Global high-resolution simulations of tropospheric nitrogen dioxide using CHASERV4.0: Response to reviewer #1

We would like to thank anonymous reviewer #1 for his or her careful reading and valuable comments, which have helped to significantly improve the manuscript. We revised the manuscript and responded to the reviewer's comments. The main changes are as follows:

1) Validation results of meteorological fields have been extended and moved to Section 3
2) An analysis of the impacts of convection and lightning NO_x has been added to Section 5.
3) An extended discussion has been added on the trade-off between horizontal model resolution and computational costs.

Individual comments (in black) and specific responses to them (in blue) are listed below. *Text (Italicized)* from the revised manuscript is in quotes.

This manuscript describes results from a decent study of the impact of horizontal resolution on model simulations, with focus on NO2 evaluated against mainly satellite observations. It illustrates the gain in performance when moving from 2.8° towards 1.1° and 0.56°, showing on global scale a relatively limited improvement in performance. Nevertheless, on a local scale generally significantly improved performance was shown mostly for the 1.1° vs the 2.8° resolution model experiments. A difficulty encountered in this system is that not only chemistry changes, but also the meteorology changes in this online system, as documented briefly by the authors. A more detailed analysis of differences (e.g.: to what extend are photolysis rates different on a high-resolution model run compared to a reference run, e.g. due to differences in clouds) would be interesting, although I can see that this may be beyond the scope of the current manuscript.

To discuss the impacts on meteorological fields more intensively, we have added validation results of outgoing longwave radiation (OLR) in Figure 1. The relevant discussion in

Section 3 has been expanded in the revised manuscript as follows:

(p. 7, l. 32–p. 8, l. 3)

"*The global mean positive bias was 80% and 50% lower at 1.1° and 0.56° resolutions, respectively, than at 2.8° resolution (Figures 1e–h), suggesting improved photolysis calculations in the high-resolution simulations. Among different regions, the positive model bias at 2.8° resolution was largest over the Maritime continent, which was reduced by 86% at 1.1° resolution and by 75% at 0.56° resolution. Over northern South America, in contrast, most of the positive biases remain at 1.1° and 0.56° resolutions.* "

Also it was shown that in particular for the O3-HOx-NOx chemistry the resolution makes a difference, considering that with NOx confined in smaller grid boxes, leads to an overall reduced efficiency in ozone chemical production, but increased stratosphere-troposphere exchange. Whereas the authors focus mainly on changes in NOx chemistry over megacity and biomass burning regions, I miss a more detailed analysis of effect of lightning NOx emissions as applied on different spatial resolution: Is it correct that with higher horizontal resolution the lightning NOx emissions will result in less efficient ozone production, and would simulations suggest that a retuning of total NOx emissions (apart from uncertainties in profile shape) may be necessary?

To discuss the impacts of convection and lightning NO_x on NO_2 and O_3 chemical production, we have added Figure 14 and the following discussions:

(p. 15, l. 34–p. 16, l. 13)

"*Figure 14 shows the spatial distributions of NO_2 partial column in the free troposphere, convective cloud updraft mass flux at 500 hPa, and vertically integrated lightning NO_x production. The simulated NO_2 partial column in the free troposphere was smaller by 17% at 1.1° resolution and by 14% at 0.56° resolution than at 2.8° resolution over the northern subtropics and midlatitudes, primarily because of smaller NO_2 concentrations above 400 hPa. These changes in the free tropospheric NO_2 were in contrast to the changes in the lower tropospheric NO_2, which were associated with suppressed* *convective cloud updraft over the continents by up to 76% at 1.1° and 0.56° resolutions over the northern subtropics and mid-latitudes. In contrast, over the Maritime continent, South America, and Central Africa, the free tropospheric NO_2 column was larger at 1.1° resolution by up to 18% and at 0.56° resolution by up to 20% than at 2.8° resolution, primarily reflecting increased NO_2 concentration between 600—800 hPa. Lightning NO_x productions are also largely different between the simulations in the tropics. Over the tropics, although the mean convective cloud updraft was weaker at 1.1° and 0.56° resolutions than at 2.8° resolution, the high resolution simulations revealed increased ice cloud in the upper troposphere and stronger (but less frequent) convection, thus increasing lightning NO_x sources especially over Asia. Meanwhile, given the same amount of lightning NO_x production (using a commonly prescribed lightning NO_x field in all the simulations), the high-resolution simulations revealed a slightly smaller ozone chemical production (by 1%) through representation of local high-concentrated NO_x plumes in July 2008 (figure not shown).*"

To obtain a reasonable lightning NO_x, we optimized the cumulus convection parameterization at each resolution to match with observed lightning flash rate, OLR, and precipitation rate based on sensitivity calculations, without applying any adjustment factors to the global lightning NO_x source amount directly.

P 9. L17: the authors relate the larger negative biases in comparison to GOME-2 observations than to OMI to difficulties in the model to capture the nocturnal thin boundary layers, associated to vertical resolution. Indeed, the number of vertical model layers is relatively small (32), but still I wonder if authors can substantiate this conclusion. Couldn't there be other reasons (missing chemistry, uncertainties in diurnal cycle in emissions, biases between OMI and GOME-2?) that could explain the discrepancies seen?

The vertical model resolution is considered to be insufficient to reproduce a thin nocturnal PBL. At the same time, as suggested by the reviewer, other factors could also contribute to the model bias. The sentences have been rewritten as follows:

(p. 10, l. 6–9)

"*The differences suggest that all model simulations underestimated high NO_2 concentrations in the morning. The underestimations could be associated with insufficient vertical model resolution for capturing thin nocturnal boundary layers, as well as uncertainties in HO_x-NO_x-CO-VOCs chemistry, NO_2 photolysis rates, and emission diurnal cycles.*"

With regard to the biases between OMI and GOME-2, the following sentence has been added:

(p. 10, l. 9–11)

"*The different model biases with respect to OMI and GOME-2 could also be attributed to the bias between these retrievals. Irie et al. (2012) concluded that the bias between these retrievals is small and insignificant for East Asia, whereas the bias between these retrievals is unclear for other regions.*"

In my opinion the Discussion section is a bit on the long side, and contains elements that may fit better in the introduction, mainly sec. 5.3 and 5.4. Also authors state in P17, L17 that high-resolution CTM's will be able to assimilate observations at nearly measurement resolution. I believe this is too optimistic, at least for global CTM's, considering the horizontal resolution of TROPOMI observations.

Some text in Sections 6.3 and 6.4 has been removed or moved to the introduction. Meanwhile, Sections 6.3 and 6.4 have been combined to reduce the text length as follows:

(p. 18, l, 26–p. 19, l. 13)

"*6.3 Application for satellite retrieval and data assimilation*

An important application of high-resolution tropospheric NO_2 simulations is to provide a priori profile information on satellite retrieval and chemical data assimilation (Liu et al., *2017). Here, we would like to discuss the potentials of the obtained results for these applications.*

Current satellite retrievals of the tropospheric NO_2 column use a priori NO_2 profiles obtained from global model simulations at relatively coarse resolutions: from TM5 at 3° × 2° in DOMINO-2 (Boersma et al., 2011) and GEOS-Chem at 2.5° × 2° in OMNO2 (Bucsela et al., 2006; Celarier et al., 2008), whereas the TROPOMI retrieval product will employ 1° × 1° resolution simulation fields from TM5 (Williams et al., 2017). To provide high-resolution (ranging from 4 km to 50 km) a priori information, several regional retrievals have employed regional models (Heckel et al., 2011; Russell et al., 2011; Lin et al., 2014), showing improvements in the retrieved fields in comparison to independent observations. High-resolution a priori fields from global CTMs are important in providing consistent global datasets.

To avoid spatial representation gaps between satellite measurements and coarse-resolution global models, super-observation techniques have been employed to produce representative data before assimilation (e.g., Miyazaki et al., 2012). The average of averaging kernel over a number of retrievals within a super observation grid does not hold any physical meaning. This may inhibit effective improvement by assimilating over regions with varying conditions. High-resolution CTMs allow assimilation of satellite measurements, with reduced representation gaps without any averages.

Because of distinct non-linearity in chemical reactions, high-resolution assimilation of satellite measurements, considering small-scale variations in background error covariance, would be essential in making the best use of observational information. High-resolution chemical data assimilation could also benefit air pollutant emission estimates (e.g., Miyazaki et al., 2014, 2017, Liu et al., 2017), especially using high-resolution measurements from future satellite missions such as TROPOMI and geostationary satellites (e.g., Sentinel-4, GEMS, TEMPO), even when model resolution is still coarser than measurement resolution through improved model processes and spatial representativeness for megacities as demonstrated by this study."

The relevant descriptions in Section 1 have been expanded as follows.

(p. 3, l. 6–16)

"*The authors demonstrated improvements in these regional retrievals using high-resolution a priori fields in comparison to the ARCTRAS aircraft observation and ground-based remote sensing MAX-DOAS through, for instance, clearer separation of* NO_2 *profiles between urban, rural, and ocean regions, and improved representations of altitude-dependent sensitivities (i.e., averaging kernels).*

Global chemical data assimilation (e.g., Inness et al., 2015; Miyazaki et al., 2015) and emission inversion (e.g., Stavrakou et al., 2013; Miyazaki et al., 2017) would also benefit from high-resolution global CTMs, through improvements in model performance (e.g., Arellano Jr. et al., 2007) and reduced spatial representation gaps between observed and simulated fields. Several previous studies (Mijling and van der A, 2012; Ding et al., 2017b; Liu et al., 2017) demonstrated the importance of high-resolution modeling in detecting small-scale NO_x *emission sources such as urban, new power plants, and ship emissions. A systematic evaluation of high-resolution model enables us to discuss application potentials of global high-resolution models to satellite retrievals and data assimilation.*"

P4, L10: A nudging to 12-hourly ERA-Interim re-analysis data is applied. Here I wonder why the authors don't use 6-hourly EI data. Are authors convinced that 12-hourly nudging is sufficiently accurate?

We have evaluated the impact of changing nudging data interval, and found that the annual RMSE of tropospheric NO_2 column against OMI retrievals differed by less than 6% between 1.1° resolution simulations with 12- and 6-hourly reanalysis over most regions. By using 12-hourly nudging interval, the model performance did not significantly worsen, whereas the computational costs (data processing and input) were reduced. Therefore, we employed a nudging to 12-hourly reanalysis.

Technical comments:

P2, L18: Suggest to change to: “High-resolution simulations can lead to improvements in two ways:”

Changed as suggested.

P3, L31: suggest to change to “…deposition is calculated…”

Changed.

P4. L6: remove ‘The’ in ‘this 43 vertical layers…’ P4. L15: “for the 2008 simulations”

Corrected.

P4.L18 “for the two study years”

Corrected.

P8, L29: “we found an increased error…”

Corrected.

P8, L30: “convection”

Corrected.

P8,L34 to P9, L17: check missing use of word “the” at several instances

We added “the” to the following sentences.

(p. 9, l. 22–23)

“*Over South Africa, the negative annual mean bias was reduced by 37% at 1.1° resolution and by 43% at 0.56° resolution, compared to 2.8° resolution, …*”

(p. 9, l. 30–31)

“*Over South America, negative bias for the annual mean concentration was 15% lower at 1.1° resolution and 12% lower at 0.56° resolution than at 2.8° resolution.*”

(p. 10, l. 2–3)

“Over Southeast Asia, RMSE for the annual mean fields was reduced by 7% at 1.1° resolution and by 5% at 0.56° resolution, compared to 2.8° resolution.”

P9, L18: suggest to reformulate to “Negative biases with respect to GOME-2 were larger than to OMI …”

Modified as suggested.

P9, L16-31: repetition of text, can be removed here.

Repetitions of text were removed.

P11, L10: “In comparison with OMI retrievals, with increasing model resolution **the** slope **for East Asia**
became…”

Corrected.

P11, L33: “The negative…”

Corrected.

P12, L4: change “chemical concentrations” to “trace gases”

Changed.

P12, L6: change “The 1.1 and 0.56” to “All”, and change “while” to “but”

Changed.

P12, L13: “within 0.5%”: are you sure about this accuracy against the observations?

The described number “0.5%” is accuracy against the observation at 800 hPa. Because the accuracies against observations vary between 800 hPa and 750 hPa, we modified this number to the corresponding accuracy range “0.5%–7%”. (p. 13, l. 4)

P15, L21: “a significant”

Corrected.

P15, L27: “simulations”

Corrected.

P15, L29: “Improve the tropospheric…”

Corrected.

P15, L32: “calculations”

Corrected.

P17, L1: Note that the TropOMI retrieval product will use TM5 on a global 1x1 horizontal resolution (Williams et al., 2017).

The sentence has been modified as follows:

(p. 18, l. 32–33)

“*, whereas the TROPOMI retrieval product will employ 1° × 1° resolution simulation fields from TM5 (Williams et al., 2017).*”

P18, L27: “captures the regional”

Corrected.

P18, L31 “points”

Corrected.

P18, L35: what is the Post-K computer?

We added the description of the post-K computer:

(p. 20, l. 28–30)

“*A post-petascale supercomputer, also known as a post-K computer, is being developed by Japan's FLAGSHIP 2020 project (e.g., Miyoshi et al., 2015), and will facilitate future studies...*”