Review 2

The paper by Plauchu et al. entitled 'NOx emissions in France in 2019-2021 as estimated by the high spatial resolution assimilation of TROPOMI NO2 observations' explores the use of TROPOMI NO2 data in combination with an inversion framework to estimate the reduction of NOx emissions due to COVID-19 lockdowns in France in 2020, with a focus on several large urban centres. The paper is very comprehensive and well-structured throughout. As the authors note, the initial results show a somewhat limited correction of the posterior by the observations due to several possible reasons. The authors then try two alternative setups that allow for a larger influence of the observations on the posterior emissions. The authors highlight further data needs and developments that could help improve the use of inversions for case studies such as these.

General comments

- Interesting and relevant addition to existing literature.
- Comprehensive description of the methodology.
- Overall, the paper is well written.

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 bold font.

Specific comments

• 281-284: Can you comment on the expected influence of using 2016 prior emissions for this inversion study? If prior emissions (for 2016) are systematically higher (~13% based on CITEPA) compared to 2019 or 2020, could this have an effect on the posterior emissions? What if you would have used 2019 prior emissions?

Our study reveal the large weight of the prior estimate of the emissions on the inversion results, at least at national scale, driven by the the respective amplitude of the prior uncertainty and observation error covariance matrices (B and R), but also by the limited satellite coverage which is not compensated by an extrapolation of the information from the observations via spatial and temporal correlations in B. Therefore, in a general way, using prior emission estimates derived from inventories for the year 2019 or 2020 rather than for the year 2016 would have led to significantly different inversion results, with lower emission values on average. However, it is difficult to make assumptions on the amplitude of these differences, which would highly vary as a function of the scales and locations, and on their impact on the inter-annual variations of the emissions between 2019 and 2021.

In particular, in experiments when reducing the amplitude of R and when focusing the analysis on cities where the signal from the anthropogenic emission is large, and on periods when the satellite coverage is good, the weight of the prior emission estimate is lower.

 369-372: Could the differences between cities also be partly due to different contributions of (heavy) industry to NOx emissions in or around these urban centres?

The INS data do not show major differences in terms of sectoral distribution between the main French cities. In all of them, more than 80 % of the emissions are due to 3 non-industrial sectors: road transport, non-industrial combustion and other mobile sources and machinery. The weight of the industrial sector (combustion in the manufacturing industry and combustion in the energy and energy transformation industry) may be higher for some cities, but these cities are not those where the slopes are the smallest. For Bordeaux, Nice, Nantes and Toulouse (cities where the decline between spring 2019 and spring 2020 is small), the share of total emissions by the industrial sector is 4.93 %, 7.15 %, 11.67 % and 11.3 % respectively.

For Paris, Lyon, Marseille and Lille (cities where the decrease between spring 2019 and spring 2020 is higher) this share is 16.55 %, 15.62 %, 10.49 % and 8.61 % respectively.

We have added a sentence in the manuscript: "These differences between cities cannot be explained by different contributions of industry to NOx emissions in or around these urban areas as INS data do not show major differences in terms of sectoral distribution between the main French cities".

Actually, the differences between cities appear to be driven by the strength of the TROPOMI signal over those cities. As depicted in Figure C5, the difference between TROPOMI NO2 TVCD over those cities between March and April 2019 and March and April 2020 is smaller than cities like Lille, Lyon, Nantes and Paris.

We have added this sentence in section 3.2.1: "The strength of the TROPOMI NO2 signal differs over these 8 French cities. Indeed, the absolute changes in NO2 TVCD in March/April 2020 compared to March/April 2019 are higher for Paris, Lille, Lyon and Nantes (see Figure C5 in Supplementary materials) than for Bordeaux, Marseille, Nice and Toulouse (i.e., 1.×10¹⁵ molec.cm⁻² or less)".

• 422: "yielding a more accurate estimate of the COVID-19 delta". The qualification of the alternative method using filtered observations being more accurate has not been made before in the paper. Can you comment on why you conclude that it is more accurate than the initial estimate? Could this lesson be generalized to other studies looking at emission variations at such high spatial and temporal resolution?

This part is not about an alternative method, but about a different way of analyzing the results as long as we do not have stronger insights on the spatial and temporal correlations of the uncertainties in the gridded emission inventories, which we now have better emphasized.

The "accurate" adjective was not the right one. We have replaced the sentence: "... yielding a characterization of the COVID-19 effects which is more consistent with the changes in emissions estimated by the CITEPA".

As detailed in section 3.2.4, some parts of France, including urban areas, suffer from the lack of observations during the COVID-19 period. Locally, for the days when no observation is available, the posterior emission estimates remain close to the prior ones. When considering the budget of the emissions over all days (with and without observations), the amplitude of the corrections to the prior estimate of the emissions driven by the satellite observations is artificially decreased by the lack of corrections during days when there is no satellite observations. Therefore, we filter out these days to better identify the corrections driven by the satellite.

The real alternative method to support a robust direct diagnostic of the monthly to annual emission budgets and changes would be to characterize in the B matrix the actual spatial and temporal correlations of the uncertainties in the gridded inventories with hourly variations used as prior estimate of the emissions by the inversions. Such a characterization would support the extrapolation in space and time of the information obtained locally and for some days from the satellite observation. However, getting suitable insight on such correlations is challenging since the usual correlation models based on assumptions of isotropy, homogeneity in space and time, and of decrease as a function of distance and time likely poorly match the actual derivation and structures of gridded inventories convolved with typical temporal cycles at diurnal to seasonal scales, which explains why a conservative configuration was used for the B matrix in this study (see section 2.5). The challenge is exacerbated when tackling a period such as 2019-2021, with lock-down measures in response to the COVID-19 crisis highly impacting the emissions and thus the structures of uncertainties

in the emission inventories over large spatial scales but limited periods. These sentences have been added in the conclusion.

We advise stepwise improvements in the configuration of B for future studies: attempting to include some temporally varying spatial and temporal correlations in B despite the current lack of knowledge to support a fully relevant characterization of these correlations, and, in parallel, increased efforts to diagnose the uncertainties in the gridded inventories used for inversions.

Technical corrections

• 10: Consider replacing "The inversions lead to a decrease..." by "The inversions suggest a decrease...".

This has been corrected.

• 18: "consistently" should be "consistent".

We have removed this part of the sentence.

20-22: Consider splitting up the first sentence of the introduction, as it is rather long, e.g., "Nitrogen dioxide (NO2) is of great interest due to its important role in many atmospheric processes with strong implications for air quality, health, climate change and ecosystems. NO2 is emitted mainly by road traffic, thermal power plants and industrial activities and produced in the atmosphere by the oxidation of nitric oxide (NO), which is emitted by the same activities".

This has been done.

27: "UE" should be "EU".

This has been corrected.

32: "reached with since" should be "reached by".

This has been done.

167: "Gloabl" should be "Global".

This has been corrected.

• 279: "with emissions higher than 72 kteqNO2 during winter" either add "per month", or "monthly emissions".

We have changed the sentence: "... with monthly emissions higher than 72 kteqNO2 during winter and equal to or lower than 66 kteqNO2 during summer".

• Figure 5: Would it be possible to show the country borders a bit more clearly in these maps?

This has been done.

404: "are about 800 kteqNO2". In lines 280-281 an average of 850 kteqNO2 is mentioned.

This has been corrected.

430: "randome" should be "random".

This has been corrected.