Author's comments in reply to the anonymous referee for "Decadal changes in global surface NOx emissions from multi-constituent satellite data assimilation " by K. Miyazaki et al.

We want to thank the referee for the helpful comments. We have revised the manuscript according to the comments, and hope that the revised version is now suitable for publication. Below are the referee comments in italics with our replies in normal font.

Reply to Referee #1

The paper analyses the changes in NOx polluting emissions, by assimilating different constituents from different satellite instruments in the chemistry-transport model. The manuscript is clear and it has nice logic flow. Because there are several works dealing with emission estimation using satellite-based observations, I would recommend stressing the added value of this approach (for example already in the abstract and in the trend calculation in Sect. 5.4), i.e. the assimilation of non-NO2 observations as compared to previous work where only NO2 is assimilated in the system. I recommend publication after addressing the following specific and technical comments.

The impact of non-NO2 measurements is discussed more carefully in the revised manuscript. Table 5 has been added to discuss the impact of non-NO2 measurements on the a posteriori emissions. Linear trend estimations from the NO2-only assimilation have been included in Table 4. The following discussions have been added to Section 5.1:

"Table 5 compares the estimated emissions between the multiple-species data assimilation and a NO2-only data assimilation. The estimated emissions differ in many regions if non-NO2 data assimilation is considered because the ratio of predicted NOx emission and NO2 column has been adjusted by non-NO2 observations. The assimilation of non-NO2 measurements leads to changes of up to about 70 % in the regional monthly-mean emissions. The estimated ten-year total regional emissions for South America and Australia are about 10 % lower in the multiple-species assimilation than in the NO2-only assimilation. The RMSE between the two estimates for the monthly total regional emissions is 15.5 % for central Africa, 16.5 % for Australia, and about 5-8 % for major polluted regions during the ten-year period. The estimated monthly mean emissions are mostly smaller in the multiple-species assimilation than in the NO2-only assimilation, especially over the tropical and southern subtropical regions such as South America, central Africa, and Australia, suggesting that NO2-only data assimilation tends to overcorrect the emissions from the a priori. The monthly total global emissions decrease by up to 6 TgN (in boreal summer) if non-NO2 data assimilation is considered. The ten-year linear trend is also different over most industrial areas (Table 4). For instance, the positive trend for India is 34.3 %/decade

in the NO2-only assimilation, which is larger than the 29.2 %/decade in the multiple-species assimilation. For the United States, the negative trend is larger in the multiple-species assimilation (-29.4 %/decade) than in the NO2-only assimilation (-23.9 %/decade). These results confirm that the assimilation of measurements for species other than NO2 provides additional constraints on the NOx emissions over both anthropogenic and biomass burning regions."

P9 L17 The larger pixel size for GOME-2 and SCIAMACHY could indeed produce a dilution effect (lower NO2 level for larger pixel) compare to the smaller OMI pixel and thus, in principle, partially reduce the difference due to the different overpass time. Could you comment about that in the text?

The concentration of individual observations over polluted regions can vary between sensors, corresponding to the pixel size. However, because we employed the super observation approach and averaged multiple observations within a large super observation grid (i.e., about 2.8 degrees) before data assimilation, the influence of different pixel size should have a small impact on the data assimilation result, assuming the super observation grid is well covered by observation pixels. To clarify this point, the sentence has been rewritten as:
"Therefore, the differences in overpass time and also in pixel size could be the principle cause of the differences between the three different satellite retrievals, although the use of super observations for all the sensors reduces the influence of different pixel sizes."

Figure 2: It is quite difficult to distinguish the differences in these maps. It could be useful to show the differences compared to the observations in the second and third row, instead of the absolute tropospheric NO2 columns. It should help in highlighting the differences.

Figure 2 has been revised to show the differences.

P11 L19 Are there any known/expected differences in the ways of reporting, that you could mention here between the a priori and the EDGAR-HTAP emission databases?
P11 L29-30 Again, is there an expected reason to explain the similarity between EDGAR_HTAP and a posteriori emissions, relative to the a priori?

To our best knowledge, no comprehensive comparison has been made between these inventories.

P18 L18 You might want to refer here to this work about in plume chemistry effect: Vinken, G. C. M., Boersma, K. F., Jacob, D. J., and Meijer, E. W.: Accounting for non- linear chemistry of ship plumes in

the GEOS-Chem global chemistry transport model, Atmos. Chem. Phys., 11, 11707-11722, doi:10.5194/acp-11-11707-2011, 2011.

Added.

P18 L32-33 It is unclear for me what do you mean for "overcorrect". Do you mean that NO2-only gives too high emission values? According Table 3, the NO2-only data assimilation almost always (except South America) gives smaller values than the full assimilation. Could you clarify?

We have extended the sensitivity calculation using NO2-measurements only. The results confirm that the estimated regional emissions are mostly higher in the NO2-only assimilation than in the multiple-species assimilation. To clarify this, the sentence has been rewritten as follows:
"The estimated monthly mean emissions are mostly smaller in the multiple-species assimilation than in the NO2-only assimilation, especially over the tropical and southern subtropical regions such as South America, central Africa, and Australia, suggesting that NO2-only data assimilation tends to overcorrect the emissions."

Table 4 and section 5.4: Do these emission trends change when NO2-only assimilation is taken into account? I would include in Table 4 also the trends with NO2-only assimilation if the differences are sizeable.

The linear trends from the NO2-only assimilation have been included in Table 4 and discussed in the revised manuscript.

How your results reported in Table 4 and Fig.8 compare with those reported as NO2 tropospheric columns (OMI Standard Product not DOMINO as in you study) by Krotkov et al. (2016) in their Fig. 8?

The following sentence has been added:
"These year-to-year variations in the observed NO2 concentrations have previously been reported by Duncan et al. (2016) and Krotkov et al. (2016)."

It could be interesting also to compare your results in China and US to the results by Liu et al. (2016) in Table S2 of their supplement. Those results are not based on data assimilation but are based on satellite data only. Liu, F., Beirle, S., Zhang, Q., Dörner, S., He, K., and Wagner, T.: NOx lifetimes and emissions of cities and power plants in polluted background estimated by satellite observations, Atmos. Chem.

Phys., 16, 5283-5298, doi:10.5194/acp-16-5283-2016, 2016.

Thank you for the information. However, Liu et al (2016) estimated emissions sources at a 40 km × 40 km scale for point source areas (power plants and cities), which is about seven time higher resolution than that of our estimates. As the estimated emissions may be sensitive to the resolution, direct comparison with their results is difficult.

Technical corrections
P1 L6 biased -> biases

Corrected.

P2 L23 add reference Krotkov et al. 2016 here too

Added.

P2 L22 Kalam -> Kalman
P6 L32 GOME-II -> GOME-2

Corrected.

P7 L18-19 This needs reference

Added.

P7 L25 You might want to mention that those resolutions are valid in nadir direction only, but get bigger on the side of the swath and actually since 2008-2009 OMI row-anomaly doesn't allow complete daily global coverage.

The following sentence has been added:
"Since December 2009, approximately half of the pixels have been compromised by the so-called row anomaly, which reduced the daily coverage of the instrument."

Table 2 Australis -> Australia (and in the other tables too)
P14 L20 Los Angels -> Los Angeles

P20 L30 There are two dots at the end of the sentence

Corrected.

Table 4: Caption: OM ->OMI

Corrected.

Table 4 Is there a reason you put Table 4 before 5 and 6 but then you refer to Table 4 only in section 5.4, after mentioning 5 and 6? Please, clarify.

Table 4 is referred before Table 5 and 6 in the revised paper,