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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluating the accuracy of NO_x emission fluxes over East Asia by comparison between CMAQ-simulated and OMI-retrieved NO_2 columns with the application of averaging kernels from the KNMI algorithm" *by* K. M. Han et al.

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First of all, thank you for your valuable comments and suggestions. Based on three reviewers' comments, we attempted to improve our manuscript by eliminating, modifying, and adding many parts from/into the original text (the added or modified parts are painted in a red color in the revised manuscript). Major changes made in the revised manuscript are as follows:

- Change of the title.

- Less emphasis on applying AKs to CMAQ model simulations.

- Restructure of the manuscript to clarify our motivations and conclusions of this study.

- More quantitative description of statistical analysis and comparison of our results with those from other studies.

- Re-calculation and re-plotting of Figures and Tables, since applying AKs were carried out over the satellite footprint.

The authors present two main topics: (1) the necessity of using averaging kernels when comparing model simulations of NO2 tropospheric columns with satellite retrievals. (2) the sensitivity of the modeled NO2 columns to simulation parameters (seasonal cycle; alternative emission inventory; reaction rate N2O5). In its current form I do not consider it fit for publication in ACP. The paper needs serious restructuring to better present its scientific relevance. The revision should be more concise.

GENERAL COMMENTS

I miss a well described motivation for the presented study. Does it concentrate on the importance of the use of averaging kernels in comparison studies (which is obvious for the satellite community, but apparently less obvious for the modelling community)? Does it want to correct previous work (e.g. by Han et al., 2011) which did not take into account the AKs? Does it want to show that emission inventories in East Asia are wrong or out-dated? Does it want to contribute to other sensitivity studies analyzing the model error in reproducing NO2 columns (e.g. Lin et al., 2012)? Does it want to do so to improve future top-down emission estimates with satellite observations? These motivations are all hidden in the text, but should be stated more clearly. The addressed scientific questions should dominate the structure of this paper and its analysis.

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Reply: Based on your comments, we tried to clarify our motivations in Sect.1. Our basic motivation/goal of this study is to evaluate the NOx emission inventories used in East Asia, applying a state-of-the-science knowledge and methods including the uses of the AKs and OMI-retrieved tropospheric NO2 columns (please, see p. 5, lines 118 – 127).

Satellite retrievals are not the truth, and can also be biased. So the cause of differences between model and observation can be found in both. Uncertainties in satellite retrievals are first mentioned in the end Section 3.2.4, but should be given a more prominent place in the analysis of the results.

Reply: We discussed the uncertainties of the NO2 retrievals in Section 2.2. The uncertainty for the tropospheric NO2 columns of the KNMI/DOMINO v2.0 used in this study is approximately 1.0×1E+15 molecule/cm2 with a 25% relative error (Boersma et al., 2011). In the revised manuscript, we also described (and further clarified) that we reduced the random and smoothing errors of the satellite NO2 columns via seasonal averaging and applying the AKs to the CMAQ model simulations, respectively. Please, check out p. 9, lines 229-234 and p. 11, lines 268-284 for further detail.

The method of applying the averaging kernels on the model simulations is only briefly explained at the end of section 2.2; Figure 4 is hardly discussed. What I understand is that the OMI observations are horizontally gridded to the model grid. Why not doing the opposite: interpolating the model values to each satellite footprint? This makes a fairer comparison as the averaging kernel is associated with the footprint area of the observation, and not with a model grid cell.

Reply: Yes, it is a better idea, although no large differences are found. We applied the AKs to the model simulations over the OMI footprint areas. The detail procedures

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were discussed in Sect. 2.2 (please, see p. 10, lines 257-267). Also, we corrected all the relevant Figures (particularly, Fig. 4) and Tables related to this issue in the revised manuscript.

SPECIFIC COMMENTS

Title: The title represents only one side of the study, and neglects the performed sensitivity analysis.

Reply: Considering two reviewers comments, we changed the title.

Abstract, P17587, line 1: Please mention which inventories are used for the evaluation.

Reply: We used three emission inventories: INTEX-B for China; CAPSS for Korea; and REAS v1.11 for Japan (Please, see p.2, lines 43-46).

Abstract, P17587, line 1: Some indication of the geographical extent of the used East Asian domain would be nice.

Reply: We put the geographical extent of our study domain into abstract (Please, see p.2, line 43).

Abstract, P17587, line 13: "28%": using which emission inventory? Can the difference be attributed to wrong emissions?

Reply: The NOx emissions from the INTEX-B, CAPSS, and REAS inventories used in this study were "possibly" underestimated in East Asia, but obviously there can also be uncertainties/errors in the satellite-derived NO2 columns, AKs, CMAQ model simulations, etc. The CMAQ-calculated NO2 columns were, on annual average, ~28%

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(in terms of “Normalized Mean Bias”) underestimated, compared with the OMI NO2 columns. We clarified this point in the revised manuscript (Please, see p. 2, lines 53-56).

1, P17588, line 14: “Han and Song, 2012” were not the first ones to find out about the importance of this removal process. Maybe an earlier reference is more appropriate.

Reply: We added more references: McConnel and McElory, 1973; Platt et al., 1984; Dentener and Crutzen, 1993; Brown et al., 2006 (p. 3, lines 82-84).

1, P17588, line 4-6: To compensate for height dependent sensitivities, the column retrieval algorithm depends on cloud information and an assumed NO2 profile. If this NO2 profile reflects reality, the retrievals can be compared directly with simulations (and are in that sense “real” or “true”). If the true NO2 profile is different, the averaging kernel of the retrieval method should be applied to the simulation to compensate for this effect.

Reply: It may be p.“17589”, lines 4-6. Thank you for your detailed comment. Yes, if we used “true/real” profiles, we would not need to apply the AKs. If the true profile is not utilized in the NO2 retrieval, the AKs should be applied to correct the systematic biases caused by unrealistic a priori assumptions. Following your comments, we added this point in the revised manuscript (Please, see p. 4, lines 103-110).

1, P17589, line 9. Consider a definition Ω instead of ΩNO2 throughout the whole paper, as NO2 columns are the only columns studied in this paper. This will increase the readability of the symbolized quantities used in the text.

Reply: Thank you for your kind suggestion. We changed them throughout the revised

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manuscript.

2, P17590, line 1-5: Leave out. This should be clear by now.

Reply: We removed it (Please, see p. 5, line140).

2.1, P17590, line 8-9: “because relatively (: : :) this year”. Better: because INTEX-B was compiled for this year.

Reply: We changed it (p. 7, lines 173-174).

2.1, P17592, line 1-2: “modeling conditions” → model setup

Reply: We changed it (p. 8, line 193).

2.1, P17593, line 20: Change x ÌC′ -x_a to x ÌC′ -x ÌC′ _a, to differentiate x ÌC′ _a being a column quantity and x_a a vector quantity.

Reply: We corrected it in Equation (2) (p. 10, line 243).

2.1, P17594, line 10: The change of the AKs over the seasons can also be related to cloud climatology, especially because it is not clear from the text that the observations in Figure 3 have been filtered for cloud radiance.

Reply: Although we filtered all the data with cloud radiance fraction (CRF) larger than 50% (i.e. it is under almost “cloud-free” condition), AKs can be influenced by the presence of some clouds. We added the parameter in the revised manuscript (Please, see p. 9, lines 224-226 and p. 10, line 255).

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3.1.1, P17595, line 3. Consider writing the section title in words instead of symbols.

Reply: Thank you again! We changed the section-title in words, instead of symbols (Please, see p. 12, lines 295-256).

3.1.1, P17595, line 20-21: “possibly” can be left out. “Han et al., 2009” were not the first ones to find out about this removal process. Maybe an earlier reference is more appropriate.

Reply: We removed the “possibly” and added more references (Please, see p. 12, lines 312-314).

3.1.1, P17596, line 6-11: Apparently this is an important motivation to conduct this study. Therefore, it should be given a more prominent position, for instance in the Abstract or Introduction.

Reply: We now mention this point for our motivations in Sect. 1 (please, see p. 4, lines 118 – 123).

3.1.1, P17596, line 13-14: “(: : :) correct previous conclusions”. This conclusion should therefore also be mentioned in the Conclusion section.

Reply: In conclusion, we mention this paragraph which is major finding in the study (see p. 26, lines 647-651).

3.1.1, P17596, line 28: Mention that the NME is defined in Table A1.

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Reply: We put the following sentence into the text. “The NME was defined in Table A1” (see p. 14 line 344).

3.1.2, P17597, line 10-11, Figure 7: I do not understand why I see in each panel so few scatter points. With a model resolution of 30 km2 and comparable OMI footprint resolution each focus region contains dozens of grid cells / observations, which sum up in a three-month period to hundreds of data pairs. Please explain in more detail how a data pair is established.

Reply: In Fig. 6, we used the season-averaged data set of the two tropospheric NO2 columns, mainly because averaging data can reduce random errors in the satellite-derived tropospheric NO2 columns. For a better understanding, we conducted the scatter plot analysis using daily data set (number of data from $\sim$300000 to $\sim$ 500000 for the entire domain) over the entire domain in Fig. S1. (Please, refer to Fig. S1 and see p. 14, lines 361-366).

3.1.2, P17597, line 20-26: A lot of different statistical quantities are introduced here; not all of them are familiar to everyone. Different quantities highlight a different aspect of how model and observation compare. Maybe it is an idea to describe in Table A1 (or elsewhere in the Appendix) for each quantity its specific use in comparing model with observation.

Reply: Thank you for your recommendation. We tried to describe the (dis)advantages in the uses of those statistical parameters. For example, there is an asymmetry problem in MNB and NMB, indicating that the overestimation (i.e., $+\infty$) are weighted more than the equivalent underestimations (i.e., - 100). MFB provides equal weight to both sides (-200 to +200). The detailed explanations were added in Appendix (Please, see p. 28, lines 683-709).

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3.1.2, P17598, line 6: “between 2 x 1015” → “between -2 x 1015”

Reply: We added the negative sign and gave specific values in both sides (Please, see p. 15, line 387).

3.1.2, P17598, line 6-7: My main interpretation of the MB results would be that for all seasons the mean bias is negative almost everywhere (except for CEC2), i.e. CMAQ,AK is smaller than OMI, a strong indication that the used NOx emission inventory is underestimating the real emissions.

Reply: Yes, it is. The negative values in the MBs indicate that the NOx emissions are possibly underestimated, compared to the real NOx emissions. We clarified this point (Please, see p. 15, line 387-389).

3.1.2, P17598, line 17-19: In the abstract and conclusion the underestimation of NOx emissions is estimated to be around 28%. This is the section about statistical analysis, so here it should give more detailed information on how this number is derived.

Reply: As mentioned in the previous comment, the CMAQ-calculated NO2 columns were, on annual average, ∼28% (in terms of the Normalized Mean Bias) underestimated, compared with the OMI NO2 columns (Please, see p. 2 lines 53-56, p. 16, lines 394-395, and p. 25, lines 618-620).

3.2: This is definitely not the first sensitivity analysis. Previous work, such as by Lin et al. (2012) should be properly discussed. May be parts of 3.2.4 can be included in such an overview. The choice of why investigating the sensitivity to parameters in Case 2,3,4 should be clearly explained. And finally, how do the sensitivity results compare

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or add to existing results?

Reply: A comprehensive sensitivity analyses were conducted by Lin et al. (2012). We briefly tried to introduce the study of Lin et al. (2012) in the revised manuscript (Please, see p. 23, lines 576-585). The reasons to choose the sensitivity parameters for Cases 2, 3 and 4 were also explained /described at p. 16, lines 411-420.

3.2.1, P17599, line 3-4: It is unclear if the imposed seasonal variation is taken the same for each emission sector.

Reply: In the sensitivity runs, we applied all the same monthly factors to the sectors of power generation, residential areas, industry, and transportation. We clarified this point in the revised manuscript (Please, check out p. 7, lines 167-168).

3.2.1, P17599, line 5-7: The larger difference found in winter time could also indicate a NOx lifetime issue of the model in colder/darker environments.

Reply: In addition to the issues of NOx emission (i.e. monthly factor and different emission inventory), the NOx lifetime can also influence the large differences during winter. That is why in Sect. 3.2.3, we explored the issue of reaction probability of N2O5 onto aerosols which is one of the most important parameters for determining the NOx lifetimes during winter. Yes, the cold environments create a favorable condition for high levels of N2O5.

3.2.1, P17599, line 18-19, Figure 1: Indicate more clearly that the monthly variation of INTEX-B is taken from Zhang et al. (2009).

Reply: We clarified this point (p. 18, line 444).

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3.2.2, P17600, line 2: Which version of the REAS inventory is used, for which base year?

Reply: We clarified it. We used the REAS v1.11 emission inventory for 2006 (Ohara et al., 2007) (Please, check out p. 18, lines 458-459).

3.2.2, P17601, line 6-7: Some words about satellite derived emission inventories seem appropriate here.

Reply: Thank you for your recommendation. The top-down NOx emissions (Martin et al., 2003; Toenges-Schuller et al., 2006; Boersma et al., 2008; Stavrakou et al., 2008; Lin et al., 2010; Ghude et al., 2013; Mijling et al., 2013) using the satellite-retrieved NO2 columns based on the mass balance approach, can be applied to the 3D-CTM over East Asia in order to reduce the spatial discrepancy between the ΩCMAQ,AK and ΩOMI. However, in the revised manuscript, we excluded this issue in Sect. 3.2.2, because the top-down NOx emission is a bit different issue from the main stream of this section. Instead, in Sect. 4 (Summary and Conclusions), we discussed this issue in the context of uncertain factors discussed in Sect. 3.2.4. We believe that this issue should be a next step we have to go forward.

3.2.3, P17601, line12-14: Not necessarily true. In winter time the increased lifetime transports NOx further away from its source. This make you more sensitive to the correctness of the meteorological fields (e.g. winds).

Reply: We eliminated the sentence of "the cold months are better for conducting this study due to the uncertain tropospheric chemistry and faster NOx loss rate during the summer", because there is another issue that there are possibly large errors related to satellite retrievals and meteorological fields during cold season (Please, refer to Sect.

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3.2.3).

3.2.4, P17603, line 8-12: “Although not shown (: : :) should be investigated further”. Leave out.

Reply: We left out this part (Please, refer to Sect. 3.2.4).

3.2.4, P17603, line 13-16: “it can be suggested”, “will/may be able to help”. This can be stated stronger.

Reply: We put a stronger statement here!! (Please, check out p. 22, lines 540-543).

3.2.4, P17603, line 26: How do k1, k2, k3, and k4 relate in magnitude? This gives information about to which mixing ratio the balance between NO2 and NO is especially sensitive.

Reply: We calculated the reaction rate constants at 298K, and put some discussions at p. 22, lines 552-560.

3.2.4, P17604, line 15-16: “The uncertainties (: : :) to some degree”. Vague. Clarify or leave out.

Reply: We eliminated this part! (Please, refer to Sect. 3.2.4).

3.2.4, P17604, line 28-29: “This is why we said that the summer was not a season of major interest in this study.” Similar statements have been at earlier points in the text. Why not centralize them (e.g. in the Introduction or in Section 2), and list all

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considerations to focus only on the winter months?

Reply: Based on another reviewer's comment, we removed these statements in the revised manuscript. In Table 1, we listed several sensitivity tests focusing on the winter season (Please, refer to Table 1).

3.2.4, P17605, line 1-12: This paragraph on retrieval uncertainties could be moved forward to Section 2.2 where the satellite product is first discussed. Maybe include some description of the retrieval error to better interpret the statistical study in Section 3.1.2: tropospheric column retrievals typically have a dominant absolute error (~0.5 x1015 molec/cm2) at low values, and have a dominant relative error (30-40%) at high values.

Reply: As mentioned in the previous response, we discussed the uncertainties in the NO2 retrieval in Sect. 2.2. The errors in the NO2 retrieval can be caused by the calculations of the AMF, spectral fitting, and stratospheric slant NO2 columns. The uncertainty for the tropospheric NO2 columns of the KNMI/DOMINO v2.0 used in this study is 1.0×1E+15 molecule/cm2 with a 25% relative error (Boersma et al., 2011). Please, see p. 9, lines 229-234 for further detail.

4, P17607, line 3-15: In my opinion, this is not a conclusion of the presented study. Instead it is an important motivation to do the sensitivity analysis in Section 3, where this text could be included in the introduction. Sensitivity studies as presented in this study improve the model (or at least improve the understanding of the model error and bias) to reproduce NO2 columns. This is very important to improve the accuracy of top-down emission estimates made with satellite observations.

Reply: As mentioned previous response, we wish to keep this paragraph here in Sect. 4 (Summary and Conclusions) to suggest the direction of our research for the next

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step.

Table 1, P17618: Indicate reference year 2006

Reply: We clarified that the target year is 2006 in Table 1.

Figure 5 and Figure 6: Consider merging the two figures in a 4 x 5 panel

Reply: As reviewer pointed out, we merged two figures into a 4x5 panel (Please, refer to Fig. 5 in the revised manuscript).

Figure 7: Indication of units

Reply: We put the unit in the Caption of Fig. 6 (Please, see p.38, line 1113).

Figure 8: Use a neutral (white) color for a value range around 0.

Reply: We changed color scales in Figs. 7 and S3 in the revised manuscript. We use white color between -1 and 1. For a better visual understanding, we used light colors for good agreements and dark colors for bad agreements (Please, refer to Figs. 7 and S3).

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