears from the error bars in the figures that the uncertainty for GOME and SCIAMACHY observations is ∼60%, rather than 30%, as discussed in the introduction. Could you give more information on these numbers and provide actual uncertainty estimates for the various instruments?

Reply: As mentioned above, the general sentence about a satellite retrieval error of about 30% has been deleted in the revised manuscript.

P14045, l28:"the difference is likely insignificant": As you probably rightly acknowledge that the difference between GOME and SCIAMACHY is insignificant, this also suggests that the difference between NO2 VCD's from those instruments and OMI is insignificant. Hence it is very hard to make quantitative statements on the diurnal variation. In fact, why not validate the models directly using MAX-DOAS if you are interested in diurnal cycles. Can you comment?

Reply: According to the reviewer's comment, we have re-examined potential biases in satellite data and now we do not think that the difference between GOME-2 and SCIAMACHY NO2 VCDs is insignificant. By this revision, we now 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.

P14047, l5: You conclude that diurnal cycle in emissions do not always produce better agreement with measurements. Could you give some more comments on this? I would believe that including a better representation of NOx emissions would be rather

important to get better match to the retrievals. But from your analysis this seems not the case, or hardly anything changes.

Reply: In the revised manuscript, we now state "Considering the diurnal variation of emissions based on the diurnal variation pattern estimated by Lin et al. (2010), the AM and PM NO2 VCD values tend to decrease and increase, respectively. In CEC (BEI), for example, the AM NO2 VCD decreases by about 7% (9%) due mainly to a reduction of nighttime emissions, whereas the PM NO2 VCD increases by about 6% (6%) due mainly to an increase in daytime emissions. The changes in NO2 VCDs are smaller than the emission changes (a ∼40% reduction at 00:00-04:00 LT and a ∼20% increase at 09:00-19:00 LT), due to a partial offset by effects of changes in daytime and nighttime emissions."

P14048, l2: "are reproduced well for all 12 regions": This seems not true: E.g. for regions ECS, SOJ and PRD, where diurnal variation is quite different. It might be better to introduce the quantitative evaluation (Tables 4/5) in the respective section (i.e., here) to diagnose the diurnal variation.

Reply: This sentence has been deleted in the revised manuscript.

P14048, l13: "larger" shouldn't this be "smaller"?

Reply: This sentence has been deleted in the revised manuscript.

P14049, l13-14: "negatives": The fact that there are 'some negatives' is not sufficient to suggest that there is an issue with the retrieval algorithm, and should be removed. Individual pixels are allowed to give negative values, to compensate for pixels with too positive values. This is simply due to the natural scatter in the observations, and relate to the observational uncertainty. At this stage one might equally well conclude that the model has problems with getting the NO2 right in winter. Please correct.

Reply: This sentence has been deleted in the revised manuscript.

P14049, l20: "It is thought. . .": It would be good to get more indications of what

changes in CMAQ chemistry when increasing the resolution. Now things are unclear and do not add to the understanding. E.g., do you see more O3 production? Do you better capture spatial variation in observed NO2 (see comment on correlation statistics)? Again, why not directly compare to MAX-DOAS observations to obtain a clear evaluation of the diurnal cycle, and impact of increased resolution?

Reply: According to this comment, we have added the sentences "At strong point sources, increasing NOx emissions at finer resolutions results in a decrease of OH and a longer NO2 lifetime (Valin et al., 2011). Indeed, for CEC (BEI) at 13:45 LT, for example, CMAQ-simulated mean OH concentration below an altitude of 1 km decreases from 8.1x10^6 (7.6x10^6) to 6.6x10^6 (5.0x10^6) molecules cm^{-3}, when the horizontal resolution is improved from 80 to 10 km. Correspondingly, the NO2 VCD increases from 5.7x10^{15} (7.3x10^{15}) to 7.2x10^{15} (15.1x10^{15}) molecules cm^{-2}." Additional revision has been made to add the figure (Fig. 3 of the revised manuscript) that compares spatial distributions of monthly-mean tropospheric NO2 VCDs from CMAQ simulations (at 4 different horizontal resolutions) and satellite observations for June 2007. Regarding the comparison with MAX-DOAS, our response is already given above.

P14050, l9: If the authors conclude that satellite observations are insufficiently constrained to be used for quantitative statements on the diurnal variation then it is questionable how we should interpret the current results, and what we can learn from this. Please comment.

Reply: In the revised manuscript, we do not conclude that satellite observations are insufficiently constrained to be used for quantitative statements on the diurnal variation, as explained above.

P14050, l20: "Quantitative agreement . . . are taken into account": what do you mean with this sentence? In what sense did you take model uncertainties into account for the quantitative agreement? It is also unclear which simulation is performing best or whether you find suggestions for possible model biases or biases in the satellite re-

trievals. It is disappointing that no reasons for the discrepancies are identified. Would you suggest that there are biases in the emissions? Biases in the retrieval? Biases in CMAQ? By just reporting the discrepancies it is difficult to learn something from this. How do other models perform? Is it a general feature? Could there be issues with representativity of the results due to small sampling areas combined with short time periods for analysis? With so many open questions it is difficult to accept this manuscript for publication. For example, it would be good to see some more basic statistics, including maps of satellite NO2 from the 3 products for June and December, and corresponding numbers for the spatial correlation of the monthly mean fields, in order to assess how this changes (improves?) with increased resolution and for different instruments, having all amongst others different pixel sizes. It could give an indication of whether the three instruments see the same features.

Reply: Following these comments, the conclusion section has been revised to clearly state "Within uncertainty in satellite NO2 VCD data, CMAQ (as the base run at a 80-km horizontal resolution) generally reproduced absolute values of monthly-mean satellite NO2 VCDs over most of 12 selected diagnostic regions." Regarding reasons for the differences between CMAQ and satellite data, we have revised the manuscript to state the need of detailed evaluation of planetary boundary layer mixing and chemical processes and suggest the use of recent available rate constants for NO2 + OH + M $\rightarrow$ HNO3 + M and/or the inclusion of HNO3-forming channel NO + HO2 $\rightarrow$ HNO3 for better simulation by a model. The present study uses only CMAQ but various sensitivity simulations have been performed. We hope that this study encourages other models to be evaluated similarly from the viewpoint of diurnal variations using multiple satellite observation data sets. Regarding the generality and the representativity, we realize that this is a case study but different sampling scales have been tested (e.g., BEI, NCP, and CEC). Also, the reasons for choosing June 2007 have been clearly stated in the text. Note that a similar underestimation of daytime NO2 losses is suggested by Itahashi et al. (2013) for other years. This is now mentioned in the revised manuscript. Moreover, revision has been made to add the figure (Fig. 3 of the revised version) that compares

spatial distributions of monthly-mean tropospheric NO2 VCDs from CMAQ simulations (at 4 different horizontal resolutions) and satellite observations for June 2007, in order to see how the spatial distributions compare among them.

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