A comparison of the impact of TROPOMI and OMI tropospheric NO2 on global chemical data assimilation: Reply to comments from anonymous referee #2

We would like to thank anonymous referee #2 for his or her careful reading and valuable comments, which have helped to significantly improve the manuscript. We have revised the manuscript corresponding to the referee's comments. Main changes we made are as follows:

1) Appendix A was added to discuss the seasonally varying bias of TROPOMI NO_2.

2) The discussion on the potential impacts of a low bias of TROPOMI in winter on the DA performance was added to Section 6.

3) The comparison of surface NO_2 concentrations derived from the control model simulation, TROPOMI DA, and OMI DA was added to Figure 9.

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

At the time of the paper submission, the version 2.2 product was not available for April—May 2018, the submitted manuscript compared TROPOMI NO_2 version 1.2.2 product during September 2018 with version 2.2 product during September 2021 in Appendix A. This comparison for different periods caused a limitation to clearly distinguish the impacts of the algorithm updates from those of inter-annual changes. After the paper submission, the S5P-PAL reprocessing product became available for May 2018—July 2021. To provide more a consistent comparison for the same time period, we have revised Appendix B (corresponding to Appendix A of the submitted manuscript) to the comparison between version 1.2beta and S5P-PAL reprocessing products for May 2018. Although this change was not requested by the referees, we think that this update provides a better implication for the impacts of algorithm updates. The paragraph has been revised as follows:

(p. 41, l. 793—809)

"*In the latest version of the TROPOMI NO_2 product, the low bias compared to OMI QA4ECV is largely improved from the previous versions (van Geffen et al., 2021). To discuss the potential impacts of the retrieval algorithm updates on the DA performance, Figure B1 compares global distributions of tropospheric NO_2 column, super-observation errors, and relative super-observation errors (i.e., errors divided by concentrations) obtained from the TROPOMI version 1.2beta product, that was used in this study, and S5P-PAL reprocessing product (processed with same processor as version 2.3.1), that was released more recently, for May 2018. The S5P-PAL reprocessing product data were obtained from the S5P-PAL data portal (https://data-portal.s5p-pal.com). The algorithm updates from versions 1.2 to 2.3 led to increases in tropospheric NO_2 column amounts typically by 6% over polluted areas due to the algorithm updates from versions 1.2 to 2.3. These increases are mainly attributable to the improved FRESCO cloud retrievals (van Geffen et al., 2021). In contrast, the relative super-observation errors* *over most regions except for the southern mid-latitudes are comparable between the products, with less than 0.2% differences in the mean relative super-observation error over 60°N–60°S. These differences are much smaller than the differences between the TROPOMI version 1.2beta and OMI QA4ECV products (by 19% in May 2018).*

The improved TROPOMI retrievals would reduce the negative bias in NO_2 concentration analysis and increase the estimated NO_x emissions for areas with weak chemical non-linearity. The increase in NO_x emissions would reduce the negative biases in ozone analysis under NO_x-limited ozone chemical regime. Meanwhile, the relative super-observation errors of TROPOMI retrievals were almost identical between versions 1.2beta and 2.3.1. This suggests that the DA efficiency, for example, to constrain detailed temporal and spatial variations, might not be largely affected by the algorithm updates."

The manuscript by Sekiya et al. compared the global chemical data assimilation results when using NO2 retrievals from TROPOMI and OMI. The TROPOMI posterior NO2 shows better agreement with NO2 observations and smaller magnitude than the OMI one. The manuscript is generally well-written. The topic fit the scope of AMT. The result is important in interpreting existing NOx data assimilations. I suggest publication after addressing the following comments.

We appreciate in careful reading and comments again.

L7, if TROPOMI NO2 is biased generally low, would the comparison with independent data improved for the wrong reason?

Negative biases in NO_2 concentration analysis against independent observations were increased by TROPOMI DA for some cases. Meanwhile, the agreements with independent observations were improved by better constraints on spatial and temporal variations in NO_2 in TROPOMI DA than in OMI DA, because of lower super-observation errors in TROPOMI than those in OMI. We added description to abstract as follows:

(p. 1, l. 9)

"*... because of better capturing spatial and temporal variability by TROPOMI DA.*"

Figure 1. Please provide the resolution these data are gridded to in the figure description.

We added the following sentence on mapping grids to caption of Figure 1:

(p. 26)

"*The values are mapped onto 0.56° resolution grids.*"

How much do precision error and the number of observations in the super-observation grid each contribute to the smaller super-observation errors in TROPOMI data?

The precision error improvements were more important than the increases in the number of observations per a grid. Meanwhile, the relative contributions of precision error improvements were smaller over polluted regions than over remote regions. The following description was added:
(p. 7, l. 208—209)
"*The improved S/N ratio and stripes contributed to about 80% and almost 100% of smaller super-observation errors over polluted and remote regions, respectively*"

Line 221-222, it would be clearer to first explain what the range of chi-square is, and what do values larger and smaller than 1 generally mean.

We have revised the explanation on meaning of chi-square values as follows and moved it to before results.
(p. 8, l. 226—228)
"*χ^2 value is used to diagnose balance between actual errors and estimated errors. When χ^2 value is larger (smaller) than the ideal value of 1, it is suggested underestimated (overestimated) background error covariance or observation errors.*"

The range of chi-square values (i.e., standard deviation) were also added:
(p. 8, l. 232—233)
"*The mean values of estimated χ^2 with standard deviation range … was 0.99±0.25 for TROPOMI DA, whereas the mean χ^2 of 1.17±0.19 for OMI DA is …*"

Figure 2, I am a bit surprised that a large portion of the TROPOMI DA improvement is over the ocean, where there is no emissions. Please explain what possibly causes this.

The DA system used in this study optimizes NO_2 concentrations and NOx emissions simultaneously, which led to the improvements over land and ocean where there is no emissions. The variables which are optimized by DA is emphasized in Section 2.3.2 as follows:
(p. 5, l. 139—140)
"*…, which optimizes ozone and related chemical species' concentrations, and ozone precursors' emissions simultaneously.*"

We also added the explanation about improvements in TROPOMI DA over oceans as follows:
(p. 9, l. 249—251)
"*Over the oceans in the tropics and midlatitudes, higher vertical sensitivity (i.e., averaging kernels) in TROPOMI than OMI in the lower troposphere and above clouds contributed to the improved performance, through ship and lightning NOx emission corrections and direct NO_2 concentration modifications.*"

L250-251, I am confused about the "regardless of the TROPOMI low bias" part. Is this only true because you calculate RMSE against the TROPOMI observations?

Yes, because the RMSE was estimated against the assimilated TROPOMI measurement, it is not affected by the TROPOMI low bias. This part was modified as follows:
(p. 9, l. 263—264)
"*... the DA efficiency by TROPOMI was evaluated based on RMSE against assimilated observation Itself. It is determined by the amount and quality of TROPOMI data, regardless of the TROPOMI low bias.*"

Figure 4, Please provide more information on what is being optimized in the DA. Are both NO2 concentrations and emissions optimized at the same time?

The DA system used in this study optimizes NO_2 concentrations and NO_x emissions at the same time. We add this explanation to Section 2.3.2 (please see our reply above).

Are emissions all in the surface layer? If not, how are they distributed vertically, and how does DA adjust emissions differently at different layers?

Anthropogenic (except for aviation), biomass burning, and soil emissions are in the lowest model layer. The following sentence was added:
(p. 5, l. 133—134)
"*These emissions are released at the lowest model layers.*"

Lightning NOx sources are vertically distributed using the C-shaped profile given by Pickering et al. (1998). The following description was added:
(p. 5, l. 134—136)
"*... the parameterization proposed by Price and Rind (1992), with the assumption for vertical distribution of lighting NOx source based on the C-shaped profile given by Pickering et al. (1998).*"

Data assimilation adjusts 3-D multiplication factors for the lightning NO production rate. The following sentence was added:
(p. 6, l. 155—157)
"*For lightning NO_x, multiplication factors for the lightning NO production rate were adjusted differently at different model layers using the method proposed by Miyazaki et al. (2014) and the background error covariance matrix.*"

L287, a similar comment as a previous one, if there are systematic low biases in TROPOMI data, why do its DA results have better agreement with independent data?

As mentioned in the second reply, TROPOMI version 1.2 data have negative biases, which led to increased negative biases in TROPOMI DA against independent surface in-situ observations for some cases. Meanwhile, TROPOMI DA better captured spatial and temporal variations mainly because of reduced TROPOMI super-observation errors associated with improved S/N ratio, increased number of observation data per a grid, and TROPOMI stripes. This part was modified to explain cause of the improvements as follows:
(p. 10, l. 302)
"*... reduced RMSE by 23% because of better capturing spatial and temporal variations, but increased ...*"

Also, we added the following sentence at the end of this paragraph to emphasis how the performance TROPOMI DA is improved.
(p. 10, l. 307—308)
"*..., while TROPOMI DA provided better constraints on spatial and temporal variations in NO_2 concentrations than OMI DA.*"

L330, could you also add a figure showing the changes in NO2 concentrations from the two DA?

We added global maps of the surface NO_2 concentration analysis derived from TROPOMI DA and the differences from the control model simulation and OMI DA to Figure 9. The corresponding description was also changed:
(p. 12, l. 357—358)
"*..., which led to smaller surface NO2 concentrations (Figure 9). These ...*"

L339-351, based on the low biases in TROPOMI NO2 retrievals and the comparisons here, what is the implication for existing DA and inversion results using this version of TROPOMI NO2?

We added the implication for DA using TROPOMI NO2 version 1.2-1.3:

(p. 13, l. 381—384)

"*Overall, these results imply that top-down NOx emission estimates using TROPOMI version 1.2-1.3 products could be affected by the TROPOMI low biases compared to OMI, while top-down estimates using TROPOMI have the potential for constraints on detailed spatial and temporal variations based on validation results (c.f., Section 3.3).*"

L446-447, would you expect the low biases in TROPOMI NOx emissions reduce using this new product, and by how much?

Based on the complemental analysis presented in Appendix B in the revised manuscript, we expect to reduce impacts of TROPOMI low bias on emission estimates if the new version products are used. This part was modified to be more quantitative, as follows:

(p. 16, l. 474—479)

"*These new versions largely remove the bias with respect to the OMI 475 QA4ECV product for all seasons, especially in winter over polluted areas (van Geffen et al., 2021). Lambert et al. (2021) and van Geffen et al. (2021) reported that the negative biases of the updated TROPOMI retrievals (versions 1.4.x and 2.x) compared to OMI are reduced to within 10%. Assuming a remaining bias of 10% compared to OMI, the improved TROPOMI retrievals would increase the estimated NOx emissions by 10–30% over Europe and eastern China in winter for areas with a weak chemical non-linearity, compared to the DA using TROPOMI version 1.2beta.*"

Reference

Pickering, K. E., Wang, Y., Tao, W.-K., Price, C., and Müller, J.-F.: Vertical distributions of lightning NOx for use in regional and global chemical transport models, J. Geophys. Res., 103, 31 203–31 216, https://doi.org/10.1029/98JD02651, 1998