

Referee #1

This paper combines multiple satellite retrievals of tropospheric NO₂, multiyear simulations with a relatively high resolution model, and an updated emission inventory to analyze the trends and variability of NO_x over East Asia. It also tries to invert NO_x emissions for 2009-2010 when the bottom-up inventory is not available. Overall I find the paper is within the scope of ACP. There are some issues to be addressed before the paper can be published in ACP.

Some results on emission trends and variability have been found in previous studies (e.g., Schneider and Van der A., 2012, JGR; Lin and McElroy, 2011, ACP; Lamsal et al., 2011, GRL; Stavrakou et al., 2008, GRL). I suggest the authors to make it clearer (especially in the introduction) how the current study improves upon the previous ones, in addition to the use of different models and emission inventories. Quantitative analysis should be included in the introduction.

I find the multiyear simulations on a relatively high resolution to be an important aspect that could be addressed in the introduction.

Model uncertainties could be discussed more thoroughly and quantitatively (if possible). Such discussions should be placed when introducing model simulations in Sect. 2.1. While this paper focuses on emission trends, an improved discussion of model settings and model uncertainty would be very beneficial.

Lightning emissions are not included, so what are the implications for model VCDs and comparisons with satellite retrieval? Adding lightning emissions would lead to more VCDs particularly in summer.

Model schemes should be clarified and citations provided. The sentence 'CMAQ was configured to use mass-conserving scheme for advection, multiscale horizontal diffusion and eddy vertical diffusion.' does not tell much.

Month variation in emissions should be presented. How about the diurnal variation of emissions in the model? This may affect the comparison results for morning time and afternoon time.

A table listing anthropogenic and natural emissions in the model will be very useful.

Are model results sampled at time and places with valid satellite retrievals? Sampling bias is important. Please also give detailed info on how the satellite pixels are gridded to 0.5 degree.

Errors in satellite retrievals should be addressed more consistently. In Sect. 2.2, satellite retrieval errors are assumed to be relatively small, which are in contrast with later sections suggesting satellite biases to be larger. Estimate of satellite errors should be made consistent, placed in Sect. 2.2, and referred to in later sections.

Results for Japan and Korea may be affected by the small domain such that the effect of model and retrieval errors may be larger (because fewer data can be used to do spatial averaging, as compared to CEC). Please discuss.

Specific comments

1) Abstract: Quantitative analyses will be useful.

We have revised the abstract to be quantitative (Please also see the comment made by Referee #2-2).

2) P11249, L20: transportation and power plants belong to ‘fossil fuel combustion’

We have removed the terms ‘transportation’ and ‘power plants’.

3) P11250, L7: If cloud cover is considered, more days are needed for global coverage

We have corrected this mistake.

4) P11251, 2nd paragraph: the paragraph is complicated. I suggest to split it. Also, 'fully utilizing' is just 'utilizing'

We have split this paragraph into two parts, and we have removed the word 'fully'.

5) Sect. 2.2. Use of averaging kernel will affect the trend analysis (e.g., Lin and McElroy, 2011).

This point has been further discussed including a comment on the discrepancies between model and satellite observations during wintertime, cited by Lin and McElroy (2011).

6) P11254, L2: unclear. Is '30-35%' found in the present study or quoted from other papers?

This sentence refers to the findings of previous studies. We have changed the citations to make this clear.

7) P11254, L14-18: Lin and McElroy (2011) first analyzed the downturn, and should be cited here.

We have cited Lin and McElroy (2011) in this line.

8) P11254, L25: the differences may also be caused by larger biases in the morning time for both modeling (due to model errors at night) and satellite (due to less valid data from SCIAMACHY, etc.).

We have added these comments.

9) P11256, L4: the satellite errors suggested here are inconsistent with Sect. 2.2.

We have added the reason for discrepancies between the model and satellite observations during winter as averaging kernel information, as cited by Lin and McElroy (2011).

10) P11256, 1st paragraph: the discussions on model and retrieval errors should be placed in Sect.2.

We would like to discuss these model and retrieval errors along with Fig. 1.

11) P11257, L1: 5.1%/yr is smaller than Lin and McElroy (2011) (8-9%/yr). Please discuss.

In the Lin and McElroy (2011) study, the trend of NO₂ VCD prior to the downturn was analyzed, and their results indicated growth of 8–9%/year. In our analysis, we have included the late-2008 period; hence, the growth rate is smaller than that in the Lin and McElroy (2011) study. We have reanalyzed the trend of NO₂ VCD before the economic downturn and revised our manuscript according to the results.

12) P11257, L20: The sentence is an overstatement, particularly for Japan and Korea.

We have removed the word ‘closely’ and replaced it with ‘generally.’

13) P11258, L1: SCIAMACHY is subject to much less valid data, and the comparison in the morning time is subject to model errors in the nighttime. Please comment.

We have now commented on this in the revised text. SCIAMACHY would be subject to much less valid data; however, as reported by Schneider and van der A (2012), the potential uncertainty associated with the combination of different datasets with differences in spatial resolution, sensor calibration, local overpass time, and retrieval algorithms is avoided by using only the SCIAMACHY data. Moreover, we used the annual mean NO₂ VCD in this inverse approach; hence, the uncertainty in the nighttime would be negligible because the model

uncertainty in the nighttime would be larger in wintertime. We confirmed that the inverse estimation based on summertime mean NO₂ VCD and wintertime mean NO₂ VCD does not differ much in this estimation based on the annual mean NO₂ VCD.

14) P11258, L11: the sentence 'because of ...' is unclear

We have removed the wording 'because of' and replaced it with 'on the basis of.' We have also revised the sentences describing our inversion results. Please also see the reply to your comments 15) and 16).

15) P11258, L18: 'the results in...' is unclear.

We have removed this sentence.

16) P11258, L18-21: the estimate should be presented with caution. The fraction of CEC to China may be changed from one year to another due to the different growth rates of NO_x emissions in different regions. Also, what is the source of '46%'?

The source of the ratio of CEC to total China (46%) is the updated REAS emission inventory used in our modeling system. As noted, the value of this ratio varied from 44.4% to 47.2%; therefore, we have reanalyzed these data and revised our manuscript accordingly.

17) P11258, L22: source of '14.3Tg'?

We have revised this value to '12.7 Tg' and cited the updated REAS inventory in (Kurokawa et al., 2013).

18) P11259, 2nd paragraph: the general discussion on China should cite other studies.

We have cited Lu et al. (2010) in this line.

19) P11259, L16: 'it is considered...variability' is unclear

We have revised this sentence.

20) P11259, L23-26: please cite previous studies on NOx trends and economic downturn.

We have cited Lin and McElroy (2011) in this line and revised the text accordingly.

21) Table 1: Pixel sizes and periods are presented incorrect/incomplete.

We have corrected the mistake.

Referee #2

This study analyzed tropospheric NO₂ trends over East Asia during 2000-2010, using observations from satellite instruments and regional model simulations with an updated emission inventory. Understanding trends in Asian NO_x emission during the past 20 years or so and the cause of biases between the bottom up emission inventory and satellite retrievals is an active research area. I believe that the most valuable aspect of this study is conducting regional-scale, full-chemistry model simulations at 80x80 km horizontal resolution for a decade. The authors could have taken advantage of this unique model experiments and conduct in-depth analysis. This study is certainly within the scope of ACP but the manuscript needs to go through major revisions before it can be published in ACP.

Major limitations of this work include: 1) Most discussions on the uncertainty of satellite measurements and model results in the current version of the manuscript are vague, i.e. mostly repeating the points already addressed in previous work and lacking a clear focus relevant to this particular model (CMAQ). 2) Different trends in morning (GOME and SCIAMACHY) versus afternoon (OMI) measurements (Page 11256 and 11257) need to be better visualized in the figure and discussed in light of the role of emissions versus lifetime. Does the model reproduce the morning versus afternoon difference? 3) The major limitation of the model set up is the exclusion of lightning NO_x emissions, which may contribute a substantial portion to tropospheric column NO₂ in some regions over East Asia. 4) Averaging kernel has not been applied in the current model evaluation. Will the application of averaging kernel reduce the wintertime biases ?

1) Title: The title of the manuscript need be concise and as short as possible. I think the current title of manuscript is really too long and somewhat confusing.

Suggested title: Tropospheric NO₂ trends over East Asia during 2000-2010: satellite observations and regional model analysis

We have revised the title as follows, "Tropospheric NO₂ trends over East Asia during 2000–2010: multi-satellite observations and a regional model analysis"

2) Abstract: Discussion of the trends needs to be quantitative. It would be much stronger if you add numbers (e.g. xx% per year) after "Rapid growth" and "Slightly decreasing"

We have revised the abstract to include a quantitative discussion.

3) Line 14, Page 11250: "Temporal variation, and interannual trends of NO_x emissions" is awkward because temporal variation on daily to seasonal time scales in NO₂ VCD can reflect changes in NO₂ lifetime rather than in emissions. Maybe revised to "long-term trends of NO_x emissions"?

We have revised this sentence according to your suggestion.

4) Line 23, Page 11250, "the anthropogenic emissions would have underestimate the growth in NO_x emissions " is odd. Change "the anthropogenic emissions" to "the bottom up emission inventory"?

We have revised the wording of this sentence.

5) Line 25, Page 11255, "the model reproduced the temporal variation in ...". Do you mean "spatial variation"?

We have revised this wording and cited the number of the relevant Figure, as requested in your comment (9).

6) Line 7-10, Page 11256, Where do you see this?

This is from the results presented in our companion paper. We have cited the paper on this line (Irie et al., 2013).

7) Line 15-18, Page 11257, Clarify what exactly Han et al (2009) found by comparing multiple emission inventory?

We have included a discussion of this point.

8) Table 1: the time period and pixel size of OMI and GOME2 are wrong.

We have corrected the mistake.

9) Figures: Throughout the discussion, it is difficult to follow which figure you are referring to in the text. The panels in the figures need to be clearly labeled using alphabet letters and referred in the text.

We have revised the text to clearly refer to the number of the figure being discussed.

10) Figure X: Wouldn't the NO₂ variability during 2000-2003 for Korea is an artifact of measurement noise due to a small sampling domain?

We have checked the pixels available during 2000–2003 that were obtained by GOME; however, no artifacts due to measurement noise were found.

11) Can you also discuss how emission trends in the updated REAS inventory compare with trends in other inventories? This can be very helpful to those who are interested in applying the REAS inventory in model simulations.

We have discussed the comparison of inventories before subsection 3.2.