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Atmospheric Chemistry and Physics Discussions

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

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

- Change of the title.

- Less emphasis on applying AKs to CMAQ model simulations. - Restructure of the manuscript to clarify our motivations and conclusions of this study.

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

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

The manuscript appears to suggest that applying averaging kernel in the application of satellite observed NO2 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 NO2 columns with satellite-retrieved NO2 columns, without using the AKs, to investigate the accuracy of bottom-up NOx emissions over East Asia. Based on this comparison, Han et al. (2011) concluded that the bottom-up NOx 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 NO2 vertical columns to compare to model simulated columns, it is OK.

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(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 NO2 columns and where AK was used in the retrieval. It seems that the concept of tropospheric NO2 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 NO2 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 NO2 profiles are not used in the DOAS NO2 retrieval process, the AKs should be applied to correct the large systematic errors caused by the unrealistic a priori assumption on the NO2 profiles (Eskes and Boersma, 2003). Clearly, it is almost impossible that the true NO2 vertical profiles are used in the tropospheric NO2 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 NO2 column products, the AMF were calculated using a priori NO2 profiles obtained from global TM4 model simulations, which are obviously not "true profiles", but the profiles from global CTM simulations with coarse resolution ($2^\circ \times 3^\circ$). 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

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with the SCIAMACHY-retrieved NO2 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 NOx 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 250-252).

In addition, the statement on P. 17595 "In the same context, more attention was paid

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to winter (spring and fall) in this study, because there are fewer uncertainties and unknowns related to the chemical NOx loss rates during these seasons." is incorrect. Although the chemical effect is less in winter (one can even argue that the uncertainty of NOx 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 NOx loss rates are so fast that the considerations of additional NOx emissions would hardly change the CTM-calculated NO2 columns (Boersma et al., 2009; Han et al., 2009). Therefore, it is difficult to evaluate NOx emissions using a comparison between the CTM-derived and satellite-derived NO2 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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