Response to reviewer #2

We thank the reviewer for careful and thorough reading of this manuscript and the thoughtful comments and suggestions. Our answers follow the reviewer's comments (in *Italics*).

General comments:

There has been many studies in the last years on the recent trends of NOx emissions over the U.S., the main motivation being the apparent important change in NO2 column trend since 2010, which obviously requires careful analysis using the available data as well as using models. The present study is useful, as it clearly shows that there is no significant discrepancy between the NEI emission trends and the different NO2 (surface and column) data, when considering only urban areas. The paper discusses the non-linear relationship between NOx emissions and NO2 abundances. Model calculations using REAM at 36kmx36 km are used to illustrate this point and show that the feedbacks are much stronger at low-NOx than at high-NOx. Although the relevance of NOx natural emissions (which obviously do not have the same trends as the anthropogenic component) is mentioned, the paper does not dwell on it.

In fact, and this is my main comment, I think clarifications are needed in order to sort out the respective roles of chemical non-linearities and the existence of the background. Both natural emissions and chemical non-linearities play their largest role during summer over rural areas, and more so in the free troposphere than near the surface. But it is not entirely clear from the paper how much these two main factors contributed to

the apparent discrepancy between the different sets of trends. This should be clarified.

Reply:

Thank you for your suggestions. Since in our trend analyses in Section 3.3, we chose urban regions with small β and γ values and had minimized the impacts of chemical nonlinearity and background sources on inferring anthropogenic NO_x emissions from satellite datasets. Silvern et al. (2019) also show that lightning NO_x and the lifetime of tropospheric NO_x have no significant trend signals from 2005 – 2017. We think you ask which factors affect β and γ more.

Due to the interactions among NO_x emissions, chemistry, and physical processes, it is difficult to completely and accurately separate the effects of all factors to β and γ values. Here, we estimated the impact of background sources and non-emission factors (transport, chemistry, and wet and dry depositions) on β and γ values and added two supplement figures (Figures S6 and S7) in Lines 105 – 143 in the revised supplement figure file. The supplement figure citation was updated in the manuscript. We also added "transport" in Line 241. Figures S6 and S7 show that the contributions of both background sources and non-emission factors to β and γ values are much more significant in low-anthropogenic-NO_x emission regions than high-anthropogenic-NO_x emission regions. In general, non-emission factors contribute more to the nonlinearity than background sources in low-anthropogenic-NO_x emission regions (Figures S7c and S7d) except for the first bin (of low local emissions) where background sources contribute more to the nonlinearity than non-emission factors at 10:00 – 11:00 LT. We added the discussion about the contributions of the two factors to β and γ values in Lines

231 – 237 and Lines 257 – 264.

Also, although the paper mentions the use of observed NO_3^- deposition trends to further support the declining trend of NOx emissions, it would be useful to incorporate more explicitly this information in the discussion.

Reply:

We mentioned nitrate wet deposition fluxes in the introduction in Lines 43 - 47 in the revised manuscript to support the decrease of NO_x emissions from the mid-2000s to the 2010s based on previous researches. Now we added a new supplement Figure S1 based on the National Acid Deposition Program (NADP) observations over the CONUS in Lines 75 – 79 in the revised supplement figure file, which shows a decrease (~ 30% - ~ 40%) of nitrate wet deposition fluxes from 2003 – 2017. In addition, we mentioned in Lines 376 - 378 in the revised manuscript that Silvern et al. (2019) used nitrate wet deposition fluxes in their analyses. Unlike the study of Silvern et al., which have multiyear simulation results and can compare model results with nitrate wet deposition flux observations, we ran 1-month simulation to show the nonlinearities among anthropogenic NO_x emissions, NO_2 surface concentrations, and NO_2 TVCDs to support the separation between urban and rural regions in our trend analyses. As discussed in Silvern et al. (2019), nitrate wet deposition fluxes are affected by both boundary NO_x and free-tropospheric NO_x, and most nitrate wet deposition flux sites are in rural regions. We didn't find any significant improvement from rural to urban regions when comparing nitrate wet deposition fluxes with coincident OMI-QA4ECV NO_2 TVCDs as shown in Figure R1 (Urban: TVCD = 1.13 × NADP + 0.13, R^2 = 0.84; Rural: TVCD =

1.49 × NADP – 0.11, R^2 = 0.82), which is a key point of our study. We suggest reading Silvern et al. paper for more details about nitrate wet deposition fluxes.

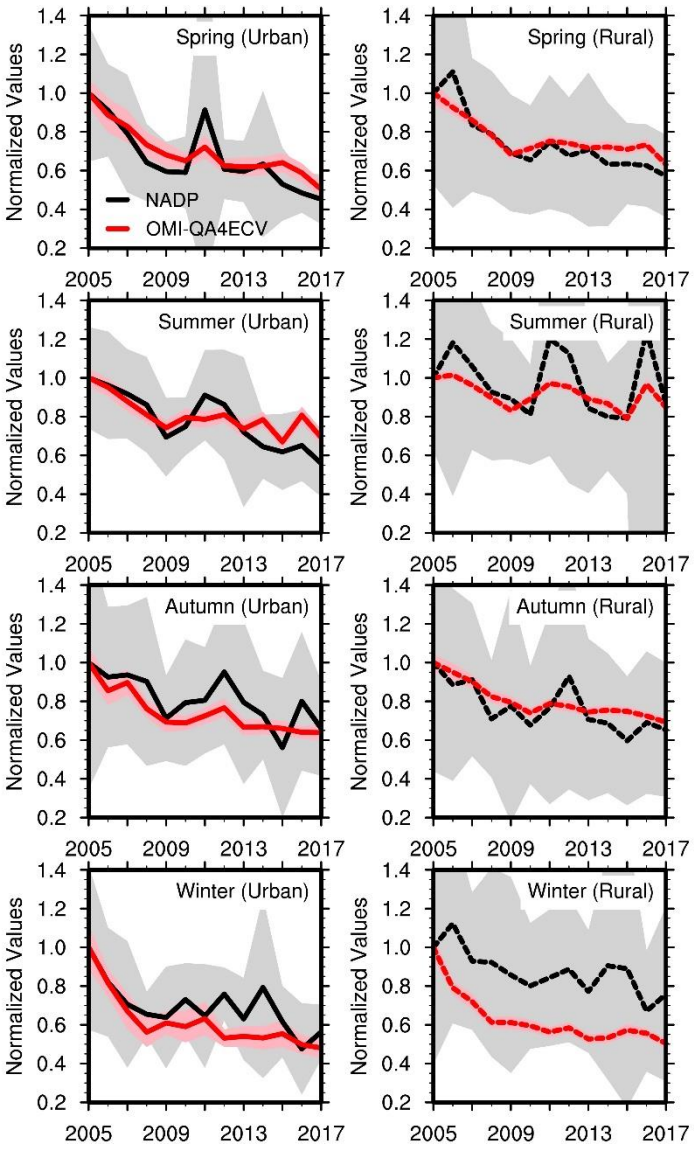

Figure R1. Relative annual variations of NADP nitrate wet deposition fluxes and coincident OMI-QA4ECV NO_2 TVCD in each season from 2005 – 2017 for urban (left panel) and rural (right panel) regions. The observation data are scaled by the corresponding 2005 values. Black and red lines denote NADP nitrate wet deposition fluxes and OMI-QA4ECV NO_2 TVCDs, respectively. Shading in a lighter color is added

to show the standard deviation of the results.

Additional (minor) comments:

- l. 34, the total of 0.24 Tg N for natural NOx emissions seems to be very low, where does it come from? I don't think NEI2014 provides this information. Please provide separately the soil, biomass burning and lightning emission information.

Reply:

Thank you for your suggestion. Unlike the natural NO_x sources from Seinfeld and Pandis (2016), which includes both lightning and soil NO_x emissions, NEI2014 only provides soil NO_x emissions calculated by the Biogenic Emission Inventory System (BEIS) but no lightning NO_x emissions (EPA, 2018). The 0.24 Tg N of natural NO_x emissions refers to soil NO_x emissions. We changed "anthropogenic and natural NO_x" to "anthropogenic and soil NO_x" in Line 35. And we provided soil and lightning NO_x emissions from REAM over the CONUS in July 2011 in Lines 112 – 115 in the revised manuscript.

- l. 64-65: there are earlier references for the effect of non-linearities on NO2 trends

Reply:

Yes, we added a citation of Lamsal et al. (2011). Please see Lines 65 – 66 in the revised manuscript.

- section 2.1 on REAM. What is the model domain?

Reply:

The model domain is shown in Figure 3, covering the CONUS. We added "the model domain of which is shown in Figure 3" in Line 95 in the revised manuscript to show the model domain.

- *l. 96: How is meteorology constrained by NCEP?*

Reply:

NCEP CFSv2 datasets provide initial and boundary conditions for our WRF simulation.

- *l. 100-102 it's a detail, but it seems a little strange that weekday emissions are based on NEI while weekend values are reduced. Isn't NEI an average?*

Reply:

Our NEI2011 emission inventory is from PNNL and has an initial horizontal resolution of 4 km. We re-gridded it to 36 km. The emission inventory was calculated by using the Sparse Matrix Operator Kernel Emissions (SMOKE) model which could produce hourly emissions for each day, thus could separate weekdays and weekends. We obtained only averaged weekday emissions from PNNL but no weekend emissions. Therefore, we scaled the weekend emissions based on previous studies (Beirle et al., 2003; Boersma et al., 2009; Choi et al., 2012; DenBleyker et al., 2012; Herman et al., 2009; Judd et al., 2018; Kaynak et al., 2009; Kim et al., 2016) and our model evaluations with observations. Currently, GEOS-Chem and CMAQ provide hourly anthropogenic emissions for each day for NEI2011 and NEI2014, respectively, such as NEI2014v2 at

https://www.acom.ucar.edu/Models/EPA/cmaq_cb6/all/. NEI2005 at ftp://aftp.fsl.noaa.gov/divisions/taq/emissions_data_2005/ also provides weekday, Saturday, and Sunday emissions separately.

- l. 105 what about lightning emissions?

Reply:

We described the method to calculate lightning NO_x emissions in Lines 107 – 112 in the revised manuscript.

- l. 148-149 the requirement that RCI > 50% is quite strict. What happens to the trends when you change that?

Reply:

When we changed the criterion to RCI < 100%, about 17% of seasonal data were removed. The following Figure R2 is for RCI < 100%. In Figure R3, we included all seasonal data with any RCI values. Generally, the trends of satellite NO_2 TVCDs over urban regions are still consistent with the trends of EPA NO_x emissions and surface NO_2 measurements in both Figure R2 and Figure R3, although there are some differences among Figure R2, Figure R3, and Figure 6 in the main manuscript. It emphasizes the selection of urban regions in trend analyses. Here, we would like to keep the RCI < 50% criterion in the main manuscript as it removes the effects of outliers.

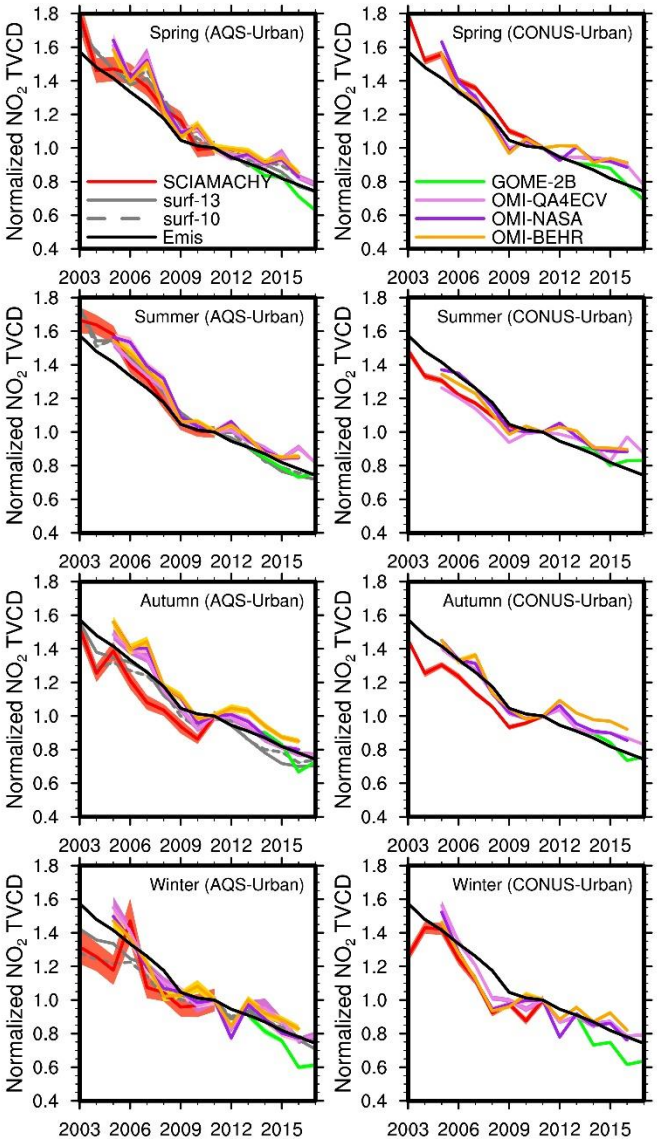

Figure R2. Same as Figure 6 in the main manuscript, but for RCI < 100%.

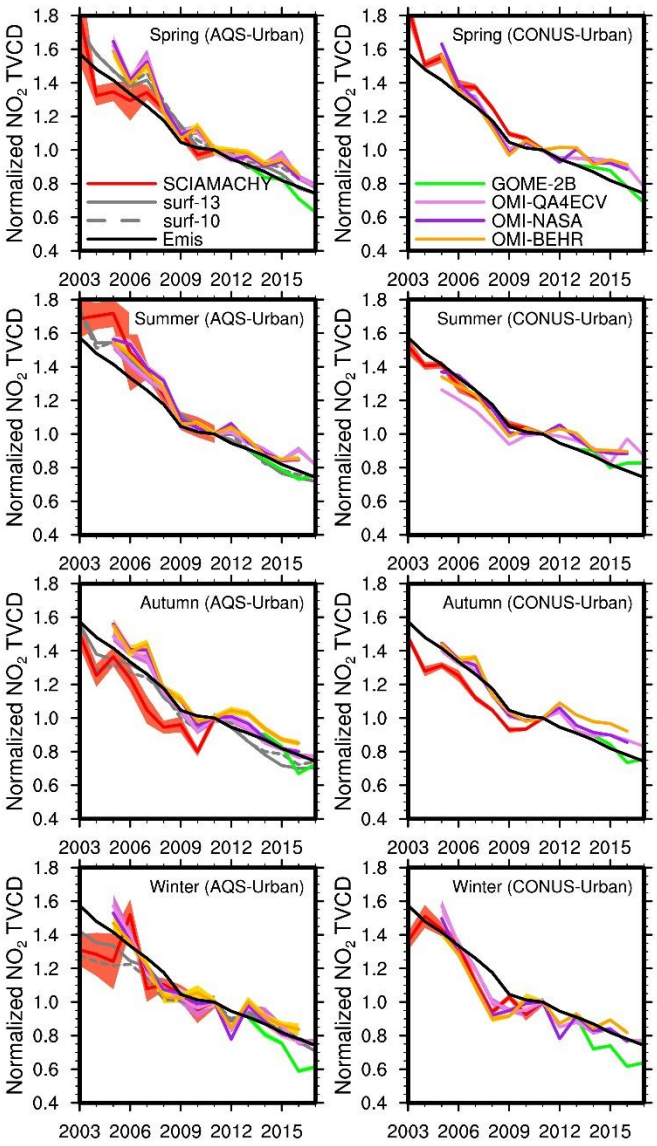

Figure R3. Same as Figure 6 in the main manuscript, but for all seasonal data with any RCI values.

- l. 184 how many measurements are rejected from this conditions on RCI?

Reply:

For surface concentrations, due to the completeness and stability of surface measurements, almost all seasonal averages (98.5%) satisfy the RCI < 50% criterion.

We added the information in Lines 194 – 195 in the revised manuscript.

- *l. 202 Are the β and γ calculated based on total emissions with or without lightning emissions? Lightning contributes significantly to the total column, but very little to surface concentrations (in part due to the vertical dependence of spaceborne instruments sensitivity).*

Reply:

Yes. Surface NO_2 concentrations are not much affected by NO_x in the free troposphere, which NO_2 in the free troposphere is an important component of NO_2 TVCD. We have discussed it in Lines 248 – 251. Both β and γ are calculated based on the emissions without lightning. The lifetime of lightning NO_x in the free troposphere is much longer than that in the boundary layer. As mentioned above, we added two supplement figures (Figures S6 and S7) to evaluate the contributions of different factors to β and γ values.

- *l. 229 "such as NOx transport from nearby regions" this is surprising since the calculated sensitivities were said to be purely local*

Reply:

In Lines 225 – 226 in the revised manuscript, we said, "Using this procedure, the effects of anthropogenic NO_x emission reduction were localized". It doesn't mean that transport effect is eliminated. Let's think about a simple example, to calculate β and γ values for a single grid cell "A", we only need to adjust the NO_x emissions of "A" but keep all other grid cells the same as before. By comparing two simulations, one with the original

emissions, the other one with grid cell “A” adjusted, we can obtain the β and γ values of “A”. Here, only the NO_x emissions of “A” are reduced in the adjusted simulation, and other grid cells are unchanged, so the emission reduction effect is localized. But transport still makes effects. Outfluxes from “A” to nearby grid cells will be different from the original simulation, as NO_x concentrations in “A” change. Our method described in Lines 216 – 225 in the revised manuscript can simulate the above procedure simultaneously for all grid cells and save computing time. This idea is different from a method widely used in previous studies by comparing one simulation with original emissions and the other one with emission reductions for all grid cells, where not only outfluxes from “A” change but also influxes to “A” are different from the original simulation. That is to say, the emission reductions of nearby grids are affecting grid cell “A”, which cannot be used to calculate local β and γ values.

l. 234 there is no "transport effect". β and γ are closer to 1 at 10-11 LT (compred to 13-14 LT) because of the weaker chemical losses.

Reply:

As we explained in the above answer, there are transport effects in the calculation of β and γ. In Line 234 in the original manuscript (Lines 251 – 253 in the revised manuscript), we were talking about the uncertainties of β and γ in each bin, and generally we don’t have enough evidence from Figure 2 to show that β and γ are closer to 1 at 10-11 LT compared to 13-14 LT.

l. 242 I suppose the "urban" definition depends on anthropogenic NOx emissions on a

specific year (and month maybe). This should be specified.

Reply:

Thank you for your suggestion. Yes, the definition is based on NEI2011, as described in Section 2.1, which provides annual average emissions for 2011 weekdays. We changed "anthropogenic NO_x emissions " to "anthropogenic NO_x emissions from NEI2011" to make it clear. Please see Lines 268 – 269 in the revised manuscript.

l. 330-332 Note that only 22 AQS sites (out of 179) are rural. Therefore, is the difference between this study and the results of Lamsal et al. and Jiang et al. really due to the selection of urban sites?

Reply:

Figure R4 shows the comparison between mean NO_2 concentrations from AQS urban sites and those from all (urban + rural) AQS sites, and there is no significant difference. Silvern et al. (2019) suggested that Jiang et al. (2018) included those sites with incomplete measurement records, which might be the reason why Jiang et al. (2018) had lower slowdown magnitude compared to our study (Table 1 in the main manuscript) and Silvern et al. (2019). The decreasing rates of AQS NO_2 concentrations in Lamsal et al. (2015) (Table 1 in the main manuscript) are smaller than our study and Silvern et al. (2019) (2005 – 2009: -6.6 ± 1.2% a^{-1}; 2011 – 2015: - 4.5 ± 1.7% a^{-1}), which might also be partly due to their different data processing procedure. We changed the original sentence, please see Lines 356 – 361 in the revised manuscript.

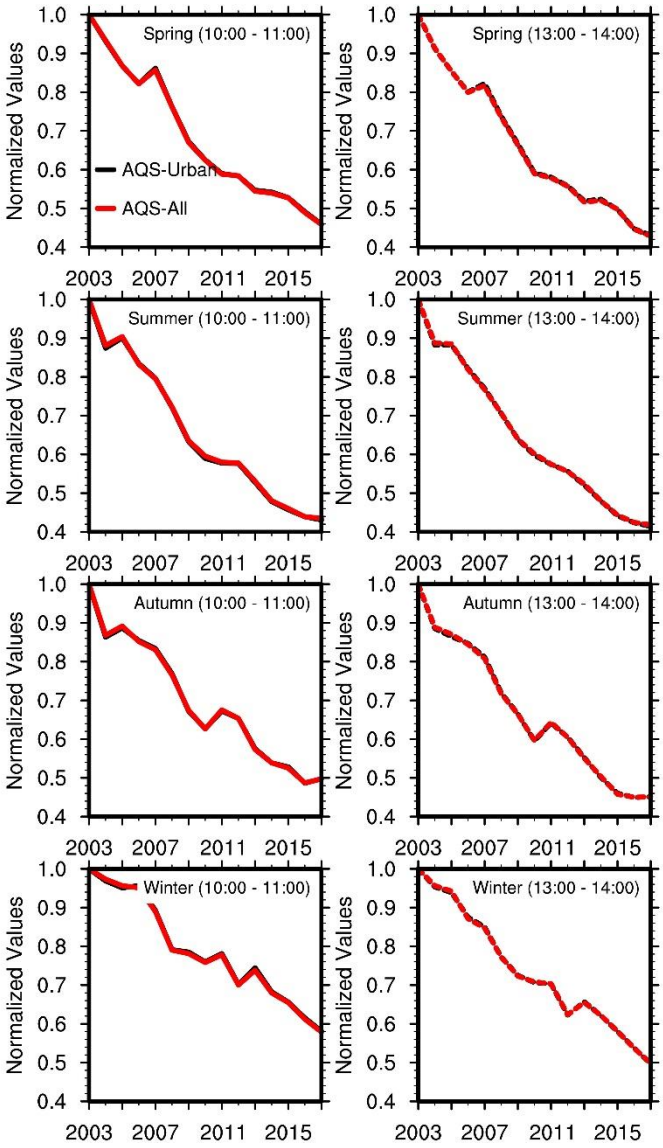

Figure R4. Relative annual variations of mean NO_2 surface concentrations from AQS sites. Black lines denote mean concentrations for only AQS urban sites, while red lines are for all AQS sites, including both rural and urban. The mean NO_2 concentrations are scaled by the corresponding 2003 values. The left column is for NO_2 concentrations at 10:00 – 11:00 LT, and the right column is for 13:00 – 14:00 LT.

l. 349-350 the sentence "They also identified model biases (...) natural emissions" is unclear, please either elaborate or delete.

Reply:

Silvern et al. (2019) shown that GEOS-Chem v11-02c underestimated NO_2 concentrations in the free troposphere compared to aircraft observations and satellite cloud-slicing results, which they thought was the reason why GEOS-Chem simulation results couldn't capture satellite NO_2 TVCD trends. We changed "natural emissions" to "missing natural emissions in the free troposphere" in Line 379 in the main manuscript.

- *l. 378-381 The nonlinear relationship of NOx with NO2 TVCD is important, but so are the effects of properly accounting for the background. The fact that spaceborne instruments have a low sensitivity close to the surface (i.e. the averaging kernels) is also important and deserves to be mentioned in this discussion.*

Reply:

Thank you for the suggestion. In this study, when we talk about nonlinearity (β and γ), we always mean any chemical and physical processes affecting the NO_2 TVCD and NO_2 surface concentrations, such as soil NO_x in the boundary layer and lightning NO_x in the free troposphere, chemistry, transport effect, and wet-dry depositions. We added other nonlinear factors in Lines 204 – 205 in the revised manuscript to make it clear. In Section 3.1, as mentioned above, now we have more discussion about the contributions of different factors to β and γ values. The low sensitivity of satellite sensors to the surface NO_x indeed emphasizes the selection of urban regions in inferring anthropogenic NO_x emissions from satellite datasets with more NO_x in the lower atmosphere compared to free troposphere to make the satellite signal meaningful to anthropogenic NO_x

emissions, but it is more related to the satellite measurement uncertainties which we have talked about in Lines 152 – 154 in the revised manuscript. We recommend reading Silvern et al. (2019) for more details about the vertical sensitivity of satellite sensors to NO_2 distributions.

Technical comments:

- in the title, "tend" should be "trend"

Reply:

Thanks. We corrected it.

- abstract line 15, add the word "bottom-up" (or "estimated') before "anthropogenic"

Reply:

The results shown in Lines 14 – 19 are based on the 1-month REAM simulation, where we indeed used the bottom-up NEI2011 emission inventory. However, the conclusions are widely applicable and not limited to NEI2011 or any other bottom-up emission inventories.

- l. 89 "mechanistic" (not "mechanical")

Reply:

Thanks. We corrected it. Please see Line 90 in the revised manuscript.

- l. 107 replace "measurements" by "sensors"

Reply:

We corrected it. Please see Line 117 in the revised manuscript.

- l. 109 add "instrument" after "SCIAMACHY"

Reply:

We added it. Please see Line 120 in the revised manuscript.

- l. 116 "These instruments measure transmitted, backscattered, and reflected radiation" is unclear

Reply:

We changed it to a simple sentence “These instruments measure backscattered solar radiation). Please see Line 127 in the revised manuscript.

- l. 126 "OMINO2" (not OMNO2")

Reply:

NASA OMI NO_2 TVCD products are named as OMNO2. Please refer to https://disc.gsfc.nasa.gov/datasets/OMNO2_V003/summary.

- l. 134 "choose" not "chose" (I guess)

Reply:

Thanks. We think it would be better to change “re-grid” to “re-gridded” in Line 145 in

the revised manuscript.

- *l. 208 add "the" before "model simulation"*

Reply:

Thanks. We added it. Please see Line 220 in the revised manuscript.

- *l. 279 -280 "sensitivities (...) to different anthropogenic NOx emissions over the CONUS" is confusing, please rephrase*

Reply:

Yes. We changed it to "We further investigate OMI-QA4ECV NO_2 TVCD relative annual variations from 2005 - 2017 over the regions with different anthropogenic NO_x emissions in Figure 5." Please see Lines 305 – 307 in the revised manuscript.

- *l. 325 insert "the" before "decreasing rates"*

Reply:

Thanks. We added it. Please see Line 353 in the revised manuscript.

- *References : use journal abbreviations, e.g. Atmos. Environ., etc.*

Reply:

Yes, we corrected it.

- *caption of Figure 5, line 672: specify the year (and month?) of the anthropogenic*

emissions used to define the groups

Reply:

Yes, we added it. Please see Line 705 in the revised manuscript.

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