

In this article Goldberg et al. present an analysis of how the tropospheric vertical column density (VCD) and surface concentrations of nitrogen dioxide (NO_2) depend on cloud coverage. The analysis is carried out over the contiguous U.S. based on measurements from the satellite instruments TROPOMI and TEMPO, in situ measurements at the surface, and simulation data from the chemistry and transport (CT) model WRF-Chem. The influence of cloud cover on these different type of measurements and simulations is often given little attention, at least in the satellite community, where rejection of cloud-contaminated data is mostly the norm. The article deals with an important topic, fits well into the scope of ACP, and presents interesting results. I recommend publication after the following points have been addressed.

Major points

I only have two major points of criticism regarding the content of the paper.

1. NO_y biases of in situ measurements with molybdenum cartridges

91 % of the instruments in the EPA AQS dataset use molybdenum cartridges, and previous studies have found that their cross-sensitivity to NO_y can be very large. Authors like Poraicu et al. (2022) and Kuhn et al. (2024) have reported overestimations in the approximate range of +20 % to +100 % based on model simulations. Examples of empirical studies addressing said issue include Lamsal et al. (2008) and Villena et al. (2012). The authors should consider

- Referencing some of this literature to give the reader an impression of how „far off“ these measurements potentially are.
- Discussing the influence of these cross-sensitivities on the presented results (e.g. in sect. 3.2). The relevant NO_y species (PAN, HNO_3 , etc.) are photo-oxidants, i.e. their concentration (and thus, the „falsely measured“ NO_2) decreases under cloud cover. In other words, cloudy scenes are not only expected to have higher NO_2 concentrations, but also less measurement bias due to NO_y . The authors must quantify this effect when comparing in situ measurements with/without clouds. This could, for example, be attempted through the WRF-Chem simulation data, which lets the authors estimate the contribution of „false“ NO_2 using the correction term given by Lamsal et al. (2008).
- In this context, the entry „V2.4 no chemiluminescence“ in Table 1 should also be discussed more directly.

2. Air mass factors in the satellite retrievals

A good explanation of the air mass factor (AMF) can be found in the TROPOMI PUM (see Eskes et al., 2022). Section 2.2 mentions the AMF, but does not go into the details, which are essential for the retrieved NO_2 VCD under cloudy conditions. In particular:

- I. 138: approximately 15 % of these -34.8 % low bias are related to the NO_2 profile shapes used to compute the AMF, see e.g. Tack et al. (2021), Judd et al. (2020), Griffin et al. (2019). TROPOMI uses NO_2 profiles from the TM5 model which has a horizontal resolution of $1^\circ \times 1^\circ$ (i.e. much lower than the actual measurement resolution).
- Section 3.3: The AMF essentially „fills up“ missing sensitivity with information from the TM5 model. In other words, if the reported NO_2 VCD changes in the presence of clouds (as shown in Fig. 5), this is does not necessarily reflect a change of actual NO_2 columns as a physical consequence of the clouds - It might just as well be caused by differences between the TM5 model (which then impacts the retrieved NO_2 VCD more) and the real world. This aspect should be explained more clearly, and the conclusions in sect. 4 should be adjusted.
- Section 3.5: This comparison is only justified if both TEMPO and TROPOMI use the same AMFs recipe (i.e. the same NO_2 a priori profiles, etc.). Is this the case?

Minor points

- I. 17: Specify that you use WRF-Chem and ERA5 model data.
- I. 26-27: This sounds as if the authors provided some form of bias-correction method or formula, which is not the case.
- I. 49-50: There are numerous more articles on the topic, see e.g. the many references given in Sun et al. (2024). Although there is no necessity to list dozens of reference, the authors might consider referencing:
- Cao et al. (2023) and Ghahremanloo et al. (2021), who focus specifically on the U.S.
 - Kuhn et al. (2024), who address the prediction of surface NO₂ with explicit consideration of the molybdenum chemiluminescence biases (see major point above)
- I. 53: better: „up to 5.5 km x 3.5 km“
- I. 77: better: „irradiation“ instead of „strength of sunlight“
- I. 72-87: The photolysis of NO₂ into NO + O should be mentioned somewhere in this paragraph, as this is the main (non-terminal) NO₂ sink associated with the high photolytic reactivity of NO₂.
- I. 92: specify „models“
- I. 93: this study does not focus only on the surface „bias“, but also column densities
- I. 95-98: I think the two points given here do not summarize the findings of this article well, because they do not (explicitly) mention the findings associated with TROPOMI and WRF-Chem.
- I. 124: column densities represent the vertically integrated concentration (not molecules per unit area).
- I. 336-341: another option for the verification of reasonable jNO₂ values would be to compare simulated NO₂/NO ratios to the corresponding in situ observations.

References:

- Lamsal et al. (2008). Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument. *Journal of Geophysical Research: Atmospheres*, 113(D16).
- Poraicu et al. (2023). Cross-evaluating WRF-Chem v4.1.2, TROPOMI, APEX, and in situ NO₂ measurements over Antwerp, Belgium. *Geoscientific Model Development*, 16(2):479–508.
- Kuhn et al. (2024). NitroNet – a machine learning model for the prediction of tropospheric NO₂ profiles from TROPOMI observations. *Atmospheric Measurement Techniques*, 17(21):6485–6516.
- Villena et al. (2012). Interferences of commercial NO₂ instruments in the urban atmosphere and in a smog chamber. *Atmospheric Measurement Techniques*, 5(1):149–159.
- Tack et al. (2021). Assessment of the TROPOMI tropospheric NO₂ product based on airborne APEX observations. *Atmospheric Measurement Techniques*, 14(1):615–646.

Judd et al. (2020). Evaluating Sentinel-5P TROPOMI tropospheric NO₂ column densities with airborne and Pandora spectrometers near New York City and Long Island Sound. *Atmospheric Measurement Techniques*, 13(11):6113–6140.

Griffin et al. (2019). High Resolution Mapping of Nitrogen Dioxide With TROPOMI: First Results and Validation Over the Canadian Oil Sands. *Geophysical Research Letters*, 46(2):1049–1060.

Eskes et al. (2022). Sentinel-5 pre-cursor/TROPOMI Level 2 Product User Manual Nitrogendioxide. Royal Netherlands Meteorological Institute. <https://sentinel.esa.int/documents/247904/2474726/Sentinel-5P-Level-2-Product-User-Manual-Nitrogen-Dioxide.pdf>

Cao (2023). National ground-level NO₂ predictions via satellite imagery driven convolutional neural networks. *Frontiers in Environmental Science*, 11.

Ghahremanloo et al. (2021). Deep Learning Estimation of Daily Ground-Level NO₂ Concentrations From Remote Sensing Data. *Journal of Geophysical Research: Atmospheres*, 126(21).