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Comment on amt-2022-176

Anonymous Referee #2

Referee comment on "Estimation of NO₂ emission strengths over Riyadh and Madrid from space from a combination of wind-assigned anomalies and a machine learning technique" by Qiansi Tu et al., Atmos. Meas. Tech. Discuss.,
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The manuscript presents an interesting new method for the determination of spatially resolved emission maps around megacities based on TROPOMI NO₂ observations, wind fields from meteorological models, and machine learning techniques. The study matches the scope of AMT. Before publication, however, major additions/extensions are needed.

The paper presents results of the new approach exemplarily for Riyadh and Madrid and states that the method "works properly and is reliable" (line 283).

However, the resulting emission maps reveal strong artefacts which are not at all mentioned in the paper.

A critical evaluation/discussion of shortcomings, artefacts, problems, and uncertainties of the proposed approach is missing.

Major concerns:

- The presented results reveal several artefacts:

- o several pixels of quite high emissions over regions without obvious NO_x sources, e.g. at 25.05°N, 46.45°E and 25.25°N, 46.45°E (Figs. 1d, 3a).

There is neither a significant NO_x emission over this area reported in Beirle et al., 2019, nor is there a local enhancement in the NO₂ column (Fig. A4).

- o a large extended area of positive emissions north of Madrid (>40.7°N), where CAMS emissions are close to zero.

While values of individual pixels still look relatively low in the color coded image, the integrated emissions >40.7°N are still considerable, and I do not think that these emissions are real.

- o generally "checkerboard-like" structures in the maps for data subsets.

This indicates a problem with the method that involves solving a linear equation iteratively. It seems that initial deviations are overcompensated in the next neighbor, than undercompensated in the 2nd next neighbor, and so on, indicating an instable system with oscillating values in the solution. I think this effect is a known problem for inverse problems, and the authors might check whether they find standard procedures for

avoiding or suppressing these oscillations.

In any case, the authors have to clearly describe the artefacts and discuss possible reasons.

- The authors should be more careful about the usage of the terms "NO₂" and "NO_x". Emissions are sometimes referred to as NO₂ and sometimes as NO_x in the manuscript. Clarify the issue of NO_x = NO + NO₂ in the beginning (emissions should refer to NO_x, while TROPOMI only measures NO₂).

Specify how you account for the missing NO in the NO_x budget.

Note that there are more "oxides of nitrogen" (line 36) such as NO₃, N₂O₅ or N₂O, which are not included in NO_x.

- In the introduction the authors give a quite high range for the lifetime of NO_x of 1-12 hours (should be "tropospheric" rather than "atmospheric" lifetimes in line 43). However, later they just use one fixed value, ignoring probable seasonal and spatial variability of the lifetime. This simplification has to be stated clearly and the impact on the resulting emission maps should at least be investigated with some simple case studies.

- The study makes several simplifications such as constant lifetime, constant wind field, no consideration of seasonal effects (which might correlate with wind direction and thus would directly affect the wind-assigned anomaly). A discussion of the impact of these simplifications, and in general an error discussion is missing.

- Finally, it is not clear to me what exactly would be the benefit of the proposed method. Quantifying megacity emissions is of course a valid goal, but this could be done with simpler methods as well. So the "extra" of the proposed method would be the generation of spatially resolved emission maps. For this purpose, a discussion of uncertainties and "reliability" of emission values for individual pixels is required. In addition, the authors should indicate concrete applications for the derived emission maps.

Further comments:

- Selection of sample locations: application of the method for Riyadh is quite straightforward due to the good observation conditions, as well as the split of wind directions almost equally in two opposite directions. But I wonder how the method should work for Madrid, as there is basically one dominating wind direction. So the wind-assigned anomaly can definitely tell you where Madrid is located, but with this "unimodal" wind pattern, I really wonder what additional information on spatial distribution of sources should be gained. There might be other megacities where the approach might be more promising.

- Line 70: "used to train": training is a crucial element of any ML, and I wondered here

against which "truth" the ML is trained. It needs Eq. 1 to understand how the "modeled truth" is constructed that is used for training. I think that this is also an important component of this approach, that a simple downwind plume model is used to construct the NO₂ distribution for the emissions from each grid pixel.

- Section 2.2: The authors describe eq. 1 as the "averaged distribution ... over a long time-period" (Line 90), but later apply this to "daily plumes". Please clarify.

- Eq. 1: what should be the meaning of the division by an angle in degree? I think this cannot be correct - is there a sin or cos missing? Otherwise, please clarify the units of all components of Eq. 1.

- Line 115: I understand the motivation for choosing $\log(w_k)$ as proxy for w_k . However, this drastically modifies the weight of the different pixels with focus on very low emission values.

The satellite measurements, on the other hand, have highest signal to noise for the pixels with very high emission values.

This has to be discussed. Have you tried to run the algorithm directly with w_k instead? This will of course result in some negative emission values, but the results for strong sources like powerplants might be more reliable.

Please also specify the exact procedure: are Eqs. 5-7 applied for w_k or actually for $\log(w_k)$? If the latter is the case, then there should also be $\log(w_k)$ written in all equations, plus an additional equation indicating how final emissions are derived (perhaps trivial, but I think very helpful for understanding what was done exactly).

- Line 115: Where is the initial epsilon coming from?

- Line 132: If the outer ring should be skipped in order to skip edge effects, the initial study area must be $(n+2) \times (m+2)$, since one pixel on each side has to be skipped.

- Line 152: In addition, for Riyadh the separation into two wind regimes suggests itself.

- Line 279: I would not agree that the spatial patterns are very well. There are some artefacts present in the presented emission maps, and the conclusions should reflect this.

- I'm no native English speaker myself. However, several sentences and formulations sound strange to me. I would recommend careful language check after dealing with the requested modifications/extensions.

Technical issues:

- Line 49: etal. -> et al.

- Line 50: TROPOMI acronym explained twice.

- Line 80: To which area are the 910,000 measurements refer to? I think it would be more useful to give a typical number how many measurements there are per 0.1° grid pixel.

- all emission maps: The emissions are given in molecules per second, but implicitly refer to the chosen grid. I.e., for 0.05° grid, numbers would be only 1/4 of the presented values.

Emission maps should thus be provided as densities (emissions per time per area).

- all maps: please choose the same lat/lon range for all plots for Riyadh and Madrid, respectively.

- Caption Fig. 1: 0.1° is coarser than most TROPOMI pixels, thus the data is not "oversampled".

- x labels of Fig. 1 (c), 2 (c), 3 (f): "TROPOMI tropospheric NO₂" is misleading here, as the shown quantity is a difference (or anomaly).

- Figs. 3 (c) and 4 (c): There are several pixels where weekend emissions are higher than on weekdays. Thus the colorbar in (c) should be symmetric around 0, including negative values as well.