Global high-resolution simulations of tropospheric nitrogen dioxide using CHASER V4.0: Response to reviewer #2

We would like to thank anonymous reviewer #2 for his or her careful reading and valuable comments, which have helped to significantly improve the manuscript. We revised the manuscript and responded to considering 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 response to them (in blue) are listed below. *Text (Italicized)* from the revised manuscript is in quotes.

Major comments:

1. Conclusion

Realistically, the computational time is trade-off. What is the authors' conclusion found through this study? On global scale comparison (judged from global RMSE), the authors concluded that 'The improvement when increasing resolution from 1.1_ to 0.56_ was limited' (P7, L27; Figs. 2-6; Table 1). On megacity levels, the authors concluded that 'These validation results demonstrate the capability of the 0.56_ simulation to represent high concentrations over strong local sources' (P11, L25-26; Figs. 7-8). I suppose that these results can be expected one, so what (or which) is the desired resolution at the current computational resources. We do not conduct 2.8_ resolution simulation? The conclusion described at Section 6 (P18, L27-31) conveys essential point in this study, so I would like to recommend this including also on Abstract.

We have added the discussion on the trade-off between horizontal model resolution and computational resource to the conclusion as follows:

(p. 20, l. 21–24)

"*The computational cost largely increases at 0.56° resolution, while the overall improvements were small at 0.56° resolution compared to 1.1° resolution except over megacities. Therefore, we consider that horizontal resolution of approximately 1° is a*

realistic option to obtain improved overall performance of global tropospheric NO_2 simulations."

The conclusion described in Section 6 (P18, L27—31) has been added to the Abstract as follows:

(p. 1, l. 6–8)

"*The 1.1° simulation generally captured well regional distribution of the tropospheric NO_2 column, whereas 0.56° resolution was necessary to improve model performance over areas with strong local sources with mean bias reductions of 67% over Beijing and 73% over San Francisco in summer.*"

In Section 5.2, the authors mention the relative computational burden compared to 2.8_ resolution simulation. The actual computational time (NOT compared as relative time) might bring us the valuable information.

We have added the actual computer time in Section 5.2 as follows:

(p. 18, l. 19–22)

"*High-resolution chemical transport modeling requires huge computational resources. Compared to the simulation at 2.8° resolution (approximately 480 s computer time for a 1-day simulation), the computational cost increased by a factor of 67 at 0.56° resolution (approximately 32000 s computer time) and by a factor of 14 at 1.1° resolution (approximately 6700 s computer time).*"

2. Model evaluation on 2014

In Section 3.3, the authors presented the model evaluation with FRAPPE aircraft measurement. This campaign is conducted on summer 2014 (P6, L29); however, the model simulation was based on 2010 emission intensity (P4, L16-17). The model evaluation should take into account the differences of emission intensity from 2010 to 2014. Detailed and careful discussion and possible differences are needed.

Based on an analysis of optimized NO_x emissions from an assimilation of satellite observations for the past decade (Miyazaki et al., 2017), we have added a discussion about NO_x emission differences between 2010 and 2014.

(p. 13, l. 31–35)

"*The 2014 simulations used the anthropogenic emission inventory for the year 2010 (c.f., Section 2.1). The optimized NO_x emission from an assimilation of multiple species satellite measurements (Miyazaki et al., 2017) suggest that surface NO_x emissions over*

the DMA in July-August increased by 7% from 2010 to 2014. The temporal variation, together with large uncertainties in the emission inventories, could explain part of the negative biases of NO and NO_2 at 800 hPa, which also affects OH, HO_2, and O_3 through subsequent chemistry processes."

Minor comments:
P1, L2: The expression of 'ranging from 0.56_ to 2.8_' impresses the resolution were varied with some intervals; but the simulation was conducted on 2.8_, 1.1_, and 0.56_. Please revise the expression to the correct usage.

The sentence has been rewritten as follows:
(p. 1, l. 2)
"*... at horizontal resolutions of 0.56°, 1.1°, and 2.8°.*"

P2, L9-31: In this context, 'high resolution' will mean 'high horizontal resolution'. Do the authors have some suggestion regarding 'vertical resolution'?

To discuss vertical model resolution, the following sentence has been added to the introduction in the revised manuscript:
(p. 2, l. 33–35)
"*Vertical model resolution could also be important through, for instance, vertical mixing between planetary boundary layers and the free troposphere (e.g., Menut et al., 2013).*"

(p. 3, l. 18)
"We focus on impacts of horizontal model resolution on global tropospheric NO_2 simulations."

P3, L10: Also, the expression of 'Three horizontal resolutions, varying from 2.8_ to 0.56_' is ambiguous. Please revise the expression to the correct usage.

This part has been rewritten as follows:
(p. 3, l. 19)
"*Three horizontal resolutions of 2.8°, 1.1°, and 0.56° ...*"

P3, L19: What is the update(s) on this version 4.0 of global chemical transport model

CHASER?

We have added the description on the differences between version 3.0 and 4.0 as follows:
(p. 3, l. 31–p. 4, l. 2)
"Several updates were made from CHASER V3.0 (Sudo et al., 2002) to CHASER V4.0, which include the consideration of aerosol species (sulfate, nitrate, ammonium, black and organic carbon, soil dust, and sea salt) and the implementation of related chemistry, radiation, and cloud processes. AGCM was also updated from the NIES/CCSR AGCM 5.7b to the MIROC-AGCM. Detailed information on the AGCM updates are provided by K-1 model developers (2004)."

P4, L17-18; P4, L26-28: GFED version 4.1 provides three-hourly fields, but the authors applied diurnal cycles described here? Why?

The model simulations employed monthly mean total emissions as a boundary condition. This is partly because we aim to optimize emission diurnal variations from data assimilation, as conducted by Miyazaki et al. (2017). We confirmed that the applied diurnal emission variability is similar to variability from GFED v4.1 3-hourly data over Central Africa and South America (Figure 1 in this document). Meanwhile, distinct differences in the diurnal emission variability functions around the GOME-2 overpass time (9:30LT) suggest that model performance could differ when using the GFED v4.1 3-hourly data in the comparison with the GOME-2 retrievals. The use of the GFED v4.1 3-hourly data is expected to decrease model negative biases against GOME-2 over Central Africa and increase model negative biases over South America. To discuss them, the following discussion has been added in the revised manuscript:
(p. 5, l. 10–13)
"*Over biomass burning regions, emission diurnal variability applied in this study is generally similar to variability from the 3-hourly GFED4.1 data, while distinct differences in relative magnitude around the GOME-2 overpass time suggest that model performance could differ in comparison with the GOME-2 retrievals when using the 3-hourly GFED4.1 data.*"

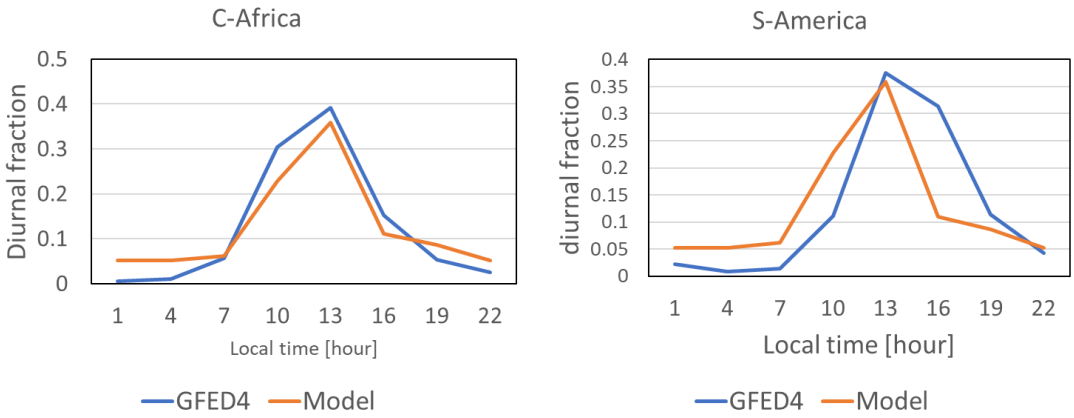

Figure 1. Diurnal emission variability functions applied for surface NO_x emissions in our model simulation (red) and those provided from GFED4.1 during June-August 2008.

P4, L29-30: Did the authors confirm that the application of the diurnal cycles of surface NOx emission can improve the simulation results on 1.1_, and 0.56_ resolution?

We confirmed the impact at 1.1° resolution but not at 0.56° resolution. A sensitivity calculation at 1.1° resolution for July 2008 suggests that the application of the diurnal cycle improves the model performance with respect to OMI over polluted and biomass burning regions (e.g., mean bias reduction by 21% over eastern China and by 32% over Central Africa). The sentences have been rewritten as follows:
(p. 5, l. 8–10)
"Miyazaki et al. (2012) confirmed that the application of this scheme leads to improvements in global tropospheric NO_2 simulation at 2.8° resolution. Improvements were commonly found in the 1.1° resolution simulation, whereas we did not evaluate the impact at 0.56° resolution."

P5, L3-27: This part includes not 'methodology' but 'results/discussion'. Some parts should be moved to appropriate locations, and reorganized as 'methodology' section. As the discussion of meteorological field, the authors showed the precipitation data with GPCP. I agree that the precipitation is one of the important parameter should be discussed; however, for gas-phase species of NO2 focused in this study, radiation will be more important because the photolysis reaction can determine the NO2 lifetime and NOx cycles. In my opinion, the discussion on meteorology can be only documented, and might not be needed as figure(s)/table(s).

The result and discussion parts in Section 2.1 have been moved to Section 3 (Validation of meteorological field) and revised as follows:
(p. 7, l. 10–11)
"*In the CTM-AGCM online framework, meteorological fields vary among different model resolutions. From sensitivity calculations, the strength and distribution of the cumulus convection were found to be sensitive to model resolution…*"

Following the comment by another reviewer, who suggested adding a more detailed analysis on meteorological fields, we have revised Figure 1 and discussion as follows. This revision contradicts your suggestion (to remove the figure), but we would appreciate your understanding.
(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; it 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."

P7, L18: The illustration of these analyzed regions in figure (e.g., on Fig. 2) is helpful.

The regions used for the model evaluation are shown in Figure 2a. The corresponding description has been added to the figure caption:
(p. 31)
"*The white square line in (a) represents the region used for the model evaluation.*"

P11, L28-29: The illustration of the Denver Metropolitan area (DMA) in figure (e.g., on Fig. 7) is helpful.

The DMA area is shown in Figure 9a in the revised manuscript. The corresponding description has been added to the figure caption:
(p. 38)
"*The DMA area is shown by the blue square line in (a).*"

P12, L16-P13, L4; Figure 11: What observation is used for these probability

distributions? Please specify.

The FRAPPÉ aircraft-campaign observation of NO at 800 hPa over the Denver Metropolitan area (DMA) is used for the probability distribution. We have modified this part to specify the observation used for the probability distribution:
(p. 13, l. 8–9).
"*Figure 11a shows the probability distribution function of NO from the FRAPPÉ aircraft observation and the model simulations at 800 hPa over the DMA.*"

P13, L6-7: From Table 2, the analyzed period will be 2008. Please specify the period in the main text.

The analyzed period has been specified as follows:
(p. 14, l. 2–3)
"*We analyzed simulated global distribution of O_3, OH, and NO_x for 2008 to characterize the resolution dependence of NO_2-related chemistry.*"

P14, L5-13: Why the tropospheric NO2 column were shown here? If the authors discussed the differences in OH and NO2, NO2 should be shown as lowermost five layers partial column as was OH.

Figure 13(a–c) has been replaced by lowermost five layers partial NO_2 column to discuss the difference between OH and NO_2 for the lower troposphere. The related description has also been modified as follows:
(p. 15, l. 6–8)
"*Figure 13 compares the spatial distribution of NO_2 and OH in the lower troposphere between model simulations. Lower tropospheric NO_2 partial columns were larger around strong source areas and smaller over rural and coastal areas around polluted regions at 1.1° and 0.56° resolutions, …*"
(p. 15, l. 11–12)
"*The differences in OH and NO_2 exhibited similar spatial patterns over polluted and biomass burning regions: e.g., r = 0.53 over the western United States, r = 0.61 over India, and r = 0.57 over South America.*"

P15, L11-12: So updated version 4.0 is not related to the improvement on the chemical kinetics?

The update does not include any improvements on chemical kinetics.

P15, L15-16: What means the differences? Anthropogenic amounts from China? What is the analyzed period?

This means a difference in the total amount of anthropogenic NO_x emission in China in 2008 between the lowest and highest inventories among the four selected inventories (REASv2.1, MEIC, EDGARv4.2, and the inventory produced by Nanjing University). This description has been clarified as follows:
(p. 16, l. 30–31)
"*The total amounts of anthropogenic NO_x emission in China in 2008 differ by 27% between two (highest and lowest) bottom-up inventories; EDGAR4.2 and MEIC (Saikawa et al., 2017).*"

P16, L17-19: Did the authors claim 'high-resolution modeling' on global scales (this will be related to Section 5.3 and 5.4)? In this manuscript, the downscaling approach was not mentioned. If we offer the improvement on megacity levels, the downscaling approach seems to be the alternate way. Especially, NO2 column is strongly related to surface NOx emissions, high-resolution over oceans might not be required (suggested from Fig. 2). Do the authors have some comments?

We have extended the discussion on advantages of global high-resolution models over methods such as downscaling and two-way nesting between global and regional models as follows:
(p. 17, l. 33–p. 18, l. 13)
"*Most previous high-resolution modeling studies have used regional models to simulate NO_2 concentration fields at high-spatial resolution, primarily focusing on urban regions, with reduced or equivalent computational costs compared to global models. Several studies demonstrated that a better representation of long-range transport of NO_x reservoir species such as peroxyacetyl nitrate (PAN) are important on simulated NO_2 in the free troposphere in remote areas (e.g., Hudman et al., 2004; Fischer et al., 2010, 2014; Jiang et al., 2016). A two-way nesting between regional and coarse-resolution global models (e.g., Yan et al., 2016) is able to consider both small-scale processes inside focusing regions and long-range transport over the globe, which has an advantage over regional models. An important advantage of global models over*

regional models and two-way nesting systems is the ability to simulate NO_2 concentration fields at high resolutions over the entire globe across urban, biomass burning, and remote regions in a consistent framework. Even over remote regions, a high-resolution simulation has the potential to improve model performance through considering the effects of non-linear chemistry in high-concentrated NO_x plumes emitted from ships and lightning (Charlton-Perez et al., 2009; Vinken et al., 2011; Gressent et al., 2016). These NOx emission sources in remote regions have significant impacts on climate and air quality (Eyring et al., 2010; Holmes et al., 2014; Banerjee et al., 2014; Finney et al., 2016). It is thus important to clarify the importance of resolving small-scale sources and plumes within a global modeling framework for better understanding of the global atmospheric environment and chemistry-climate system."

Figure 10: This figure presented the comparison with observation over the Denver metropolitan area, so please specify the simulation period explicitly.

We have added the period to the caption of figure 10 as follows:
(p. 39)
"… over the Denver metropolitan area (39-41°N and 103-105.5°W) during the FRAPPE period (from July 16 to August 18, 2014).".

Figure 12: What makes the OH increment over high altitude over southern hemisphere?

We have added this explanation to the revised manuscript:
(p. 15, l. 4–5)
"A large relative OH increment was found over the Antarctic, because weak ultraviolet radiation led to small OH concentrations during a polar night."

Table 2: These statistical scores averaged over global might be helpful to understand the improvement according to the resolution change. Why so large MB and RMSE are found on 100 hPa comparison?

We have added the statistical scores averaged over all available ozonesonde, and the corresponding description:
(p. 14, l. 17–20)
"*Overall, RMSE with respect to the globally available ozonesondes was reduced with increasing resolution (by up to 8.1 ppbv) at 850 hPa and 500 hPa. In contrast, at 300*

hPa, RMSE increased at 0.56° (by 1.2 ppbv) and 1.1° (by 9.4 ppbv) resolutions, reflecting larger RMSE at 0.56° and 1.1° resolutions in the high-latitudes of both hemispheres."

The observed and simulated ozone concentrations are large at 100 hPa. The relative values of MB and RMSE with respect to the observed concentrations are comparable between 100 hPa and the other pressure surfaces, except at the southern high latitudes, where the relative MB is larger by a factor of 2 at 100 hPa than that at the other altitudes.

Technical comments:
Figure 1: The color bars might be understood, but it will be better to fit the corresponded figures.

Modified.

Figure 2: The color bars might be understood, but it will be better to fit the corresponded figures. Specific indication by using (a) to (h) will be better.

Modified.

Figure 3 to Figure 6: Specific indication by using (a) to (h) will be better not using column and row expressions, or remove (a) to (k) because (a) to (k) were not used in the main text (P8, L1-P9, L17)

Removed the specific indications.

Figure 7: The color bars might be understood, but it will be better to fit the corresponded figures. Specific indication by using (a) to (h) will be better. The coastline of map in first column should be emphasized to be distinguished. Typo of 'Dever' on (i).

Modified.

Figure 8: Typo of 'Shengzhen' in the figure.

Corrected.

Figure 9: Specific indication by using (a) to (h) will be better.

Modified.

Figure 10: Specific indication by using (a) to (j) will be better.

Modified.

Figure 13: The color bars might be understood, but it will be better to fit the corresponded figures.

Modified.

References

Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal changes in global surface NOx emissions from multi-constituent satellite data assimilation, Atmos. Chem. Phys., 17, 807–837, doi:10.5194/acp-17-807-2017, 2017.