

review of "Estimation of NO₂ emission strengths over Riyadh and Madrid from space from a combination of wind-assigned anomalies and machine learning technique" by Tu et al

Anonymous Referee #1

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 paper "Estimation of NO₂ emission strengths over Riyadh and Madrid from space from a combination of wind-assigned anomalies and machine learning technique" by Tu et al presents a new simple method to obtain the tropospheric NO₂ emission strengths and their spatial patterns derived from the TROPOMI observations. It relies on wind-assigned anomalies and machine learning (ML) technique (the so-called Gradient Descent) and it is applied to 2 cases (Riyadh and Madrid), which have already been used for emission estimates by past literature, to which they compare to in a few words. They also present the week-end effect and the impact of the COVID at the 2 locations.

The paper is well written and easy to follow, the study is interesting and in the scope of the journal, but the method is only briefly described relying a lot on references, and only describing the technical implementation, and no error estimation is mentioned.

I recommend publication after some revision, including some more discussion and some testing cases (some are suggested below) to provide an estimation of the uncertainty of the method.

Specific comments:

The paper to my point of view lacks some discussion about the importance of the choice of different parameters, that here have been fixed "once for all", as coming from a reference (sometimes on a quite different topic), e.g. the choice of the wind (line 85) is the ERA5 at 330m coming from an "empirical choice based on Tu et al (2022b) ...", but where the focus is methane. In the discussion of the Riyadh results (sect 3.1) the output value is compared to Beirle et al. 2019, using a different wind source (ECMWF) and height (450m) and no discussion at all is made to help the reader knowing if these choices have a (large) impact or not. Similarly, when discussing the differences with previous results from Beirle 2011 and 2019 only a sentence "The difference might be due to the different study periods and methods used." (line 162) is mentioned. Also for the Madrid case (line 184: "This discrepancy is partly due to the different 185 periods, methods, and data sets used."). The author could make some tests on either different heights, or different wind source and give an estimation of the final outcome value.

To estimate the impact of the selected choices, I would suggest the following tests:

- use 1 year of S5p data instead of 3. Has this a big impact? Is this helping the coherence with the Madrid results, where yearly inventories are available?
- test another wind height (why not the surface? as the NO₂ is a short-live species, the NO₂ will follow the orography)
- what is the impact of S5p in term of pixel resolution (ie wrt to OMI larger pixels used in Beirle 2011 for Madrid)? Could a test be made by resampling S5p to OMI resolution and seeing the impact on the outcome emission estimate?
- be careful that the S5p NO₂ dataset is an aggregate of different versions (010202, 0103xx since 20/3/2019 and 0104xx since 29/11/2020, see ROCVR here: <https://mpc-vdaf.tropomi.eu/index.php/nitrogen-dioxide>). This should be mentioned in Sect 2.1 (v1.4 has an important change in the FRESCO cloud algorithm leading to larger NO₂ columns, see e.g. Van Geffen et al., 2022) and a test on the impact of the change of version could also be interesting (as the remote and urban NO₂ columns changes differently, is this leading to a different spatial result of the emissions?). The different versions should be added in figure A9 with the S5p NO₂ time-series.

Other minor comments:

About the COVID impact, the illustrations are interesting, but the context of other studies could be done better. Some studies explicitly mention Madrid, in maps or tables (eg Beuwens et al 2020, table 1 and Levelt et al., 2021, figure 3) and could be discussed.

Some of the figures in the annexe are "quick and dirty" (or give this feeling at least). They seem like simple print-screens, without any latitude & longitude coordinates, etc (figures A5, A7).

Technical comments and corrections:

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- line 33: "Though our analysis is limited to two cities as testing examples, the method has proved to provide reliable and consistent results." --> what are the errors and limitations of the method? this is not presented in the manuscript!
 - line 34 (and in the conclusions): "Therefore, it is expected to be suitable for other trace gases and other target regions." --> be a bit less optimistic, here only the 2 "easier" cases have been presented, but there can be challenges for other places/gases.
 - Sect 2.1: give more details on the TROPOMI NO₂ data used. Which version? OFFL or a reprocessed? which version number? (at least mention that different versions exist and give references)
 - lines 85-88: see comment above about wind selection. Methane has a much longer lifetime than NO₂ (about 10 years vs a few hours), so explain why the choices made for methane are still ok for NO₂ or give some estimation of the uncertainty related to this choice.
 - line 96: how much impact has the choices of tau (4h for Riyadh and 7 for Madrid) on the result? what are the ideas to estimate this value for other trace gases or other target regions? (to follow my comment on the 2nd bullet here above!)
 - lines 130-133: I don't understand very well the scope of increasing the area, and then removing the "outmost ring" (ring is a bit misleading as the ROI is a rectangle)

- lines 141-145: although this paragraph is given to explain things, although for a non-expert on ML as myself, it is just creating confusion, with a lot of different names (without explanations or references), to end up with "This practice is not necessary with our problem." --> why?
- line 157: suggestion to "The estimated emission strengths based on the ML model *(Fig. 1d)* show a very similar spatial pattern to the results in Beirle et al. (2019) (Fig. 2)."
- line 163: see comment above about discussing impact of different choices and tests that could be made to estimate uncertainty of the present method.
- line 184: "with a spatial resolution of $0.05^\circ \times 0.1^\circ - 0.1^\circ \times 0.1^\circ$ in longitude and latitude" --> I don't understand this resolution notation.
"on a yearly basis" --> what year is considered in CAMS-REG-AP? or, if different years are available, can a test be made with only one year of TROPOMI to see if the year-to-year variability is similar?
- line 183: comment on the possible OMI vs TROPOMI resolution impact (see proposed test above). An error estimation would really help disentangle choices made with this approach and impact of time-periods (2005-2009 vs 2018-2021). How are the trends in NO₂ around Madrid ? (see eg Georgoulias et al 2019 Fig 5)
- line 190: what do you mean by "With an expectation, these actions may help to decrease NO₂ concentrations by ~25% in the central area by 2020." ? We are in 2022!
- line 197: mention that the airport is presented on Fig2d with a triangle.
- line 203: what does mean the 1/2 subscript on the different directions? it is also present in Fig A3, but not in its caption.
- line 227: suggestion "The ML-estimated emission strengths *for Madrid* are presented in Figure 4."
- line 228: suggestion "However, for weekends, the northeastern regions *(close to the airport)*, far away from the city center, are the main sources,..."
- line 259: "The NO₂ emission estimate in the urban area *of Madrid* is about..."
- lines 260-267: add more discussion of other lockdown studies (see above some suggestions). (Do the same for Riyadh)
- line 282: same as said before "This difference might be due to the different study period and methods"--> this is too general. some investigations should be performed to give at least some error estimate/ quantification of impact of some of the choices made here.
- line 305: "But, it is can be applied to other key gases such as carbon dioxide or methane, and in other regions" --> as mentioned for the introduction, for me this is too general/optimistic. In my view carbon dioxide or methane have very different life-times and the urban emissions should be disentangled from the background. If you feel differently, please provide some supporting comments to your sentence.

References:

Levelt, P. F., Stein Zweers, D. C., Aben, I., Bauwens, M., Borsdorff, T., De Smedt, I., Eskes, H. J., Lerot, C., Loyola, D. G., Romahn, F., Stavrou, T., Theys, N., Van Roozendaal, M., Veefkind, J. P., and Verhoelst, T.: Air quality impacts of COVID-19 lockdown measures detected from space using high spatial resolution observations of multiple trace gases from Sentinel-5P/TROPOMI, *Atmos. Chem. Phys. Discuss.* [preprint], <https://doi.org/10.5194/acp-2021-534>, in review, 2021.

Bauwens, M., Compennolle, S., Stavrou, T., Müller, J.-F., van Gent, J., Eskes, H., et al. (2020). Impact of coronavirus outbreak on NO₂ pollution assessed using TROPOMI and OMI observations. *Geophysical Research Letters*, 47, e2020GL087978.

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