

Review #2

We wish to thank the referee for his/her helpful comments. His/her full comments are copied hereafter in normal black font, and our responses are inserted in between in blue and bold font.

This manuscript presents NO_x emission inversion experiments driven by two different satellite NO₂ column observational data sets. By performing inversions for 2019 using prior emissions for 2005, this study addresses the question to what extent bottom-up reported NO_x emission reductions can be reconciled using satellite NO₂ observations in an emission inversion. This study is an important contribution to the literature and is well within the scope of ACP, with the community increasingly focusing on reconciling top-down and bottom-up emission estimates. The methodology is mostly scientifically sound, but the presentation of the methodology and results in the manuscript is unclear. This paper can be considered for publication in ACP after a revision focused primarily on restructuring the manuscript and improving the clarity of writing. Please find below the specific issues to be addressed in a revised manuscript.

We thank the referee for his/her positive general comments.

Methodological issues:

- While I agree with the authors that soil emissions are a relatively minor term on annual time scales, this is not the case for the spring and summer months when biogenic and agricultural emissions peak and anthropogenic emissions are lowest. Previous studies (e.g., Silvern et al. 2019) found that accounting for soil emissions is important to reconcile simulated and observed NO₂ column trends. Please discuss the effect of omitting agricultural soil emissions on the estimated NO_x emission reduction trends.

We agree with the reviewer about the importance of accounting for soil emissions to reconcile simulated and observed NO₂ column trends, as shown by Silvern et al. (2019). This is in agreement with our study, published in 2021, confronting trends in OMI NO₂ tropospheric columns over the 10-year 2008–2017 period in Europe to those of tropospheric columns simulated with the regional chemistry-transport model CHIMERE and state-of-the-art emission estimates for anthropogenic and biogenic NO_x emissions (Fortems-Cheiney et al., 2021).

Actually, we do take soil emissions from MEGAN into account, as explained in Section 2.2. The impact of agriculture on soil emissions is neither provided by the TNO inventory nor by MEGAN. Therefore we do not include a specific agricultural soil NO_x emissions component in our prior estimation of the NO_x soil emissions. However, the atmospheric inversions controls these soil emissions (separately from the anthropogenic emissions) and could have increased them if it had supported the decrease of the misfits to the satellite observations above agricultural areas, which was not the case. As stated in Section 3.4,

« the annual budget for biogenic emissions are only changed by a few percent both by the OMI and TROPOMI inversions. »,

In this study, the analysis are focused on the quantification of the decrease of NO_x European emissions in 2019 compared to 2005. As the anthropogenic emissions strongly contribute to the total NO_x emissions in Europe even in summer, we assume that the differences between the 2005 and the 2019 budgets are mainly due to anthropogenic emissions and not to the biogenic ones. Finally, the level of distinction between the anthropogenic and biogenic emissions in the inversions is one of the sources of uncertainty in the estimate of the anthropogenic emissions: as such, it is indirectly assessed together with other sources of uncertainty in the evaluation with comparisons to the TNO inventory.

We have added these information in the conclusion: “As the anthropogenic emissions strongly contribute to the total NO_x emissions in Europe even in summer, we assume that the differences between the 2005 and the 2019 budgets are mainly due to anthropogenic emissions and not to the biogenic ones. However, the level of distinction between the anthropogenic and biogenic emissions in the inversions is one of the sources of uncertainty in the estimate of the anthropogenic emissions. The corrections provided by the inversions to the prior emissions can also be limited by the cloud coverage affecting the OMI or TROPOMI observations, by errors in the OMI or TROPOMI data and by errors in the CTM. Finally, the set-up of error covariance matrices could also have a strong impact on the emission estimates resulting from the inversions.”

- In my view, this study is missing a section on the performance of the CIF-CHIMERE system using the two assimilated datasets. Please include this, e.g. by presenting error reductions or a comparison with independent (surface) observations.

The derivation of the statistics of uncertainty in the emission estimates from the inversion (“the posterior uncertainty”), and thus of the “error reduction” (the relative difference between prior and posterior uncertainties) is highly challenging when solving high dimensional inversion problems in variational mode (Rayner et al. 2019). In principle, the posterior uncertainty can be derived approximately using a Monte Carlo approach (Chevallier et al. 2007). However, because of the large computational cost of such a Monte Carlo framework, we have chosen not to perform it.

The comparison between our type of regional CTM (here at 0.5° resolution) and typical in situ air quality observations in view to evaluate the emission estimates, and consequently the validation of our inversions using such data or the assimilation of such data can be complex due for example to the difficult representation of these observation close to the surface and to source in the model. In addition, as described by Lamsal et al. (2008), the instrument most commonly used for routine measurements of NO_2 is a chemiluminescence analyzer, a measurement technique which exhibits significant interference from other reactive oxidized nitrogen-containing species (NO_z) such as peroxyacetyl nitrate (PAN) and nitric acid (HNO_3). It could consequently result in an overestimation of ambient NO_2 concentrations (Savas et al. 2023). Specific studies are

needed to properly handle such a use of the in situ observations, which explains why we have a long list of publications focusing on the use of the satellite data (see our introduction, van der A et al., 2008; Lamsal et al., 2011; Mijling et al., 2012; Mijling et al., 2013; Miyazaki et al., 2017; Plauchu et al., 2024; van der A et al., 2024).

Furthermore, here, the analysis are focused on the assessment of the ability to provide information on the decrease of NO_x anthropogenic emissions in 2019 compared to 2005 based on satellite NO₂ observation (that from the OMI and TROPOMI instruments). Given their spatial coverage and spatial resolution, and their vertical representativity, these satellite observations— are likely more suitable for inversions at the targeted scales than the surface measurements in the area of analysis. This rationale is now better expressed with our new title and in our revision of the introduction.

Chevallier, F., F.-M. Bréon, and P. J. Rayner (2007), Contribution of the Orbiting Carbon Observatory to the estimation of CO₂ sources and sinks: Theoretical study in a variational data assimilation framework, *J. Geophys. Res.*, 112, D09307, doi:[10.1029/2006JD007375](https://doi.org/10.1029/2006JD007375).

Lamsal, L.N.; Martin, R.V.; van Donkelaar, A.; Steinbacher, M.; Celarier, E.A.; Bucsela, E.; Dunlea, E.J.; Pinto, J.P. Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument. *J. Geophys. Res.* 2008, 113, D16308.

Rayner, P. J., Michalak, A. M., and Chevallier, F.: Fundamentals of data assimilation applied to biogeochemistry, *Atmos. Chem. Phys.*, 19, 13911–13932, <https://doi.org/10.5194/acp-19-13911-2019>, 2019.

Writing issues:

- The title does not reflect the contents of the paper: TROPOMI observations were not available in 2005, and the paper does not focus on detecting NO_x emission decreases but rather on reconciling top-down- and bottom-up-derived emission trends. Please revise.

We agree that the title of the article was potentially misleading and we have changed it. It is now: “Assessing the ability to quantify the decrease of NO_x anthropogenic emissions in 2019 compared to 2005 using OMI and TROPOMI satellite observations”.

- Methodology: there is little coherence between these subsections.

Explanations have been added at the beginning of Section 2 to show the coherence between the subsections: “The principle of our traditional atmospheric inversion approach is to correct « a priori » estimates of the emission maps, also denoted “prior emissions”, to reduce differences between atmospheric observations and their simulations with a chemistry-transport models (CTMs) fed with the estimates of emission maps. It relies on a specific Bayesian inversion algorithm, on satellite observations of the atmospheric densities of NO₂, and on a regional CTM, as shown in Figure 1. We detail these different components of our inversion framework in Section 2: the CIF-CHIMERE variational

inversion system, the prior estimates of the NO_x emissions in Europe for the year 2005, the configuration of the CHIMERE CTM and of its adjoint code for the simulation of NO₂ concentrations and of their sensitivity to the emissions estimates, and the OMI-QA4ECV and TROPOMI satellite observations for the year 2019. The process ensuring a suitable comparisons between the simulations and the satellite observations -the aggregation of the observations into super-observations and the application of the averaging kernels from the satellite retrievals to the vertical columns of the model simulations - are also explained. Finally, we provide details about our configuration for the inversion of NO_x emissions over Europe.”

The title of Section 2.4.3 has also been changed for clarity, it is now « Choices made to ensure a consistent comparison between simulated and observed NO₂ TVCD » and no more « Comparison between simulated and observed NO₂ TVCDs ».

- Please consider giving more descriptive names to your inversion experiments and including a conceptual diagram summarizing your methods.

As recommended, the names of the experiments have been changed in the text and in the figures. We now use the terms “optimistic” or “conservative”, respectively when the super-observation uncertainty is reduced compared to that of individual observations or not. Due to these new names, few sentences have been slightly changed in Section 3.3 to simplify the reading.

We have also included a diagram providing an overview of the inversion and experimental frameworks, and of the results in the new Figure 1.

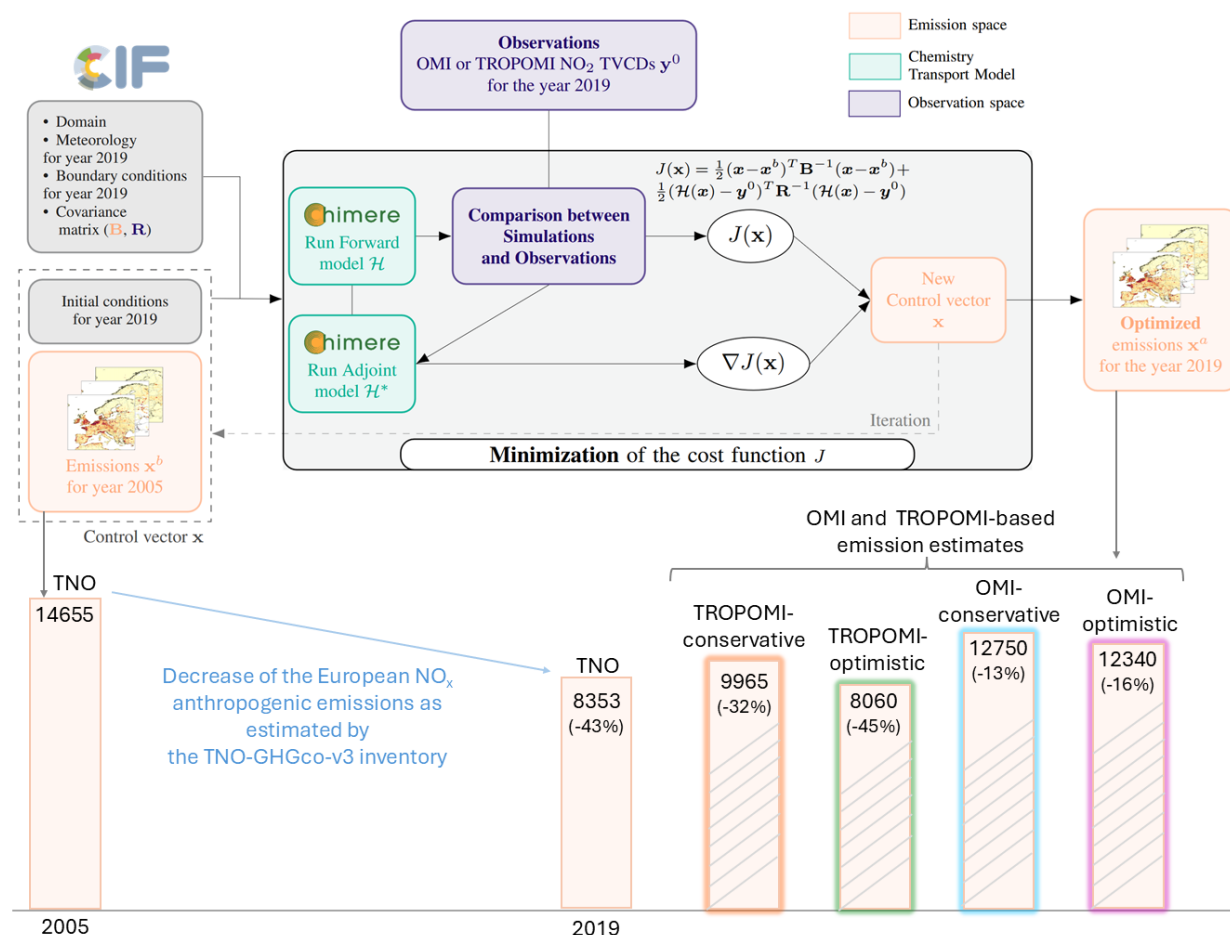


Figure 1. Simplified scheme of the iterative minimization in the CIF-CHIMERE inversion system and illustration of the decrease of the European NO_x emissions estimated both by TNO and by the OMI and TROPOMI-based inversions. The different inversions are described in Table 1. The numbers express the anthropogenic NO_x emission estimates for the EU-27+UK area in kTqNO_2 . The numbers in brackets express the difference of the anthropogenic NO_x emission estimates for the EU-27+UK in 2019 compared to 2005, in %.

Minor issues:

Introduction: please introduce non-anthropogenic NO_x emissions before the first use of biogenic emissions in line 55.

This has been done.

Section 2.1: please add more detail on the CIF-CHIMERE setup. Since NO_x chemistry is non-linear, how do NO_x emission changes impact NO_2 columns via the chemistry scheme?

Our inversion system provides the advantages of simulating NO_2 concentrations and the sensitivities of NO_2 tropospheric columns to surface emissions at a relatively high spatial resolution with a chemistry scheme, based on the Eulerian regional CTM CHIMERE and the MELCHIOR-2 chemical scheme, and their adjoint codes.

The MELCHIOR-2 chemical scheme used here indeed accounts for non-linear relationships between NO_x (NO and/or NO₂) emission changes and NO₂ TVCDs via reactions with hydroxyl (OH) radicals but also with other direct or indirect NO_x sinks associated with other species, such as ozone (O₃) or the HO₂ radical. These information have been added in Section 2.3.

L217-218: what is the goal of reprocessing the NO₂ column observations using meteorological data?

These goals are explained in the user's guides of OMI and TROPOMI retrievals. For example, according to Lambert et al. (2024), the TROPOMI NO₂ retrieval explicitly accounts for temperature effects by using the co-located temperature profiles in troposphere and stratosphere from the ECMWF meteorological analyses in the DOAS retrieval of the slant column density.

Lambert, J.-C., A. Keppens, S. Compennolle, K.-U. Eichmann, M. de Graaf, D. Hubert, B. Langerock, A. Ludewig, M.K. Sha, T. Verhoelst, T. Wagner, C. Ahn, A. Argyrouli, D. Balis, K.L. Chan, M. Coldewey-Egbers, I. De Smedt, H. Eskes, A.M. Fjæraa, K. Garane, J.F. Gleason, F. Goutail, J. Granville, P. Hedelt, K.-P. Heue, G. Jaross, Q. Kleipool, M.L. Koukouli, R. Lutz, M.C. Martinez Velarte, K. Michailidis, A. Pseftogkas, S. Nanda, S. Niemeijer, A. Pazmiño, G. Pinardi, A. Richter, N. Rozemeijer, M. Sneep, D. Stein Zweers, N. Theys, G. Tilstra, O. Torres, P. Valks, J. van Geffen, C. Vigouroux, P. Wang, and M. Weber : Quarterly Validation Report of the Copernicus Sentinel-5 Precursor Operational Data Products #24: April 2018 – August 2024. S5P MPC Routine Operations Consolidated Validation Report series, Issue #24, Version 24.00.00, 212 pp., 16 September 2024

L350-351: wouldn't OMI also underestimate columns in polluted areas, due to the coarse-resolution a priori NO₂ profiles? Why is this problem larger for TROPOMI?

Indeed, it is probable that OMI also underestimates NO₂ columns in polluted areas, as OMI and TROPOMI both use the same coarse-resolution a priori NO₂ profiles. Nevertheless, these coarse-resolution a priori NO₂ profiles can have a stronger impact on the TROPOMI columns than on the OMI ones, as TROPOMI has a finer spatial resolution than OMI.

OMI vs TROPOMI evaluations have shown that TROPOMI provides smaller NO₂ TVCDs compared to OMI over polluted areas, at least with previous TROPOMI versions (Sekiya et al. 2022 ; Lambert et al. 2021). In addition, recent evaluations of the TROPOMI RPRO v2.4 product against MAXDOAS observations still indicate significant biases of TROPOMI NO₂ TVCDs of typically -40% over highly polluted areas (Lambert et al., 2023; van Geffen et al., 2022b).

The differences between OMI and TROPOMI NO₂ TVCDs probably come from differences in their retrieval algorithms, for example from different cloud pressure retrievals or from different albedo datasets, as detailed in Section 2.4.

L355-360: please explain better what is meant with ‘an indirect comparison of OMI and TROPOMI using the CHIMERE CTM as an intermediate’. I am also missing this paragraph’s key message and contribution to the subsection.

OMI and TROPOMI retrievals do not perfectly coincide in terms of spatio-temporal and vertical representativeness. One can attempt to compare, directly, retrievals from OMI and TROPOMI whose footprints overlap and whose observation time is close, ignoring the differences in terms of vertical sensitivity (AKs). But a more suitable “indirect” comparison can be made by confronting OMI retrievals to CHIMERE in one hand, and TROPOMI retrievals to CHIMERE in the other hand (in both cases: applying the corresponding AKs to CHIMERE), and analysing whether the average signs and levels of differences to CHIMERE are consistent when analysing OMI vs. TROPOMI dataset. This indirect comparison using the CHIMERE CTM as an intermediate benchmark highlights the impact of the different OMI and TROPOMI AKs on the CHIMERE simulations.

These explanations have been added in Section 3.1: “Since the OMI and TROPOMI AKs are different, and since the spatio-temporal samplings of the two data sets are also different, a direct comparison between the OMI and TROPOMI datasets could be complex or misleading. The confrontation of the OMI retrievals to CHIMERE in the one hand, and of the TROPOMI retrievals to CHIMERE in the other hand can be used as an indirect but more suitable comparison between the OMI and TROPOMI datasets.”

L394-395: this seems specific to the experiment setup with a missing agricultural soil emission term.

See our answer to the general comment about the biogenic emissions.