The work presented by R. Pope et al. in the study "Exploiting satellite measurements to reduce uncertainties in UK bottom-up NOx emission estimates" shows new results in the NOx emission estimates based on satellite observations over the UK.

I was interested to see the results since this subject, i.e. to derive emissions with satellite observations and without the use of a chemical transport model, is becoming more and more popular and represents a fast and efficient way to provide these estimates.

While this topic is more and more studied in several countries (e.g. the USA, China, India, France to cite a few), it is also the first time, to my knowledge, such study is done in the UK.

However, I have major concerns in the results published in this work.

1) The use of the NAEI16 should be discussed and explained a bit more by the authors. It is a 3-yr old dataset and the authors have decided to interpolate this emission inventory to represent 2019.

The NAEI19 is available as explained in the text. I also understand it was not feasible to use this new inventory at the beginning of the study. But there is no explanation on the interpolation used.

Since the NAEI19 is available (and other more recent years too), a minimum should be to check and present an evaluation of their estimated 2019 emission in comparison to the official NAEI19 and see if their assumption is correct or not.

I guess it will not change the main conclusion of the paper, but I think the assumptions used by the authors are critical and need more explanations.

2) The authors should also justify their criteria used for the TROPOMI data. They used a quality flag of 0.5 with additional criteria. However, the ESA does not recommend using the data with a quality flag lower than 0.75. This higher value in the quality flag avoids the cloudy scenes and the problematic retrievals.

Loads of studies use this flag at 0.75. To cite a few, there are:

- Bierle et al. 2019 Science: https://www.science.org/doi/10.1126/sciadv.aax9800

- Zhao et al. AMT 2020: https://doi.org/10.5194/amt-13-2131-2020

- Ialongo et al., Atmos. Env. 2021 : https://doi.org/10.1016/j.aeaoa.2021.100114

- Lange et al. ACPD 2021 : https://doi.org/10.5194/acp-2021-273

Moreover, there is the change in the cloud filter in 2019 which impacts the data with a quality flag of 0.5.

Thus, I think the TROPOMI values presented in this study are overestimated.

3) The method used by the authors to estimate the NOx emissions is sensitive to several parameters, such as the BL height (they have chosen the levels between 1000 and 850 hPa) and the coordinates of their point source (city). A sensitivity analysis could be done, or at least a discussion should be added.

4) I do not see a clear conclusion about the results given for the different wind directions. If I take the example of London, the emission rate varies from 36.60 to 58.70 mole/s.

The result based on the S direction for London is not presented (Table 1). Is it because there is not enough data to have a representative estimate? Based on the sentence in line 400, I guess the S wind direction for London does not frequently occurs.

I have the same comment for the other cities, why did the authors provide an estimate only for one or two wind direction(s)? Their method seems very sensitive to the wind direction used.

5) The conclusions are based on a comparison between one emission year (2016 scaled to represent 2019) with ~ 2 years of TROPOMI data (Feb 2018-Jan 2020). Why the authors did not use only the year 2019 for the observations?

This difference should be better stated through the manuscript, i.e., in the abstract, in the figures (in the axis and the caption of Fig.5; in a legend for the maps and on the axis in Fig. 3). This gives a confusing message.

Minor comments:

- All the figures are hard to read and some of them lack explanations. For example in Fig .4 , a better legend explaining the curves should be given.

- In the introduction, lines 68-69, the correct sentence should be something like:

"Until recently, spatial verification of NAEI... (Tsagatakis et al., 2021)."

https://uk-

air.defra.gov.uk/assets/documents/reports/cat09/2107291052_UK_Spatial_Emissions_Methodology_ for_NAEI_2019_v1.pdf

That's true, this report is not an evaluation of the emission maps with satellite observations, but these satellite observations are now included in the analysis.

- The authors should provide the reference of the ECMWF wind data they have used.

- I do not understand the sentence in line 210 "...and τ of 3.8 hours (7.0 and 2.6 hours)". Does it mean the value ranges from 2.6 to 7 with a mean equal to 3.8 hours?

- Line 392: "Our methodology was applied to 12 city sources where sources had suitable downwind TCNO2 enhancements...". What does a "suitable downwind enhancement" mean? What was the criteria to choose these 12 cities?

- The last sentence of the conclusion is too general. The readers will be interested to know what is needed to improve the NOx estimates with the satellite observations, or how can we improve the NAEI with this work?

- Figs 1 and 2. It will be interested to know the number of days and the number of pixels used in the graph. It will give an idea of the representativeness of the results.

- Fig. 5: Does the lifetime represent the mean calculated lifetime with the different estimates? If yes, it should be explained in the caption.

In Table 1, the NO2 lifetime for London can be: 3.8, 8.3 and 5.3 hr. In Fig 5, it seems to be larger than 6 hours.

- Lines 423-425: "As the input emissions for GEOS-Chem come from the NAEI (2016 NAEI emissions scaled to 2019), any inconsistencies between simulated and observed NO2 potentially indicates discrepancies in the underlying emission." That's not fully true. Inconsistencies can also be due to the GEOS-Chem model (deposition, chemistry, dispersion) and/or uncertainties in the TROPOMI observations. The inconsistencies may also come from the way the authors gridded the NAEI or how they vertically distributed the emissions in the model.

- Note the weblink: <u>http://www.temis.nl/airpollution/no2.html</u> does not work (checked on 16 September).