Review "Variability of nitrogen oxide emission fluxes and lifetimes estimated from Sentinel-5P TROPOMI observations", by Lange et al.

The study by Lange et al. applies the EMG method to TROPOMI NO2 observations to estimate NOx emissions and lifetimes in various urban areas and sources. They study in detail the variability, seasonality, trends and geographical dependence of the NOx emissions and lifetimes. The method is robust and has been applied to satellite data in the past, with the added value of detailed analysis and other sources being analyzed in this study. The paper is well written and well structured, with quality figures supporting the methods and discussion provided in the text.

Although results and conclusions are presented in a clear way, there are some points that need to be addressed to make the conclusions more substantial and robust. So I would recommend for publication once the comments below are addressed by the authors.

General comments

In general, the introduction could be improved. Currently the concepts are mixed, and it is not to the point. After reading it, it is not clear what the study is going to be about, and what the research adds to the current literature that is discussed is lacking. It starts with some chemistry, then lifetime, emission estimates, seasonal, day to day, trends, then covid and then back to lifetimes to then introduce the satellite measurements that have already been mentioned at different points in the text before. In the last paragraph you explain what you do in the study but still go back to literature, so not to the point. I think the text and ideas are there, but somewhat disorganized.

Sect. 4.5 and the discussion on the Covid effects on NOx emissions is vague. You argue that EMG method accounts for meteorology and other possible effects on NOx emissions, but the arguments do not hold equally for the three cities that are discussed and different months. Only months where results are supported by the covid hypothesis are brought forward, which makes conclusions less solid. Many cities have been analysed in literature to study covid effects on air pollution, so the fact that only three are presented here is also a sign that the method does not work in other cities. If this is not the case, then this should also be stated in the manuscript.

The results discussed in other sections may be affected by Covid, but there is no mention to that. How are the summer to winter ratios affected by covid? How is the comparison to EDGAR affected by the supposedly lower emissions when restrictions due to covid where active?

Sect. 4.6: The 43-63% uncertainties are higher than the 1-sigma uncertainty provided by the EMG method (e.g. Table 1 uncertainties are ~ 5 %). The high uncertainties need to be reflected in the uncertainty values you provide for the emissions and lifetimes, and discussed in this section.

Specific comments

Introduction

Line 101: Is not only urban areas that are the target of the study, right? Same at the beginning of Sect. 5.

Why you do not discuss Beirle et al. (2021) in the Introduction? Is it because those are only point sources?

Data

Line 154 when referring to Fig. 1: "The red circles mark regions with higher NO2 than their surroundings and are analyzed in this study." Is this the reason why you choose these regions? Now it reads as it would be like that, but there are many more places with "higher NO2 than the surroundings".

Wind data and ozone mixing ratio: one is interpolated to the exact overpass time and the other to "typical mean early afternoon overpass time". Why this difference?

Line 191: were "new sources" found in Georgoulias et al. (2019)? Or was it positive trends as well as negative ones, that also reversed during the last decades? Could you use the trends from 2015-2017 from this study to extrapolate EDGAR inventory?

Sect. 3.1.: why you do not choose any region in Australia? Southern Hemisphere is underrepresented in your selection.

Pleases mention here the > 2m/s applied to the overall filtering criteria.

Have you looked at boundary layer height information? 100 m is probably too low, so you might introduce a systematic error here.

Results and discussion

Sect. 4.1 & Sect. 4.2: How do your summer estimates agree with Goldberg et al. (2019)? Are there any temperature anomalies in the areas you study that could point to an over/under estimation of the emission inventories based on the threshold temperatures assumed for heating/air conditioning?

Sect. 4.3: Lifetimes need to be validated in order to discard unrealistic effects on the NOx emissions and ratios shown in Sect. 4.2. Also, in view of the last paragraph of Sect .4.4. Or at least make clear that the uncertainties associated to the lifetimes is much higher than 1-sigma given by the method, which results in important uncertainties in the emission estimates.

Sect.4.4: line 420: what if you only take into account summer data as in Goldberg et al. 2019?

Sect. 4.5: line 469: this can be known by looking at meteorology anomalies in this period.

Line 475: two years cannot be considered a "trend". Has the economy changed so much that is noticeable in emissions?

Line 483: This would indicate that the lockdown was much stricter in May compared to October, so these estimations might be affected by other factors rather than covid, which could point to the lifetime that is estimated being too low.
What is the explanation for higher emissions in Feb. 2020 compared to 2019?

Technical corrections

Throughout the manuscript, please modify "nitrogen oxide" by "nitrogen oxides" and "emission" by "emissions" when necessary. NOx is plural, so NOx <u>are</u> emitted (e.g. page 1, line 15).

Please define acronyms first time they are mentioned (e.g. abstract lines 1-2 TROPOMI, NOx)

Abstract: "is ECMWF ERA 5" relevant? Just "wind data" could be sufficient, as there is no need to go into details in the abstract.

Line 90: "but analyses with GEOS-Chem…": reader does not necessarily know what GEOS-Chem is, so please specify.

Line 92: do you mean "measurements of NO2" instead of "measurements of NOx"?

Line 116: "(" missing

Line 152: "removes part of the scenes" -> "removes the scenes"

Line 215: facilities - > facilitates