Response to Reviewer #1:

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 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_2O_5 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 NO_2 profiles.

Below is a summary (Table 1) of the comparison studies between satellite-retrieved and CTM-calculated NO_2 columns over "East Asia", but all the studies were conducted "without the applications of the AKs".

Table A1. Several comparison studies over East Asia without the application of AKs

References	Model (CTM/Meteorological Model)	Models used in NO_2 retrieval	Satellite data (algorithm)
Ma et al., 2006	RADM/MM5	MOZART	GOME (Bremen)
Uno et al., 2007	CTM based on CMAQ/RAMS	MOZART	GOME (Bremen)
He et al., 2007	CTM based on CMAQ/RAMS	MOZART (Richter et al., 2005)	GOME & SCIAMAHCY (Bremen)
Shi et al., 2008	CMAQ/MM5	Described ambiguously	SCIAMACHY (KNMI)
Han et al., 2009	CMAQ/MM5	MOZART	GOME (Bremen)
Han et al., 2011	CMAQ/MM5	TM4	OMI (KNMI)

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 (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 611-614).

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 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 440-445 and 448-452).

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 chemistry (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.

Reply: Reviewer's point is definitely right! Although the CMAQ-calculated NO_2 columns were, on annual average, ~28% (in terms of Normalized Mean Bias) smaller than the OMI NO_2 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 52-54 and p. 25, lines 615-617).

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.

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 ist 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 NO_2 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 NO_2 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 NO_2 columns" (Please, refer to Sects. 3.1.1 and 3.2.3).

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.

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 611-615). We intended that the NMEs between the $\Omega_{CMAQ,AK}$ and Ω_{OMI} (AKs applied) decreased, for example, from ~98% to ~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 NO_2 column" by "two tropospheric NO_2 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

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 arguement 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: We rewrote many parts in Abstract. The statement was clarified in the revised manuscript. 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 162-164). 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. 17, lines 420-424).

2.1.7 Abstract, line 18
I don't understand the difference between different monthly variation and different strengths of the NO_x emissions.
Reply: We rewrote many parts in Abstract. In the revised manuscript, we tried to clarify it to remove this ambiguity (Please, see p. 17, lines 420-424, and p. 18, lines 455-458).

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.
Reply: We changed 'NO_x' to 'column NO_2' (Please, see p. 4, lines 94 and 99).

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 $30\times30km^2$, 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 147-151 and 151-153).

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 253-263). 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 $13\times24km^2$ at nadir, because towards the edges of the scan, the spatial resolution becomes significantly lower.

Reply: We corrected it (see p. 8, line 203).

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. 8, lines 213-214).

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 220-221).

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
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 NO_2 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 NO_2 columns are greatly larger than OMI NO_2 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 313).

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.
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 NO_2 is larger than OMI NO_2, 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 NO_2 columns (< 1×10^{16} molecules cm^{-2}) in Fig. 6, most data are scattered above the 1:1 line. In contrast, in the large values of NO_2 columns (>1×10^{16} molecules cm^{-2}), most data are scattered below the 1:1 line. We believe that the large values of the OMI NO_2 columns made the low regression slop (<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
- Add a note to the Figure that red and blue colors indicate under and overestimation of the actual NO_2 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 poor agreements (Please, see p.15, lines 375-376).

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 454-455).

3 Small Corrections
3.1 Introduction, p. 17588
3.1.1 l. 10
in East Asia insted of in East Asian
Reply: We corrected it (Please, see p. 3, line 78).

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.

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, lines 88-90).

3.1.3 l. 22
remove also
Reply: We removed it (see p. 4, line 91).

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.
Reply: We added those references in the revised manuscript (Please, see p. 4, line 97).

3.2 Introduction, p. 17589
3.2.1 l. 8
The authors should specify what exactly they mean by Ω_{NO2}, i.e., if they refer to to total or tropospheric columns.
Reply: We clarified the definition of Ω that indicates "tropospheric NO_2 vertical columns" from CTM simulations and satellite observations (Please, see p. 4, line 105-106).

3.2.2 l. 11–12
interpreting [. . .] Ω_{NO2} [. . .] near the surface doesn't make any sense, as Ω_{NO2} is a quantity integrated over the whole troposphere.
Reply: We corrected it (Please, see p. 4, lines 109-111).

3.2.3 l. 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 l. 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 122-123 and p. 2, lines 43-44).

3.2.5 l. 20
remove also
Reply: We removed "also" (see p. 5, lines 125-127).

3.3 p. 17590
3.3.1 l. 2–3
Tropospheric columns? Total columns?
Reply: We removed the sentence (Please, see p. 5, lines 138-139). We clarified it in the revised manuscript (p. 4, lines 105-106).

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.
Reply: Thank you for this kind comment. We revised it (Please, see p. 12, line 294).

3.5 p. 17596
3.5.1 l. 1–2
I don't understand why high values would be better for a comparison study.
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 NO_2 columns" (Please, refer to Sects. 3.1.1 and 3.2.3).

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.
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. 23, lines 584 and 586).

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.
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 130-131, p. 18, lines 457-458, and p. 25, line 620).

3.8.2 l. 17–22

The authors should make a clear statement which N_2O_5 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 N_2O_5 onto aerosols, the NO_2 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 640-642).

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Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131-5153, doi:10.5194/acp-9-5131-2009, 2009.

Response to Reviewer #2:

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 manuscript appears to suggest that applying averaging kernel in the application of satellite observed NO_2 columns is so new that it warrants a journal publication (with which I cannot agree). It seems that a main justification is in line 10-14 on P. 17596, "However, such a comparison without applying the AKs is like comparing apples and oranges, and is not reasonable. Such studies have been conducted over East Asia, with misleading conclusions (e.g. Ma et al., 2006; He et al., 2007; Uno et al., 2007; Shi et al., 2008; Han et al., 2009, 2011)." I strongly suggest that the authors read the papers from the other groups carefully before stating that those papers erred. Among the cited papers, Uno et al. (2007), for example, compared the retrieved tropospheric vertical columns with the model simulations, which is very appropriate. The authors did not seem to know that AK has already been used in the retrieval of the vertical columns.

In line 6-10 on P. 17596, the authors stated "Previously, Han et al. (2009, 2011) also compared the CMAQ-calculated NO_2 columns with satellite-retrieved NO_2 columns, without using the AKs, to investigate the accuracy of bottom-up NO_x emissions over East Asia. Based on this comparison, Han et al. (2011) concluded that the bottom-up NO_x emissions used in the CTM simulation over East Asia could be overestimated." While it is common knowledge that the AK-type observation sensitivity corrections on satellite data are absolutely needed, if the authors were using retrieved tropospheric NO_2 vertical columns to compare to model simulated columns, it is OK. (The profile error is another matter.)

I strongly suggest that the authors read carefully the early papers by Martin and coauthors to understand the difference between slant and vertical tropospheric NO_2 columns and where AK was used in the retrieval. It seems that the concept of tropospheric NO_2 vertical column retrieval was misunderstood. Another possibility is that the paper suggests that AK should be used when comparing to satellite-derived tropospheric slant columns, which seems rather obvious and there is no need to write a journal paper for that.

Reply: We have believed that we understood the processes of the NO_2 retrieval and the use of averaging kernels (AKs). We do not know which Martin et al.' paper reviewer mentioned. But, it is obvious that if true/real NO_2 profiles are not used in the DOAS NO_2 retrieval process, the AKs should be applied to correct the large systematic errors caused by the unrealistic *a priori* assumption on the NO_2 profiles (Eskes and Boersma, 2003). Clearly, it is almost impossible that the true NO_2 vertical profiles are used in the tropospheric NO_2 column

retrieval process, since they are always changing in time and space. This is the usefulness of the applications of the AKs. Unfortunately, we did not apply the AKs in our previous study (Han et al., 2011). Thus, we wish to correct our previous conclusions, applying the AKs to the CMAQ model simulations in this study. In the KNMI/DOMINO v2.0 NO_2 column products, the AMF were calculated using *a priori* NO_2 profiles obtained from global TM4 model simulations, which are obviously not "true profiles", but the profiles from global CTM simulations with coarse resolution (2°×3°). We therefore have to apply the AKs to our study. The applications of the AKs in our study have been fully confirmed with other colleagues (recent personal communications with Drs. H. Eskes and K. F. Boersma at KNMI; Prof. R. Cohen at UC Berkeley).

Reviewer also mentioned that Uno et al.'s work (2007) was correct. In fact, Uno's group published their recent work, fully collaborating with "our lab" (actually, we gave them several suggestions to use the AKs). In their recent paper, they also applied the SCIAMACHY-derived AKs to their CMAQ model simulations and they compared them with the SCIAMACHY-retrieved NO_2 columns. Please, refer to Itahashi et al. (2014).

There are recent publications in which the AKs have been applied. We leave these examples for your references (Herron-Thorpe et al., 2010; Lamsal et al., 2010; Huijnen et al., 2010; Ghude et al., 2013; Zyrichidou et al., 2013).

In addition, if reviewer tried to mention Martin et al. (2002) and Palmer et al. (2001), please, take a look at p. 1286, lines 3-12 and p. 1290, lines 7-16 of Eskes and Boersma (2003). As used in our study, applying the AKs can provide an alternative way to Palmer et al. (2001)'s and Martin et al. (2002)'s works. Even, Martin's group has recently used the AKs (please, refer to Lamsal et al., 2010; supplementary materials of Kharol et al., 2013).

As a side note, the usage of English can be improved in this manuscript. "Accuracy" has a well-defined meaning in science. I don't think that a comparison between satellite and model columns can be used to evaluate the accuracy of NO_x emissions (as stated in the title).
Reply: Thank you for your comment. Considering reviewers' comments, we changed the title.

In the abstract, AKs cannot be retrieved from a retrieval algorithm. On P. 17594, the wording of "under-sensitive" and "over-sensitive" should be changed.
Reply: Indeed, the AKs can be provided by retrieval and instrument groups without direct involvement of 3D-CTMs (Eskes and Boersma, 2003). Regarding "over- and under-sensitivity", we changed the words (see p. 10, lines 246-248).

In addition, the statement on P. 17595 "In the same context, more attention was paid to winter (spring and fall) in this study, because there are fewer uncertainties and unknowns related to the chemical NO_x loss rates during these seasons." is incorrect. Although the chemical effect is less in winter (one can even argue that the uncertainty of NO_x chemistry in winter is larger), the transport uncertainty is much larger in winter than summer. To understand emissions, winter is not a better season to use satellite measurements than summer. The MM5-CMAQ (4.7.1) modeling system is getting long in the tooth. The authors should consider updating the modeling system.

Reply: During summer, the NO_x loss rates are so fast that the considerations of additional NO_x emissions would hardly change the CTM-calculated NO_2 columns (Boersma et al., 2009; Han et al., 2009). Therefore, it is difficult to evaluate NO_x emissions using a comparison between the CTM-derived and satellite-derived NO_2 columns "during summer". This is what we wish to say here! Also, as reviewers pointed out, there are other uncertainties related to the issues of pollutant transport and satellite errors during winter. However, we are sure that summer is not a better season for this comparison study. It may be a controversial issue. We thus eliminated that "the cold season are better for conducting this kind of comparison study" in the revised manuscript (Please, refer to Sects. 3.1.1 and 3.2.3).

In the modeling system, the MM5/CMAQ v4.7.1 is somewhat old, but still viable (although we are currently using WRF model). However, in this study, four-dimensional data assimilation (FDDA) using observation data set was carried out in order to prepare more accurate meteorological fields. The meteorological fields were used and evaluated in our previous study (Park et al., 2011). Also, in the CTM, we modified the SAPRC-99 mechanism to consider the OH recycling in isoprene chemistry which is a hot issue. It was successfully evaluated and applied in our another previous study (Han et al., 2013).

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Response to Reviewer #3:

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 NO_2 tropospheric columns with satellite retrievals. (2) the sensitivity of the modeled NO_2 columns to simulation parameters (seasonal cycle; alternative emission inventory; reaction rate N_2O_5). 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 NO_2 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.

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 NO_x 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 NO_2 columns (please, see p. 5, lines 117 – 125).

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 NO_2 retrievals in Section 2.2. The uncertainty for the tropospheric NO_2 columns of the KNMI/DOMINO v2.0 used in this study is approximately 1.0×10^{15} molecule cm^{-2} 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 NO_2 columns via seasonal averaging and applying the AKs to the CMAQ model simulations, respectively. Please, check out p. 9, lines 226-230 and p. 11, lines 264-280 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 were discussed in Sect. 2.2 (please, see p. 10, lines 253-263). 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 NO_x 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 NO_2 columns, AKs, CMAQ model simulations, etc. The CMAQ-calculated NO_2 columns were, on annual average, ~28% (in terms of "Normalized Mean Bias") underestimated, compared with the OMI NO_2 columns. We clarified this point in the revised manuscript (Please, see p. 2, lines 52-54).

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 81-83).

1, P17588, line 4-6: To compensate for height dependent sensitivities, the column retrieval algorithm depends on cloud information and an assumed NO_2 profile. If this NO_2 profile reflects reality, the retrievals can be compared directly with simulations (and are in that sense "real" or "true"). If the true NO_2 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 NO_2 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 102-109).

1, P17589, line 9. Consider a definition Ω instead of Ω_{NO2} throughout the whole paper, as NO_2 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 manuscript.

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

Reply: We removed it (Please, see p. 5, line138).

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 170-171).

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

Reply: We changed it (p. 8, line 190).

2.1, P17593, line 20: Change $\hat{x} - x_a$ to $\hat{x} - \hat{x}_a$, to differentiate $\hat{x}_a$ being a column quantity and x_a a vector quantity.

Reply: We corrected it in Equation (2) (p. 10, line 239).

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 221-223 and p. 10, line 251-252).

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, line 291).

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 308-310).

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. 5, lines 117 – 122).

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 644-648).

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

Reply: We put the following sentence into the text. "The NME was defined in Table A1" (see p. 14 line 341).

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 km^2 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 NO_2 columns, mainly because averaging data can reduce random errors in the satellite-derived tropospheric NO_2 columns. For a better understanding, we conducted the scatter plot analysis using daily data set (number of data from ~300000 to ~ 500000 for the entire domain) over the entire domain in Fig. S1. (Please, refer to Fig. S1 and see p. 14, lines 357-362).

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 680-706).

3.1.2, P17598, line 6: "between 2×10^{15}" → "between -2×10^{15}"

Reply: We added the negative sign and gave specific values in both sides (Please, see p. 15, line 383).

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. $\Omega_{CMAQ,AK}$ is smaller than Ω_{OMI}, a strong indication that the used NO_x emission inventory is underestimating the real emissions.

Reply: Yes, it is. The negative values in the MBs indicate that the NO_x emissions are possibly underestimated, compared to the real NO_x emissions. We clarified this point (Please, see p. 15, line 383-385).

3.1.2, P17598, line 17-19: In the abstract and conclusion the underestimation of NO_x 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 NO_2 columns were, on annual average, ~28% (in terms of the Normalized Mean Bias) underestimated, compared with the OMI NO_2 columns (Please, see p. 2 lines 52-54, p. 16, lines 390-391, and p. 25, lines 615-617).

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 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 573-582). The reasons to choose the sensitivity parameters for Cases 2, 3 and 4 were also explained /described at p. 16, lines 404-416.

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 164-165).

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

Reply: In addition to the issues of NO_x emission (i.e. monthly factor and different emission inventory), the NO_x 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 N_2O_5 onto aerosols which is one of the most important parameters for determining the NO_x lifetimes during winter. Yes, the cold environments create a favorable condition for high levels of N_2O_5.

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 440).

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 454-455).

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 NO_x 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 NO_2 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 $\Omega_{CMAQ,AK}$ and Ω_{OMI}. However, in the revised manuscript, we excluded this issue in Sect. 3.2.2, because the top-down NO_x 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 NO_x 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 NO_x 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. 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. 21, lines 537-540).

3.2.4, P17603, line 26: How do k_1, k_2, k_3, and k_4 relate in magnitude? This gives information about to which mixing ratio the balance between NO_2 and NO is especially sensitive.
Reply: We calculated the reaction rate constants at 298K, and put some discussions at p. 22, lines 549-557.

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 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 $\times 10^{15}$ molecules cm^{-2}) 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 NO_2 retrieval in Sect. 2.2. The errors in the NO_2 retrieval can be caused by the calculations of the AMF, spectral fitting, and stratospheric slant NO_2 columns. The uncertainty for the tropospheric NO_2 columns of the KNMI/DOMINO v2.0 used in this study is 1.0×10^{15} molecule cm^{-2} with a 25% relative error (Boersma et al., 2011). Please, see p. 9, lines 226-230 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 NO_2 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 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 × 5 panel
Reply: As reviewer pointed out, we merged two figures into a 4 × 5 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 1110).

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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