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Comment on acp-2021-268

Anonymous Referee #1

Referee comment on "A satellite-data-driven framework to rapidly quantify air-basin-scale NO_x emissions and its application to the Po Valley during the COVID-19 pandemic" by Kang Sun et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-268-RC2>, 2021

Review of 'A Satellite Data-Driven Framework to Rapidly Quantify Air Basin-Scale NO_x emissions and its application to the Po Valley during the COVID-19 Pandemic' by Sun et al.

The paper by Sun and co-authors presents a study into changes in NO_x emissions inferred from OMI and TROPOMI NO₂ retrievals between 2005 and 2020 over the Po Valley. The theoretical basis for the inversion approach relies on a textbook-type one box model that is ventilated by wind; the stronger the wind, the more loss of pollution to outflow rather than to chemical loss, and stronger loss overall. The authors exploit this relationship between NO₂ levels and wind speed and show it to be captured by OMI and TROPOMI NO₂ observations and ECMWF wind data.

The method to derive NO_x emissions relies on a Bayesian framework with a priori climatologically established relationships between OMI NO₂ columns and wind speeds, and furthermore relies heavily on fine-tuning using pre-existing emission estimates. The authors explain this quite well in the manuscript, but they are optimistic in calling their approach 'fast' or capable of providing 'timely' updates to emissions given the sophisticated efforts they require to arrive at the final results. These are nevertheless interesting, and I recommend publication in ACP after the following concerns have been addressed.

Major comments

- P6, Eq. (4): the assumption that wind efficiently ventilates pollution from the Po Valley is rather questionable. Mountains are surrounding much of the valley, so that with winds from the west, the east and the south, air pollution can be expected to accumulate and circulate within the basin, rather than to be transported away from it.

The authors should provide a convincing justification for why Eq. (4) would still hold over the Po Valley. To me it seems only true if the winds are coming from the north and blow out pollution to the Mediterranean Sea.

- The claim that the method can also be applied to other regions would need to be put to the test. Given the concern above, it might have been better to test the technique on a polluted area not surrounded by mountain regions, such as the Ruhr Area or the British Midlands.
- NO_x lifetime changes with changing wind speed (Valin et al., 2012; Lorente et al. [2019]), but the method proposed by the authors fits only one lifetime for different wind speed levels. This seems to be an internal inconsistency in the method (Figure 3 and Eq. (5)) and the authors need to quantitatively explain how they circumvent this problem.
- I'm missing a discussion of the role of soil NO_x emissions in the Po Valley. These are likely not fully represented in the bottom-up inventories, but since satellite NO_2 measurements observe contributions from all sources, including the sizeable soil NO_x source in the agricultural hotspot of the Po Valley (e.g. Visser et al. [2019]), this may well lead to discrepancies between the top-down and bottom-up NO_x emission estimates.
- The other weak point is the assumption that the $\text{NO}_x:\text{NO}_2$ ratio in the polluted boundary layer can be taken as fixed and always have a value of 1.32. The authors should verify the validity of this assumption over the Po Valley, i.e. if there are not trends in the $\text{NO}_x:\text{NO}_2$ ratio following from the reductions in NO_x emissions.

Specific comments

L59: it is better to refer to the direct source of information (e.g. Schenkeveld et al. [2017]) on the row anomaly instead of a paper referring to that source.

L63: 'largely' ... besides difficulties with a priori profile shapes, there may also be issues with surface albedo and cloud parameters in the TROPOMI NO_2 AMF calculation.

L112: it is unclear how the uncertainty in the satellite NO_2 values has been used. Moreover, it is not recommended to weigh columns according to their uncertainties because the most polluted scenes would then have less influence on the averages. See e.g. discussions on how to calculate representative averages in Miyazaki-papers and Boersma et al. [2016].

Figure 4 and 6: can the authors qualitatively explain why April/May has such high NO_x emissions and such a low NO_x lifetime?

L275: indications for shorter lifetimes have also been found over The Netherlands by Zara et al. [2021].

L305-306: can the authors explain what it is in the instrument performance that causes the reduction in DOFS? Increased noise leading to larger SCD uncertainties? Or would it rather have to do with data availability?

L309: please quantify what "match closely" means here.

L332: suggest to be careful with words such as "timely". This remains to be seen, the current method may give faster results than bottom-up estimates, but given the heavy level of tuning and need for climatologies to be developed, it cannot be called "fast".

References

Boersma, K. F., Vinken, G. C. M., and Eskes, H. J.: Representativeness errors in comparing chemistry transport and chemistry climate models with satellite UV-Vis tropospheric column retrievals, *Geosci. Model Dev.*, 9, 875-898, doi:10.5194/gmd-9-875-2016, 2016.

Miyazaki, K., Eskes, H. J., and Sudo, K.: Global NO_x emission estimates derived from an assimilation of OMI tropospheric NO₂ columns, *Atmos. Chem. Phys.*, 12, 2263-2288, <https://doi.org/10.5194/acp-12-2263-2012>, 2012.

Schenkeveld, V. M. E., Jaross, G., Marchenko, S., Haffner, D., Kleipool, Q. L., Rozemeijer, N. C., Veefkind, J. P., and Levelt, P. F.: In-flight performance of the Ozone Monitoring Instrument, *Atmos. Meas. Tech.*, 10, 1957-1986, <https://doi.org/10.5194/amt-10-1957-2017>, 2017.

Visser, A. J., Boersma, K. F., Ganzeveld, L. N., and Krol, M. C.: European NO_x emissions in WRF-Chem derived from OMI: impacts on summertime surface ozone, *Atmos. Chem. Phys.*, 19, 11821-11841, <https://doi.org/10.5194/acp-19-11821-2019>, 2019.

Zara, M., Boersma, K. F., Eskes, H., Denier van der Gon, H., Vilà-Guerau de Arellano, J., Krol, M., van der Swaluw, E., Schuch, W., and Velders, G. J. M.: Reductions in nitrogen oxides over the Netherlands between 2005 and 2018 observed from space and on the ground: decreasing emissions and increasing O₃ indicate changing NO_x chemistry, *Atm. Environ. X*, 9, 100104, <https://doi.org/10.1016/j.aeaoa.2021.100104>, 2021.