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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.

Anonymous Referee #1

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The paper *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 Han et al. is a detailed description of a comparison study between modelled and measured NO_2 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 models
3. The influence of the used emission inventory on the model results
4. The influence of the used N_{2O5} 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.

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 NO_2 columns*. Maybe the authors should chose 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.

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

Point 4 is also interesting and valids publication.

1.2 Inference from NO_2 columns to NO_x emissions

Throughout the manuscript, the authors repeatedly do inference from the observed NO_2 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., 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 NO_2 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 chem-

istry (i.e., VOCs), it is impossible to infer directly and quantitatively from measured NO_2 column differences onto inaccuracies in the used NO_x emission databases.

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 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.

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 ist better for comparisons.

2 Specific comments

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.

2.1.2 Abstract, line 10

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

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.

2.1.4 Abstract, line 11

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

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 arguement and should be removed.

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?

2.1.7 Abstract, line 18

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

2.2 Introduction, p. 17589

2.2.1 l. 1

All these studies have been about satellite measurements of tropospheric columnar NO_2, not of mixing ratios of NO_x.

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?

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.

2.4.2 Stratospheric correction

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

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.

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 legent into each of the seven plots.
- in the Figure caption, give reference to Fig. 2 for the region definitions

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.

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

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.

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

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 NOx, 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.

3 Small Corrections

3.1 Introduction, p. 17588

3.1.1 l. 10

in East Asia insted of *in East Asian*

3.1.2 l. 20

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

3.1.3 l. 22

remove *also*

3.1.4 l. 27

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

3.2 Introduction, p. 17589

3.2.1 l. 8

The authors should specify what exactly they mean by $\Omega_N O_2$, i.e., if they refer to to total or tropospheric columns.

3.2.2 l. 11–12

interpreting [. . .] Omega$_{NO2}$ [. . .] near the surface doesn't make any sense, as $\Omega_N O_2$ is a quantity integrated over the whole troposphere.

3.2.3 l. 17

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

3.2.4 l. 18

The authors write *[. . .] the accuracy of **the** bottom-up NO$_x$ emissions*. What is *the*? Which dataset do the authors evaluate?

3.2.5 l. 20

remove *also*

3.3 p. 17590

3.3.1 l. 2–3

Tropospheric columns? Total columns?

3.4 p. 17595

3.4.1 l. 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.

3.5 p. 17596

3.5.1 l. 1–2

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

3.6 p. 17600

3.6.1 l. 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.

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.

3.8 p. 17606

3.8.1 l. 3

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.

3.8.2 l. 17–22

The authors should make a clear statement which N_{2O5} 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.

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