We wish to thank the referee for his/her helpful comments and for this positive acknowledgment of the improvements of our manuscript. The full review is copied hereafter and our responses are inserted in bold. We want to recall that our examples had been chosen to illustrate, for GMD, the parameters of the system configuration and the way the code works. We present a state-of-the-art system for regional inversions of reactive species emissions and we will provide fully comprehensive scientific study on NO_x and CO inversions in the near future and in dedicated journals.

The authors state that "the biases between OMI and simulated NO2 tropospheric columns are a complex topic (...) Addressing it properly is thus clearly out of the scope of this paper." This is true, but still, the inversion of emissions is expected to bring the model much closer to the observations even when the model and data are flawed. This article should show that the behavior of the inversion system is well understood, which I am not convinced.

In the NOx inversion E, the simulated NO_2 columns are increased by only about 10% over northern Germany and the Netherlands (based on Table 5 and Figure 6), despite large emission increments (>20%, possibly much more). Why this lack of sensitivity?

The inadequacies of the improvements found in this study are not related to our CHIMERE simulations only [Huijnen et al., 2010; Miyazaki et al., 2017; Souri et al., 2020; Elguindi et al., 2020]. For example, Miyakazi et al., [2017] found positive increments higher than 40% over parts of Western Europe. However, they do not improve the bias between the simulations and the OMI observations (mean bias of -0.45 10^{15} molec.cm^{-2} with and without data assimilation, see their Table 2).

We have performed a test to explain this lack of sensitivity. We have simulated the NO_2 tropospheric columns with biogenic emissions from MEGAN and the anthropogenic emissions from the TNO-GHGco-v2 inventory (called PRIOR in Figure 1). We have also simulated NO_2 columns with anthropogenic emissions increased by a factor 3 (called PRIORx3 in Figure 1). The ratio between these two simulations shows strong non-linearities, blurring the multiplicative effect of our increments and explaining the lack of sensitivity. By increasing NO_x anthropogenic emissions, NO_2 tropospheric columns can be strongly increased (Figure 1c) and can exceed the observations values for particular pixels (e.g., for 8 pixels in the purple box of our Figure 6 in the draft). NO_2 tropospheric columns can also be decreased, or only slightly increased as it is seen for example over rural areas over Spain (Figure 1c). On average, it tends to increase the concentrations by a factor that is much smaller than the factor of increase in the anthropogenic emissions. However, the patterns where the posterior tropospheric columns exceed the observations or, on the opposite are decreased or only slightly increased, explain why the inversion system does not attempt at increasing further the average level of the concentration (to decrease further the general bias to the observations), even though it accounts for the impact of non-linearities in the chemistry through the use of the M1QN3 minimization algorithm.

Several studies have reported that strong non-linear relationships exist between NO_x emissions and satellite NO_2 columns [Lamsal et al., 2011; Vinken et al., 2014; Li and Wang, 2019]. This reveals that a fully comprehensive scientific study is required, by

analyzing the NO_x lifetime through processes such as the NO_2+OH reactions and/or the reactive uptake of NO_2 and N_2O_5 by aerosols [e.g. Lin et al., 2012; Stavrakou et al., 2013].

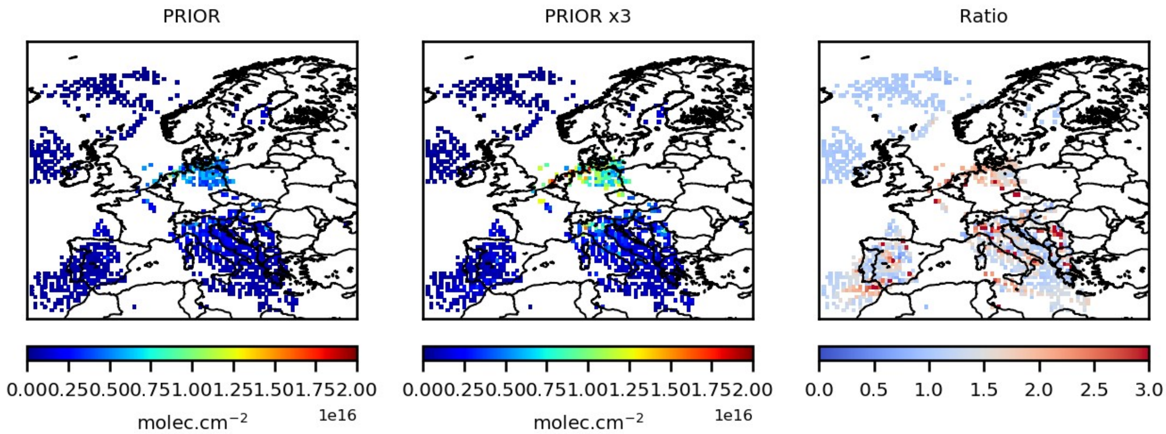

Figure 1. *NO_2 collocated tropospheric columns left) simulated by CHIMERE using the prior TNO-GHGco-v1 emissions, middle) simulated by CHIMERE using the prior TNO-GHGco-v1 emissions increased with a factor 3 in 10^{16}molec.cm^{-2} and right) ratio between these two fields, at the 0.5°x0.5° grid-cell resolution, the 19th, February 2015.*

It is stated on l. 476-482 that the discrepancies might have different causes including biases in the observations, in the emissions, and in the model. Nevertheless, the basic assumption of inverse modeling is that errors in the emissions play the dominant part. Therefore, we expect a substantial reduction of the bias after inversion, at least if the observations do not have huge uncertainties. What are the relative uncertainties in the NO_2 observations used here?

We agree. We normally expect a reduction of the bias after inversion if the observations do not have huge uncertainties. As seen in Figure 2a, the bias between the simulation and the observations is indeed reduced with posterior emissions, in particular when the OMI uncertainties are the lowest, over parts of Spain, Italy and northeastern Germany (Figure 2b). Nevertheless, the bias reduction is not as substantial as expected due to high non-linearities linked to the NO_x chemistry (as explained just above). Inferring NO_x emissions is therefore challenging.

We have added this Figure 2 in the manuscript (as Figure 9). We have also added details in the text: "Over this area (see the purple box in Figure 6), **where the OMI uncertainties are lower than 50% (Figure 9b),** the mean bias between the simulation and the observations has been reduced by about 24% when using the posterior emissions (mean bias of 1.9x10^{15} molec.cm^{-2} against 2.6x10^{15} molec.cm^{-2} with the prior emissions, Table 5**, Figure 9a).**

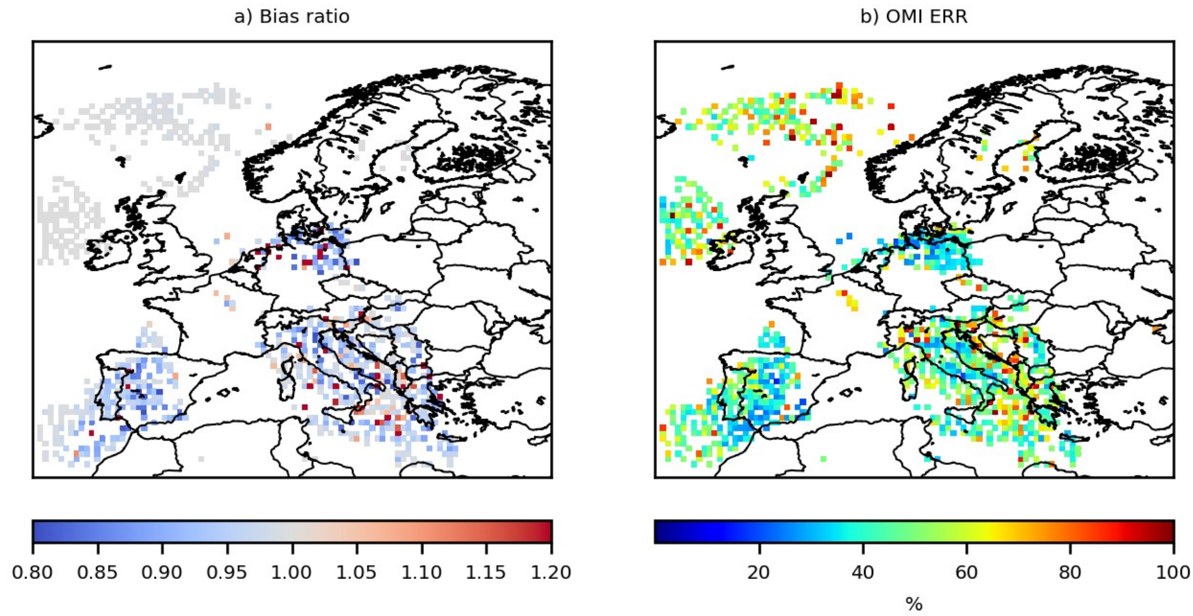

Figure 2. *a) Bias ratio between CHIMERE simulations using the posterior emissions against prior TNO-GHGco-v1 emissions compared to the OMI-QA4ECV-v1.1 observations. All ratios lower than 1, in blue, demonstrate that posterior emissions improve the simulation compared to the prior ones. b) OMI uncertainties, in %, the 19th, February 2015.*

Given the relatively long NOx lifetime in winter, there could be a strong dependence of the columns on the emissions during the preceding days. Does the system account for that?

As presented in the study with 1-day inversion for NO_x, the system does not account for the dependence of the columns to emissions of preceding days. We agree that the assimilation window will be widened in the near future. Nevertheless, as suggested by the sensitivity test with a 3-day inversion in Figure 3, this cannot explain the small reduction of the bias after inversion.

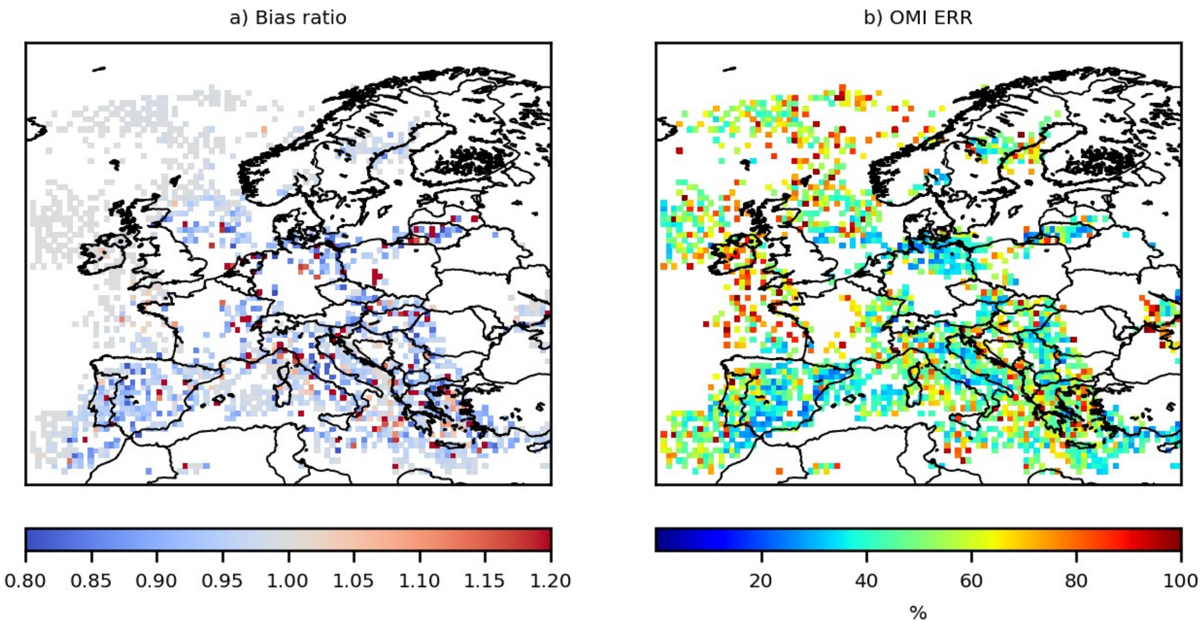

Figure 3. *Same as Figure 2 but for 3 days from 19th to 21st February 2015.*

The fact that only few iterations were needed to reach near-convergence (Table 3) indicates that the errors in the observations are very high, and this could partly explain the poor performance of the inversion. Please clarify.

We do not agree with this statement. The fact that only few iterations were needed to reach near-convergence does not indicate that the errors in the observations are very high (as seen in Figure 2b). It only indicates that M1QN3 easily reaches the vicinity of the minimum of the cost function (as seen in Figure 4).

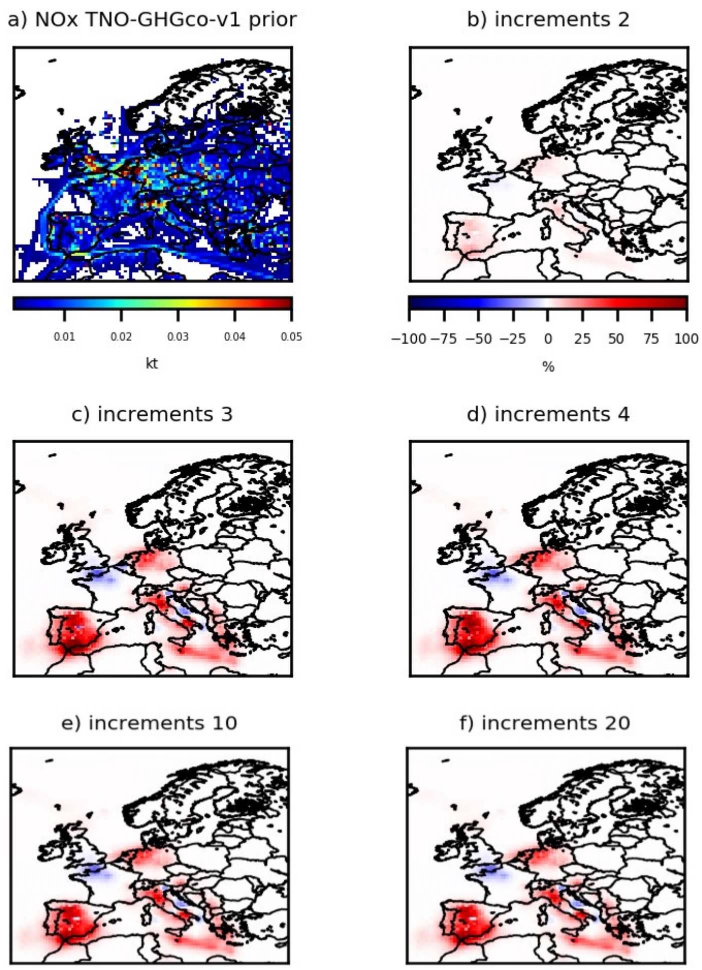

Figure 4. *a) TNO-GHGco-v1 NO_x anthropogenic prior emissions, in ktNO_2/grid-cell and increments provided by the inversion at different stages of the inversion b) during the 2sd forward-adjoint cycle, c) the 3rd, d) the 4th, e) the 10th and f) the 20th, with constraints from OMI the 19th, February 2015, in %.*

Something makes it difficult for the inversion system to reproduce the observations. All I ask is that the cause(s) for that inability are identified.

As shown with different sensitivity tests here, the difficulty of the inversion system to reproduce the observations is mainly explained by strong non-linear relationships existing between NO_x emissions and satellite NO_2 columns.

We have added a sentence at the end of the abstract: "We reported strong non-linear relationships between NO_x emissions and satellite NO_2 columns, now requiring a fully comprehensive scientific study."

We have added sentences in the text about the likely causes explaining the discrepancies between simulations and satellite observations at the end of the subsection 4.2.4: "We have performed a test to explain this lack of sensitivity. We have simulated NO_2 columns with anthropogenic emissions increased by a factor 3 compared to the simulation in Figure 6a. The ratio between these two simulations shows strong non-linearities, blurring the multiplicative effect of our increments and explaining the lack of sensitivity (not shown). By increasing NO_x anthropogenic emissions, NO_2 tropospheric columns can be strongly increased and can even exceed the observations values for particular pixels. NO_2 tropospheric columns can also be decreased or only slightly increased. On average, it tends to increase the concentrations by a factor that is much smaller than the factor of increase in the anthropogenic emissions. However, the patterns where the posterior tropospheric columns exceed the observations or, on the opposite are decreased or only slightly increased, explain why the inversion system does not attempt at increasing further the average level of the concentration (to decrease further the general bias to the observations), even though it accounts for the impact of non-linearities in the chemistry through the use of the M1QN3 minimization algorithm. The biases between OMI and simulated NO_2 tropospheric columns are a complex topic that is not related to our CHIMERE simulations only [Huijnen et al., 2010; Souri et al., 2020; Elguindi et al., 2020]. **Several studies have indeed already reported that strong non-linear relationships exist between anthropogenic NO_x emissions and satellite NO_2 columns [Lamsal et al., 2011; Vinken et al., 2014; Miyazaki et al., 2017; Li and Wang, 2019]. This reveals that a fully comprehensive scientific study is required, by analyzing the NO_x lifetime through processes such as the NO_2+OH reactions and/or the reactive uptake of NO_2 and N_2O_5 by aerosols [e.g. Lin et al., 2012; Stavrakou et al., 2013]. "**

We also added sentences in the conclusion of our paper: "We show the potential of PYVAR-CHIMERE, with inversions for CO and NO_x illustrated over Europe. PYVAR-CHIMERE will now be used to infer CO and NO_x emissions over long periods, e.g. first for a whole season or year and then for the recent decade 2005-2015 in the framework of the H2020 VERIFY project over Europe, and in the framework of the ANR PolEASIA project over China, to quantify their trend and their spatio-temporal variability. **Nevertheless, as we have reported strong non-linear relationships between NO_x emissions and satellite NO_2 columns, a fully comprehensive scientific study is required, by analyzing the NO_x lifetime through processes such as the NO_2+OH reactions and/or the reactive uptake of NO_2 and N_2O_5 by aerosols [e.g. Lin et al., 2012; Stavrakou et al., 2013]. Biogenic emissions will be also further studied to better understand the relationship between NO_x emissions and NO_2 spaceborne columns."**

References

Lamsal, L. N., Martin, R. V., Padmanabhan, A., van Donkelaar, A., Zhang, Q., Sioris, C. E., Chance, K., Kurosu, T. P., and Newchurch, M. J.: Application of satellite observations for timely updates to global anthropogenic NO_x emission inventories, *Geophys. Res. Lett.*, 38, L05810, doi:10.1029/2010GL046476, 2011.

Li, J. and Wang, Y.: Inferring the anthropogenic NO_x emission trend over the United States during 2003–2017 from satellite observations: was there a flattening of the emission trend after the Great Recession?, Atmos. Chem. Phys., 19, 15339–15352, https://doi.org/10.5194/acp-19-15339-2019, 2019.

Lin, J.-T., Liu, Z., Zhang, Q., Liu, H., Mao, J., and Zhuang, G.: Modeling uncertainties for tropospheric nitrogen dioxide columns affecting satellite-based inverse modeling of nitrogen oxides emissions, Atmos. Chem. Phys., 12, 12255–12275, https://doi.org/10.5194/acp-12-12255-2012, 2012.

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

Stavrakou, T., Müller, J.-F., Boersma, K. F., van der A, R. J., Kurokawa, J., Ohara, T., and Zhang, Q.: Key chemical NO_xsink uncertainties and how they influence top-down emissions of nitrogen oxides, Atmos. Chem. Phys., 13, 9057–9082, https://doi.org/10.5194/acp-13-9057-2013, 2013.

Vinken, G. C. M., Boersma, K. F., Maasakkers, J. D., Adon, M., and Martin, R. V.: Worldwide biogenic soil NO_x emissions inferred from OMI NO_2 observations, Atmos. Chem. Phys., 14, 10363–10381, https://doi.org/10.5194/acp-14-10363-2014, 2014.