

Interactive comment on “Evaluating the accuracy of NO_x emission fluxes over East Asia by comparison between CMAQ-simulated and OMI-retrieved NO₂ columns with the application of averaging kernels from the KNMI algorithm” by K. M. Han et al.

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Received and published: 21 October 2014

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:

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- 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 paper Evaluating the accuracy of NO_x emission fluxes over East Asia by comparison between CMAQ-simulated and OMI-retrieved NO₂ columns with the application of averaging kernels from the KNMI algorithm by Han et al. is a detailed description of a comparison study between modelled and measured NO₂ columns over East Asia. While the paper points out some interesting aspects, I believe that the scientific value of the study in its present form is only fair, as the authors put too much emphasis on trivial aspects and somewhat hide the scientifically valuable parts behind technicalities. Furthermore, I disagree with the main inference performed by the authors. I suggest the study to be accepted for publication in ACP, provided the following points are addressed properly.

1. General comments

1.1 Scientific relevance

The present study constitutes of four points:

1. The importance of using AKs when comparing model results to satellite measurements
2. The importance of using the correct seasonal variation in the NO_x emissions in the

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models

3. The influence of the used emission inventory on the model results

4. The influence of the used N2O5 reaction mechanism on the model results

Point 1. in itself is trivial. Anyone familiar with satellite retrievals knows about the importance of the vertical measurement sensitivity. However, given that the authors use this section 3.1.1 to correct conclusions they drew in a previous study, I can see the value in publishing this. However, the authors should focus on the scientifically interesting part of the results, namely the comparison of the measured OMI columns to the modelled columns (with an AK applied). This is interesting. The fact that the AKs improve the results is non-surprising and should not be emphasized so much.

Reply: Thank you for your comments. We tried to less emphasize the importance of using AKs in the revised manuscript and also removed some strong statements on this point in the conclusions of Sect. 3.1.1. As reviewer mentioned, using the AKs is familiar to satellite retrieval (or remote sensing) groups. In fact, our work was also initiated from several advices given by a satellite retrieval specialist. However, the concept of the AKs is not very familiar and quite obvious to majority of chemistry-transport modelers and some atmospheric scientists, particularly in East Asia. Even, in the review process of this manuscript, reviewer can witness that using the AKs is still arguable to some atmospheric scientists. Because of these reasons, we decided to leave some statements on the importance of using the AKs in this type of studies. Also, we added some paragraphs to mention that the application of the AKs is for reducing the smoothing errors, caused mainly by the biases in a priori vertical NO₂ profiles. Table S1 (in the supplementary material) is a summary of the comparison studies between satellite-retrieved and CTM-calculated NO₂ columns over “East Asia”, but all the studies were conducted “without the applications of the AKs”.

In addition, we also left some descriptions on the comparison with and without the application of the AKs in order to correct our wrong conclusions in the previous study

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(Han et al., 2011). The correction was also stated in the sections of Summary and Conclusions (Please, refer to Sect. 3.1.1 and see p. 25, lines 614-617).

Point 2 in itself is also trivial. Given a short-lived species as NO_x, it is obvious that getting the seasonal variation in NO_x emissions right is crucial to get accurate model results. Again, the authors use this section 3.2.1 to correct previous results of their own, so I can see the value in publishing this. But the authors should focus more on the result which seasonal variation leads to the best agreement between modelled and measured NO₂ columns. Maybe the authors should choose one reference seasonality (i.e., the one giving the best agreement), and then state, for each different seasonality, the degree by how much the agreement worsens. The fact that the seasonal variation is important is trivial.

Reply: Thank you for your comment. In the revised manuscript, we chose one reference monthly variation of NO_x emissions from Zhang et al. (2009) that actually produced better results than those from Han et al. (2009). Then, we put some statements that our previous results/conclusions would be wrong. In addition, some statistical analyses were also conducted (Please, see p. 18, lines 444-449 and 452-454).

Point 3 is indeed interesting; the fact that INTEX-B leads to better agreement than REAS is noteworthy.

Point 4 is also interesting and validates publication.

1.2 Inference from NO₂ columns to NO_x emissions

Throughout the manuscript, the authors repeatedly do inference from the observed NO₂ column differences onto the NO_x emissions underlying the model simulations. In doing so, the authors fail to properly acknowledge that this inference is quite challenging, due to the importance of, among others, meteorological variability (see, e.g.,

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10.5194/acp-10-2491-2010) and the importance of the NO_x lifetime. For example, in the Summary (p. 17605, l. 24–25), the authors write [. . .] NO_x emissions were [. . .] 28% [. . .] underestimated in East Asia. However, the present study does not allow this conclusion. A valid conclusion would be that the measured NO₂ columns were underestimated by that amount, and that this underestimation is likely to be caused by an underestimation in the used NO_x emission datasets. However, the methodology used in this study does not allow to quantitatively assess the amount of underestimation of the NO_x emission datasets! Due to a) the importance of meteorology and the like (see above) and b) the uncertainty in other trace gas emissions related to NO_x chemistry (i.e., VOCs), it is impossible to infer directly and quantitatively from measured NO₂ column differences onto inaccuracies in the used NO_x emission databases.

Reply: Reviewer's point is definitely right! Although the CMAQ-calculated NO₂ columns were, on annual average, ~28% (in terms of Normalized Mean Bias) smaller than the OMI NO₂ columns, it does not directly indicate that the NO_x emission fluxes were 28% underestimated, because of many uncertainties in other NO_x chemistry-related trace gas emissions, missing chemistries in CMAQ model, meteorological fields, etc. This is also what we wished to say in the original manuscript, but our intentions were not conveyed well. Anyhow, we tried to reflect reviewer's points in the revised manuscript (Please refer to p. 2, lines 53-56 and p. 25, lines 618-620).

1.3 Title

The title could be a better description of the paper's contents. Without reading the manuscript, the reader doesn't know the accuracy of which NO_x emission fluxes are being evaluated. Which kind of emission fluxes, bottom-up or top-down? Which dataset? To my understanding, it is not possible to speak of accuracy of emission fluxes; one can only speak of accuracy of a certain dataset of emission fluxes. As it turns out, the article does not assess NO_x emission fluxes at all (it cannot, at least not

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quantitatively; see my point above). Also, the AKs should not be emphasized in the title so much, as using them is a scientific necessity and not an improvement.

Reply: Considering reviewer's comments, we changed the title.

1.4 Summer/Winter

The authors repeatedly claim that cold months are better for [comparison studies] due to the uncertain tropospheric chemistry and faster NO_x loss rates during the summer (p. 17601, l. 12–14). I disagree with the authors, because they neglect the possibly higher uncertainties in the OMI data in winter. See, e.g., Figure 6 in 10.1029/2005JD006594. A revised manuscript should state this issue and should refrain from proclaiming that winter is better for comparisons.

Reply: Again, thank you for your useful comment. During summer, the NO_x loss rates are so fast that the considerations of additional NO_x emissions would hardly change the CTM-calculated NO₂ columns (see Boersma et al., 2009; Han et al., 2009). Therefore, it is difficult to evaluate the NO_x emissions using a comparison between the CTM-derived and satellite-derived NO₂ columns during summer. This is what we wished to say here! Also, as reviewers pointed out, there are other uncertainties related to the issues of pollutant transport and satellite errors during winter. We tried to reflect these points in the revised manuscript. However, we are still sure that summer is not a better season for this comparison study. We eliminated the description of "the cold months are better for conducting this study due to the uncertain tropospheric chemistry and faster NO_x loss rate during summer" and "higher values would be better for a comparison study between CMAQ and OMI-derived NO₂ columns" (Please, refer to Sects. 3.1.1 and 3.2.3).

2 Specific comments

C8335

2.1 Abstract, p. 17587

2.1.1 Abstract, lines 7–10

The authors speak of an improvement in the comparison between measurements and simulations, but they don't explicitly state which of the two simulation datasets they take as reference. While this is implicitly clear, I believe that the authors should make an effort and be as explicit as possible, to reduce possible ambiguities.

Reply: We rewrote many parts in Abstract. The statement can be found in the section of Summary and Conclusion in the revised manuscript (Please, see p. 25, lines 614-617). We intended that the NMEs between the Ω CMAQ,AK and Ω OMI (AKs applied) decreased, for example, from $\sim 98\%$ to $\sim 40\%$ during winter in East Asia, compared with the NMEs between the Ω CMAQ and Ω OMI (AK not applied).

2.1.2 Abstract, line 10

Replace "Also, the two" by "Also, measured and simulated"

Reply: We replaced "two NO₂ column" by "two tropospheric NO₂ columns from the CMAQ model simulations and OMI observations" (Please, check out p. 2, lines 48-51).

2.1.3 Abstract, line 11

What is meant by "(R=0.71–0.94)"? Please be explicit about what the range is supposed to mean.

Reply: "R=0.71-0.96" indicates that the correlation coefficients ranges from 0.71 to 0.96. We clarified it in the revised manuscript (Please, see p. 2, lines 48-51).

2.1.4 Abstract, line 11

C8336

The authors write of NO_x emissions used, but they don't say which NO_x emissions were used.

Reply: In this study, we evaluated the NO_x emissions from INTEX-B, CAPSS, and REAS v1.11 inventories over East Asia. We clarified the point in the revised manuscript (Please, see p. 2, lines 44).

2.1.5 Abstract, lines 14–17

The authors basically state that /some overestimates [of NO_x emissions] [. . .] can be influenced by [. . .] the strength of the NO_x emissions/. That's a trivial nonsense argument and should be removed.

Reply: We eliminated the statement.

2.1.6 Abstract, lines 17–19

Does this mean that in their base run, the authors used seasonally flat NO_x emissions? Why would one start with this in the first place?

Reply: In our base-case run, we used the monthly variations from Zhang et al. (2009) for China and from Han et al. (2009) for Korea and Japan (Please, see p. 6, lines 165-167). In our sensitivity run (Case 2), we applied the monthly factors from Han et al. (2009) for China, instead of those from Zhang et al. (2009) (Please, see p. 2, lines 58-61 and p. 17, lines 424-428).

2.1.7 Abstract, line 18

I don't understand the difference between different monthly variation and different strengths of the NO_x emissions.

C8337

Reply: In the revised Abstract, we tried to clarify it to remove this ambiguity (Please, see p. 2, lines 61-64, p. 17, lines 424-428, and p. 18, lines 459-463).

2.2 Introduction, p. 17589

2.2.1 I. 1

All these studies have been about satellite measurements of tropospheric columnar NO₂, not of mixing ratios of NO_x.

Reply: We changed 'NO_x' to 'column NO₂' (Please, see p. 4, lines 95 and 100).

2.3 Section 2.1

The authors should be more explicit about the horizontal and temporal resolution of the input datasets. They state that the CMAQ model runs on 30x30km², but the following points are important and should be explicitly stated:

- What is the horizontal resolution of the emission datasets?
- Which year do the emission datasets represent?
- Do the emission datasets show seasonal behaviour, or is it just one value per grid box?

Furthermore, the authors should describe their collocation criteria for model grid boxes and satellite measurements. Do they bin the satellite observations into the model grid? Or do they interpolate from the model grid to the spacetime coordinates of the satellite measurements? If so, how?

Reply: We clarified the horizontal resolutions, base year, and seasonal factors of the emissions in the revised manuscript (Please, see p. 6, lines 150-153 and 1533-155).

C8338

We applied the AKs to the model simulations over the OMI footprint areas. The detail method was discussed in Sect. 2.2 (please, see p. 10, lines 257-267). Accordingly, we corrected all the relevant Figures (particularly, Fig. 4) and Tables related to this issue in the revised manuscript.

2.4 Section 2.2

2.4.1 OMI spatial resolution

The authors really should state the OMI spatial resolution as up to 13x24km² at nadir, because towards the edges of the scan, the spatial resolution becomes significantly lower.

Reply: We corrected it (see p. 8, line 206).

2.4.2 Stratospheric correction

The authors should state that the TM4 CTM used for stratospheric correction assimilates the OMI measured slant columns.

Reply: We added this point (Please, see p. 9, lines 216-217).

2.4.3 Data filtering based on surface albedo

The authors don't state which surface albedo dataset is being used. Specifically, it is unclear whether they use a climatological dataset or actual measurements; consequently, it is unclear if measurements affected by snow/ice cover on the surface are being excluded from further analysis.

Reply: We added the dataset used for surface albedo, which is from the OMI observations, too (Kleipool et al., 2008) (Please, see p. 8, lines 223-224).

C8339

2.5 Figure 3

- provide x labels also for the right column of plots
- place the legend outside the first (top-left) plot and into the empty space on the bottom right, or put a legend into each of the seven plots.
- in the Figure caption, give reference to Fig. 2 for the region definitions

Reply: We corrected the x-labels, legend, and figure caption, as reviewer pointed out (Please, refer to Fig. 3 and figure caption).

2.6 Section 3.1.1, p. 17596

2.6.1 l. 3–4

CMAQ NO₂ columns are not greatly larger [. . .] over the entire domain. According to Fig. 5, this is only the case for strong sources regions. For the background regions and over the Oceans (apart from continental outflow), I don't see significant differences.

Reply: The CMAQ NO₂ columns are greatly larger than OMI NO₂ columns over all the analysis regions except for the DM (entire domain). We corrected this point in the revised manuscript (Please, see p. 13, line 317).

2.7 Figure 6

I'm unhappy with the colorscale in Fig. 6. The gray color for values between -4 and 0 is quite distinct from both the blues for values < -4 and the yellows/reds for values > 0. Consequently, the gray suggests that it's a neutral color, while in fact, the zero is between the gray and the yellow. I suggest the authors change the used colorscale so that a neutral color like gray is used for small absolute values, symmetrically around

C8340

zero, e.g., from -2 to +2.

Reply: Based on your comment, we changed color scales of Fig. 5-d and 5-e in the revised manuscript. We used a gray color for the range between -2 and 2 (Please, refer to Fig. 5)

2.8 Figure 7

I have trouble understanding Figure 7. For example, looking at the DJF values for region SB, the slope is 0.98. On the other hand, comparing to Fig. 6b, virtually all of region SB in DJF is yellow, i.e., > 0. If for the whole region, CMAQ NO₂ is larger than OMI NO₂, how can it be that the regression slope is still < 1.0? I urge the authors to double-check that their calculations are correct.

Reply: In order to clarify this point, we added the y-intercepts. In the low values of NO₂ columns (< 1 × 10¹⁶ molecules/cm²) in Fig. 6, most data are scattered above the 1:1 line. In contrast, in the large values of NO₂ columns (> 1 × 10¹⁶ molecules/cm²), most data are scattered below the 1:1 line. We believe that the large values of the OMI NO₂ columns made the low regression slope (< 1.0) and large y-intercept in the SB region (Please, refer to Fig. 6).

2.9 Figure 8

Again a comment about the color scale: At first sight, the reader is a bit challenged with understanding this plot. I would suggest two things:

- Invert the color scale for R and IOA such that good values are lighter and bad values are darker.
- Add a note to the Figure caption / discussion that light colors show good agreement and dark colors show bad agreement

C8341

- Add a note to the Figure that red and blue colors indicate under and overestimation of the actual NO₂ columns for the appropriate measures.

Reply: Based on two reviewers' comments, we changed color scales in Figs. 7 and S3 in the revised manuscript. We use white color between -1 and 1. For the sake of readers' understanding, we clarified that light colors are good agreements and dark colors are bad agreements (Please, see p.15, lines 379-380).

2.10 Section 3.2.2

The authors write that the REAS inventory does not include monthly variation (l. 5–6 on p. 17600). I'm confused by this statement. When looking at the REAS v2.1 data files for NO_x, they do indeed contain 12 values, one for each month. So I disagree with the authors' statement in the current form and urge them to use the seasonal variation present in the REAS emission data. If the authors happen to have used an older version of REAS which may did not include seasonal variation, they should explicitly say so and give reference to the version they used. Along these lines, the authors should clearly state the version numbers of the emission datasets they used. For example, the INTEX-B v1.1 data files which I can download on the web do not contain seasonally varying NO_x emissions.

Reply: We used the REAS v1.11 emission data. The REAS v1.11 (annual) emission data does not include seasonal variation of the NO_x emissions. So, we clarified this point (Please, see p. 18, lines 458-459).

3 Small Corrections

3.1 Introduction, p. 17588

3.1.1 I. 10

C8342

in East Asia insted of in East Asian

Reply: We corrected it (Please, see p. 3, line 79).

3.1.2 I. 20

future GAINS simulations sounds like the authors refer to GAINS simulations run in the future, however I doubt this is what they mean.

Reply: We intended that several emission scenarios are applied to GAINS simulations for the target years between 2015 and 2035. We think that the "future" does not need to be mentioned (see p. 3, line 90).

3.1.3 I. 22

remove also

Reply: We removed it (see p. 4, line 92).

3.1.4 I. 27

The authors should also list some more recent references, e.g., 10.1029/2012JD017571 and 10.5194/acp-13-4145-2013.

Reply: We added those references in the revised manuscript (Please, see p. 4, line 98).

3.2 Introduction, p. 17589

3.2.1 I. 8

C8343

The authors should specify what exactly they mean by ΩNO_2 , i.e., if they refer to total or tropospheric columns.

Reply: We clarified the definition of Ω that indicates “tropospheric NO₂ vertical columns” from CTM simulations and satellite observations (Please, see p. 4, line 106-107).

3.2.2 I. 11–12

interpreting [. . .] ΩNO_2 [. . .] near the surface doesn't make any sense, as ΩNO_2 is a quantity integrated over the whole troposphere.

Reply: We corrected it (Please, see p. 4, line 111).

3.2.3 I. 17

The authors have not defined DRF before (they defined ADRF on p. 17588, but not DRF).

Reply: We removed the sentence in the revised manuscript.

3.2.4 I. 18

The authors write [. . .] the accuracy of the bottom-up NO_x emissions. What is the? Which dataset do the authors evaluate?

Reply: In this study, the comparison study was carried out in order to evaluate the performances of the NO_x emissions of INTEX-B, CAPSS, and REAS v1.11 inventories in East Asia. We clarified which emission inventories were evaluated in the revised manuscript (Please, see p. 5, lines 124-125 and p. 2, lines 43-44).

C8344

3.2.5 I. 20

remove also

Reply: We removed “also” (see p. 5, line 127).

3.3 p. 17590

3.3.1 I. 2–3

Tropospheric columns? Total columns?

Reply: We removed the sentence (Please, see p. 5, lines 140-141). We clarified it in the revised manuscript (p. 4, lines 106-107).

3.4 p. 17595

3.4.1 I. 6

It is unclear what the authors mean by December–February 2006. The use of the – implies a range over three consecutive months, but the start of that range (December 2006) is after the end of the range (February 2006). The authors should re-phrase as January, February, and December of 2006 if that's what they mean.

Reply: Thank you for this kind comment. We revised it (Please, see p. 12, line 298).

3.5 p. 17596

3.5.1 I. 1–2

I don't understand why high values would be better for a comparison study.

C8345

Reply: As responded to previous comment, we eliminated the description of “the cold months are better for conducting this study due to the uncertain tropospheric chemistry and faster NO_x loss rate during the summer” and “higher values would be better for a comparison study between CMAQ and OMI-derived NO₂ columns” (Please, refer to Sects. 3.1.1 and 3.2.3).

3.6 p. 17600

3.6.1 I. 21

The authors should specify what exactly they mean by underestimated by a factor of ~0.9. So was the underestimation by 90% or by 10%? This is not clear from the authors' formulation.

Reply: We intended that it is underestimated by ~10%. We removed the sentence in the revised manuscript.

3.7 p. 17604

3.7.1 "geogenic" emissions

The authors repeatedly speak of geogenic emissions. I've never heard this term before; to my knowledge, the term biogenic NO_x emissions is commonly used in the literature for emissions from soils.

Reply: We changed the term, “geogenic NO_x emissions” to “biological NO_x emissions from soils” (see p. 24, lines 587 and 589).

3.8 p. 17606

3.8.1 I. 3

C8346

Whenever the authors write strength of NO_x emission, they should add that this means that they actually use a different emission inventory. From just reading strength of NO_x emissions, the author is lead to wonder what the authors exactly mean. For example, the authors could have scaled the used emission datasets, and the reader is left to guess what the authors want to say.

Reply: In this study, the strength of NO_x emissions means “different emission inventory”. Here, we applied the REAS v1.11 emission inventory over China instead of the INTEX-B emission inventory for sensitivity runs. We clarified the meaning of the strength of NO_x emission in the revised manuscript (Please, see p. 5, lines 132-133, p. 18, lines 459-460, and p. 25, line 623).

3.8.2 I. 17–22

The authors should make a clear statement which N₂O₅ parameterization leads to the best agreement, or which parameterizations lead to bad agreements. As it stands currently, the author cannot tell from the summary alone.

Reply: Based on the sensitivity tests with different reaction probability of N₂O₅ onto aerosols, the NO₂ columns with the Schemes II, III, and IV resulted in the best comparisons with the OMI observations. We stated this in the revised manuscript (Please, see p.26, lines 643-645).

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/14/C8330/2014/acpd-14-C8330-2014-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 17585, 2014.