Response to Reviewers

Response to comments from Reviewer #1

[Comment 1] *l 27: "We find a 38 % increase of tropospheric NO_2 in version 1.4 due to improved FRESCO-wide cloud retrieval, and a 14 % increase in version 2.2 due to adjusted surface albedo for cloud-free scenes."*

This result is very similar to the conclusion drawn in paper of van Geffen, 2022, where the retrieval changes in version 2.2 (and 1.4) are discussed. Also the comparison against OMI-QA4ECV are presented in this paper.

[Response] Thanks for your comments. We really appreciate the study on the impact of TROPOMI NO_2 v2.2 retrieval improvements on a global scale by van Geffen et al. (2022). In this work we follow previous studies and evaluate the impact of TROPOMI NO_2 v1.3-2.4 retrieval improvements over China by using TROPOMI, OMNO2 and QA4ECV OMI data. We have extended the study period to TROPOMI NO_2 v2.4, find an increase by 27-40 % of tropospheric NO_2 with the introducing of v2.4 over vegetation. Furthermore, we find that TROPOMI v1.3-2.2 data shows strongest tropospheric NO_2 seasonal variation compared to OMNO2 and QA4ECV OMI data, and this seasonal effect was enhanced with the tropospheric NO_2 retrieval version upgrades. Lastly, we conduct a correction for the underestimation of TROPOMI tropospheric NO_2 in the previous version retrievals, and find a 33 % overestimation of NO_2 reduction during the COVID-19 lockdown over China when using TROPOMI data before and after the activation of the NO_2 version 1.4.

[Comment 2] *l 29: "We show that the upgrade to version 2.4 with new DLER surface albedo, led to an increase by 3 x 10^{14} molecules cm^{-2} of tropospheric NO_2 over vegetation."*

The paper includes only two months of data of version 2.4, which I would judge is not enough to document the impact of the upgrade to v2.4. Possible weather influences are not discussed by the authors. The increase of NO_2 over vegetation has been *discussed in the release documentation, the ATBD and the readme file. As indicated below, I was not convinced by the analysis on this topic.*

[Response] Thanks for the comments. We have extended the TROPOMI NO_2 v2.4 data series to December 2022, and add a comparison of daily TROPOMI tropospheric NO_2 columns in December of 2020 (v1.4), 2021 (v2.2) and 2022 (v2.4) over Fujian province and China, as well as a discussion on the impacts of TROPOMI NO_2 v2.4 retrieval over vegetation in winter month. Therefore, in this manuscript we investigate the impacts of TROPOMI NO_2 v2.4 improvements under conditions with lush vegetation (summertime) and withered vegetation (winertime) over high vegetation coverage and the whole China. We find that from v1.4 to 2.2 the TROPOMI tropospheric NO_2 enhancement over China (7 %, 2.30 $\times$ 10^{14} molecules cm^{-2}) is greater than over Fujian which is the province with the highest vegetation coverage in China (1 %, 0.17 $\times$ 10^{14} molecules cm^{-2}) in December, similar as the comparison between them in August. However, from v2.2 to 2.4 the TROPOMI tropospheric NO_2 column in December is decreased over the whole China (20 %, 6.87 $\times$ 10^{14} molecules cm^{-2}) but increased over Fujian (4 %, 1.14 $\times$ 10^{14} molecules cm^{-2}). We infer that the impact of the TROPOMI v2.4 improvements with the DLER surface albedo on NO_2 column enhancement is relatively stronger over higher vegetation coverage, under the condition with lush vegetation and low NO_2 level in summer. Furthermore, in winter when the condition has been changed with withered vegetation and high NO_2 emission, the impact of TROPOMI DLER in NO_2 v2.4 retrieval is even more obvious. Please see Line 352-356, Lie 368-380 and Figure 4 in the revised manuscript.

TROPOMI NO_2 issue 2.2 product documentation (S5P-MPC-KNMI-PRF-NO_2) (released in July 2022) tested the impact of DLER in v2.4 for September 2020 on a global scale, suggested that the impact on tropospheric NO_2 over Europe, North America and East China is relatively minor, but is substantial over vegetated regions like South America or Central Africa. Our manuscript provides a validation and extension of the tested result in S5P-MPC-KNMI-PRF-NO_2 document, as well as a quantitative measurement of the impact of TROPOMI NO_2 v2.4 improvements on the basis of actual observation data.

[Comment 3] *l 31: "we demonstrate that TROPOMI data shows strongest tropospheric NO_2 seasonal variation compared to OMNO2 data and QA4ECV OMI data, and this seasonal effect was enhanced with the tropospheric NO_2 retrieval version upgrades": In the paper by van Geffen it was also reported that the largest increases occur in wintertime, so not really a new results.*

[Response] Thanks for your comment. We really appreciate the research on the impact of TROPOMI NO_2 v2.2 retrieval upgrade on a global scale by van Geffen et al. (2022). van Geffen et al. (2022) suggested that TROPOMI tropospheric NO_2 v2.2 data is larger than v1.x data, depending on the level of pollution and season, with the largest impact occurs in wintertime. In this manuscript we make some tentative attempts and research on the impacts of TROPOMI NO_2 v1.3-2.4 retrieval upgrades, we find that TROPOMI v1.3-2.2 data shows strongest seasonal variation of tropospheric NO_2 compared to OMNO2 and QA4ECV OMI data, and the improvements in the TROPOMI NO_2 retrieval upgrades lead to form stronger effect of tropospheric NO_2 seasonal variation.

[Comment 4] *l 33: "we arrive at a correction for the underestimation of TROPOMI NO_2 column in previous versions"*

In this respect the paper is a bit late. Such corrections are no longer relevant given the S5P-PAL (available since december 2021) and the reprocessing of v2.4 (available since March 2023). Such corrections have been introduced before, e.g. Riess et al, 2021.

[Response] Thanks for the comments. Riess et al. (2022) used TROPOMI NO_2 v1.2-2.1 and QA4ECV OMI data, applied an artificial neural network, investigated the impact of the COVID-19 pandemic on ship NO_2 pollution over European seas, and found that NOx emissions from ships reduced by 20-25 % during the pandemic. In this work we use TROPOMI NO_2 v1.3-2.4, OMNO2 and QA4ECV OMI data, derive the changes of TROPOMI tropospheric AMF from v1.3-2.4, investigate the impact of the COVID-19 lockdown on NO_2 column over China, we also obtain the expected NO_2 reduction during the lockdown over China retrieved from TROPOMI, OMNO2 and QA4ECV OMI data, and find a 33 % overestimation of NO_2 reduction during the lockdown over China when using TROPOMI data before and after the activation of v1.4.

[Comment 5] *l 35: "We also find a 33 % overestimation of NO_2 reduction during the COVID-19 lockdown over China when using TROPOMI data before and after the activation of the NO_2 version 1.4." This follows directly from the large increase introduced in v1.4 and is a bit a trivial result. A better analysis of the COVID period was a main motivation to launch the S5P-PAL reprocessing.*

[Response] Thanks for the comments. On the one hand, although the upgrade to TROPOMI v1.4 with the improved FRESCO cloud retrieval can lead to a significant increase of tropospheric NO_2 as compared with the previous version. Quantitative measurement the impact of TROPOMI v1.4 retrieval improvements on NO_2 column changes during the COVID-19 lockdown over China is valuable for the application of TROPOMI-derived NO_2 measurement.

On the other hand, TROPOMI operational, OMNO2 and QA4ECV OMI data, instead of S5P-PAL data, are used to investigate the COVID-19 lockdown on NO_2 pollution over China in this manuscript is mainly due to the following reasons. Firstly, the aim of our manuscript is to evaluate the impacts of TROPOMI NO_2 v1.3-2.4 retrieval improvements over China, and thus, the S5P-PAL NO_2 dataset, which is a reprocessing of the TROPOMI official NO_2 data product, is not used in this manuscript as a reference for comparison. We appreciate the contribution of this dataset in supporting research on the impact of the COVID-19 lockdown by satellite-derived NO_2 observations, and will concern this dataset in the future work on NO_2 monitoring during the lockdown. Secondly, up to date OMI and TROPOMI have been the main data sources in satellite monitoring of NO_2 (Biswal et al., 2021), the retrieval of tropospheric NO_2 from QA4ECV OMI proceeds along the same lines as from TROPOMI, and is similar in many aspects (Riess et al., 2022). Considering the

QA4ECV OMI NO_2 data product is available before 30 March 2021, OMNO2 data is used to compare with TROPOMI v2.2 (from July 2021-June 2022) and v2.4 (from July 2022-) data. Therefore, in our manuscript the comparisons of TROPOMI NO_2 v1.3-2.4 data with OMNO2 and QA4ECV OMI NO_2 data can already reflect the impacts of TROPOMI NO_2 retrieval improvements from v1.3-2.4.

[Comment 6] *Why do the authors focus only on China? The author team is from China, but the paper is submitted to an international journal. It would be just as relevant to know the impacts over Europe, USA, the tropics etc. As indicated by Van Geffen et al., 2022, the impact seems to be quite dependent on the region.*

[Response] Thanks for the comments. In this manuscript we focus on investigating the impacts of TROPOMI NO_2 v1.3-2.4 improvements on NO_2 column changes over China. We select China as the study area because that China is one of the regions with heaviest NO_2 pollution in the world, monitoring NO_2 columns over China can well demonstrate its spatial-temporal change characteristics, and furthermore, quantitative measurement of NO_2 column changes in TROPOMI different version periods over China can clearly reflect the impacts of these version improvements on NO_2 retrieval.

[Comment 7] *Why is a reference to the S5P-PAL dataset (https://data-portal.s5p-pal.com/products/no2.html) not included? This PAL dataset was generated to remove the jumps between versions, e.g. for Covid studies.*

[Response] Thanks for your comment. Firstly, OMI and TROPOMI have been the main data sources in satellite monitoring of NO_2 to date, Retrieval of tropospheric NO_2 from QA4ECV OMI proceeds along the same lines as from TROPOMI. Thus QA4ECV OMI data is widely used as reference in previous studies on investigations of impacts of TROPOMI NO_2 version upgrades (Riess et al., 2022, van Geffen et al., 2022). Secondly, since the QA4ECV OMI NO_2 data is available before 30 March 2021, in order to investigate the impacts of TROPOMI v2.2 (from July 2021-June 2022) and v2.4 (from July 2022-) on NO_2 retrieval, OMNO2 data is used to compare. Lastly, the S5P-PAL NO_2 dataset is a reprocessing of the TROPOMI official NO_2 data product, while our work aims to evaluate the impacts of TROPOMI NO_2 v1.3-2.4 retrieval improvements over China, thus S5P-PAL NO_2 data is not applied to compare with TROPOMI operational NO_2 data. Overall, comparisons of TROPOMI NO_2 v1.3-2.4 data with OMNO2 and QA4ECV OMI data, instead of S5P-PAL data, have been able to well accomplish the evaluation of the impacts of these different retrieval version improvements.

[Comment 8] *There is a new reprocessing available since March 2023, covering the full mission duration, see https://sentinels.copernicus.eu/web/sentinel/-/copernicus-sentinel-5-precursor-full-mission-reprocessed-datasets-further-products-release.*

The seasonality linked to the DLER albedo update can be studied with this new dataset. I realise that the paper was written before the reanalysis became available, so this is not a main reason for my negative judgement.

[Response] Thanks for your comments. Copernicus Sentinel-5 Precursor full mission reprocessed datasets have not released when we completed this manuscript. We will concern these new datasets in the future work of air pollution monitoring.

[Comment 9] *The paper focusses on China, but the POMINO product is not mentioned at all (http://www.pku-atmos-acm.org/acmProduct.php/#TROPOMI). This is a clear omission.*

[Response] Thanks for your comment. OMI and TROPOMI have been the main data sources in satellite monitoring of NO_2 to date, because the retrieval of tropospheric NO_2 from QA4ECV OMI proceeds along the same lines as from TROPOMI, thus QA4ECV OMI data is used in this work on investigations of impacts of TROPOMI NO_2 version upgrades. Moreover, the QA4ECV OMI NO_2 data product is available before 30 March 2021, so OMNO2 data is also used to compare with TROPOMI data in the v2.2 (from July 2021-June 2022) and v2.4 (from July 2022-) periods. Additionally, the POMINO-TROPOMI product is retrieved from the TROPOMI instrument and based on calculation of tropospheric AMFs by applying a radiative transfer model LIDORT. But in this work we focus on evaluating TROPOMI's capability to detect tropospheric NO_2 in different version retrievals itself, and thus the POMINO-TROPOMI product, which is also derived by TROPOMI, is not applied in this manuscript. We appreciate the contribution of this dataset in satellite-derived NO_2 monitoring for China, and will concern this dataset in the future work of NO_2 pollution monitoring in China.

[Comment 10] *It was surprising that Lamsal et al, https://doi.org/10.5194/amt-14-455-2021, is not cited for v4 of OMNO2A.*

[Response] Done as suggested. We have reviewed and cited the related findings in the paper of Lamsal et al. (2021) accordingly. Please see Line 206 in the revised manuscript.

[Comment 11] *There is no reference to the product readme file and user manual documents of TROPOMI NO_2: these documents inform users of the updates of the processor and main impacts and are therefore relevant for this paper.*

[Response] Thanks for your comment. The product readme file and user manual document of TROPOMI NO_2 (e.g. S5P-MPC-KNMI-PRF-NO_2 issue 2.2 document) is already cited in our manuscript. Please see Line 170-172, Line 293-295, and Line 322-324 in our manuscript.

[Comment 12] *As indicated by the authors, the v2.4 upgrade is "not well documented" in the peer-reviewed literature. I can sympathise with this statement. But this version is new, and is also used in the recent reprocessing. Did the authors get in contact with the retrieval team, which would be a normal step to take?*

[Response] Thanks for your comment. We have dug up the information on TROPOMI NO_2 v2.4 retrieval improvements from official ESA channels, including TROPOMI ATBD tropospheric and total NO_2 (S5P-KNMI-L2-0005-RP) issue 2.4 document, S5P-MPC-KNMI-PRF-NO_2 issue 2.2 document, and some other relevant papers (e.g. Tilstra et al., 2017).

[Comment 13] *The analyses presented by the authors are rather straightforward, consisting of simple comparisons of averages of the tropospheric column and the tropospheric air-mass factor between different retrieval versions. A more in-depth analysis of the differences is missing, and the general conclusions broadly agree with what has already been reported before.*

[Response] Thanks for your comments. In this work we evaluate the impacts of TROPOMI NO_2 v1.3-2.4 retrieval upgrades over China, quantitative measure these impacts on NO_2 column changes, and also make deep analysis and discussions on these impacts on NO_2 retrievals. Take the impact of TROPOMI NO_2 v2.4 retrieval upgrade as an example, the DLER surface albedo using in TROPOMI v2.4 accounts for the directionality or viewing-angle dependence of the scattering at the surface, especially over vegetation in the near infrared. Thus according to this strong effect of the DLER over vegetation, we evaluate to the new DLER surface albedo in and its impact on the TROPOMI NO_2 columns, to better understand the detection of NO_2 under condition of vegetation coverage. Moreover, considering weather variability may play a big role in vegetation coverage changes, we compare the TROPOMI tropospheric NO_2 VCD daily data in August (condition with lush vegetation) and December (condition with withered vegetation) of 2020 (v1.3, 1.4), 2021 (v2.2) and 2022 (v2.4) over Fujian province (the province with the highest vegetation coverage in China), as well as over China. We find that from v1.3 to 2.2 to 2.4, over China the August TROPOMI tropospheric NO_2 column is increased by 9 % (1.06 $\times$ 10^{14} molecules cm^{-2}) and 5 % (0.73 $\times$ 10^{14} molecules cm^{-2}) respectively, and in comparison, the increase in August TROPOMI tropospheric NO_2 column over Fujian from v1.3 to 2.2 is relatively minor (2 %, 0.35 $\times$ 10^{14} molecules cm^{-2}), but the tropospheric NO_2 enhancements over this region from v2.2 to 2.4 are presented, with a substantial increase (16 %, 2.42 $\times$ 10^{14} molecules cm^{-2}). The impact of the TROPOMI v2.4 improvements with the DLER surface albedo on NO_2 column enhancement is relatively stronger over higher vegetation coverage, under the condition with lush vegetation and low NO_2 level in summer. Furthermore, in winter when the condition has been changed with withered vegetation and high NO_2 emission, the impact of TROPOMI DLER in NO_2 v2.4 retrieval is even more obvious. For instance, from v1.4 to 2.2, the TROPOMI tropospheric NO_2 enhancement over China (7 %, 2.30 $\times$ 10^{14} molecules cm^{-2}) is greater than over Fujian (1 %, 0.17 $\times$ 10^{14} molecules cm^{-2}) in December, similar as the comparison between them in August. However, from v2.2 to 2.4 the TROPOMI tropospheric NO_2 column in December is decreased over the whole China (20 %, 6.87 $\times$ 10^{14} molecules cm^{-2}), but increased over Fujian (4 %, 1.14 $\times$ 10^{14} molecules cm^{-2}). Please see Section 3.2 in the revised manuscript.

[Comment 14] *The data series stops in September 2022. This is a very short period (only two months) to make any statements on the version 2.4 data. Because the albedo climatology is available on a monthly basis, it would be important to document a full year of data.*

[Response] Thanks for your comment. We have extended the data series to December 2022, and add comparisons of TROPOMI NO_2 and OMI NO_2 in December of 2020-2022 over Fujian province and China, as well as a discussion on the impacts of TROPOMI NO_2 v2.4 improvements during withered month. Thus, the impacts of TROPOMI NO_2 v2.4 improvements under conditions with lush vegetation (summertime) and withered vegetation (winertime) over high vegetation coverage and the whole China are all investigated. Please see Line 364-380 and Figure 4 in the revised manuscript. In addition, since TROPOMI NO_2 v2.4 data is in operation from July 2022, a full year of this data can not be obtained to date.

[Comment 15] *I found the analysis of the impact over vegetation not convincing: How can we compare relative differences in Fujian and the entire China? Many aspects may play a role here. Furthermore, the analysis is limited to one month, which is not convincing as weather variability may play a big role. So I do not think the authors have presented enough evidence to quantify the increase due to v2.4.*

[Response] Thanks for your comments. On the one hand, because the DLER surface albedo using in TROPOMI v2.4 accounts for the directionality or viewing-angle dependence of the scattering at the surface, especially over vegetation. We select Fujian province which is the province with the highest vegetation coverage in China as the study area to investigate the strong effect of the DLER over vegetation, and here we also measure the impact of the DLER in TROPOMI v2.4 on NO_2 retrieval over the whole China as a reference. On the other hand, we have extended the data series to December 2022, and add comparisons of TROPOMI NO_2 and OMI NO_2 in December of 2020-2022 over Fujian province and China, thus the impacts of TROPOMI NO_2 v2.4 improvements under conditions with lush vegetation and withered vegetation are both investigated, and the relative differences of TROPOMI NO_2 column changes due to the introduction of v2.4 over Fujian and the entire China are derived. Please see Line 364-380 and Figure 4 in the revised manuscript.

[Comment 16] *Concerning the seasonality, section 3.3: I could not reconcile the results of Figure 5 with figure 6, for QA4ECV OMI. Is there a mistake in figure 5, since the seasonality seems much too small?*

[Response] Thanks for your comments. Sorry for the confusion caused by the unclear expressions of the figure captions of Figure 5 and 6. We have revised these figure captions in the revised manuscript. In our manuscript Figure 5 shows the result of NO_2 seasonal variation for the whole China retrieved by TROPOMI, OMNO2 and QA4ECV OMI data. From Figure 5 it can be clearly seen that over China compared to OMNO2 and QA4ECV OMI data, TROPOMI data shows strongest seasonal variation of tropospheric NO_2 columns, and the extents of the observed NO_2 changes in winter or summer month retrieved from TROPOMI exceed those retrieved from OMI. In addition, although QA4ECV OMI follows a more similar NO_2 retrieval algorithm to TROPOMI relative to OMNO2, the increase in winter and decrease in summer of NO_2 observed with QA4ECV OMI (-0.5 % and -5 %) are even smaller than those observed with OMNO2 (4 % and -18 %) over China. Taking this into account, we conclude that the FRESCO-wide cloud algorithm using in the NO_2 retrieval has a positive impact on the clear demonstration of seasonal variation of TROPOMI tropospheric NO_2.

Figure 6 demonstrate the result of NO_2 time series for the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) region in China by using TROPOMI, OMNO2 and QA4ECV OMI data. From Figure 6 it can be clearly seen that over these three regions with high NO_2 pollution, The characteristics of NO_2 seasonal variation derived from TROPOMI, OMNO2 and QA4ECV OMI data are all significant. We calculate the differences between TROPOMI and QA4ECV OMI tropospheric NO_2 daily VCDs for each selected pollution region and each month from November 2019 to November 2020, we find that compared to the QA4ECV OMI NO_2 retrieval, the FRESCO cloud algorithm using in the TROPOMI NO_2 retrieval has a strongly positive impact on the tropospheric NO_2 seasonal cycle, especially in high pollution regions.

Reference

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