

Replies to comments provided by Referee #2

Ref.: essd-2024-410

Title: **Global gridded NO_x emissions using TROPOMI observations**

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Earth System Science Data

Type: Research article

We would like to thank the reviewers for their careful reading, that led to interesting comments. Reviews have been addressed in the revised manuscript and commented in this document. For a better readability, reviewer comments are highlighted in grey in this document, while answers are highlighted in light yellow.

Rey-Pommier et al. present a global dataset of monthly mean gridded NO_x emissions derived from TROPOMI measurements.

The authors apply a flux-divergence method which has been presented in previous studies, but, to my knowledge, has not yet been used to compile such an emission dataset on global scale. Thus, the dataset is generally of interest and matches the scope of ESSD.

However, in the current version, I see two major shortcomings that need to be resolved before publication on ESSD:

1. The dataset contains extreme outliers, which I can only explain with some bug(s) in the processing chain, and
2. The discussion of uncertainties misses some of the most important factors (choice of wind altitude, background correction, air mass factors).

For further details and additional comments see below.

Methodology

Horizontal transport

The authors do not directly state which altitude is selected for horizontal wind fields. From the context ("assumption of a stationary state and a pollution concentration close to the ground") I conclude that surface winds have been considered, which is probably not the best choice:

- power plants etc. emit from stacks of altitudes up to some hundred meters.
- even emissions at surface (e.g. from car exhausts) are usually rapidly mixed within the boundary layer. Thus, wind fields for a typical altitude within the boundary layer would be more appropriate, as shown in several previous studies. In any case, the authors should
 - explicitly state which altitude was chosen for wind fields,
 - justify that choice and
 - quantify the uncertainty associated to that choice (by comparison to alternative processing with different altitude).

→ This was not precised in the first version of the manuscript – The first two pressure levels (975 hPa and 1000 hPa) are used for the wind field **w**, hence the calculation of a mean horizontal

wind within a layer of about 350 m above the ground. For ground wind w_g in the topography-correcting term, only the first pressure level (1000 hPa) is used. The text has been changed in the revised version of the article manuscript to precise this.

Noise

The presented emission data is very noisy at high latitudes, in particular over ocean. This is shortly mentioned in the manuscript, and the authors explain it with the low amount of available data. However, there seem to be some issues in the processing that cause quite extreme outliers: For instance, in the January data at (2348, 582), corresponding to 143.6°W, 56.8°N (northern Pacific), emissions are 20 Pmolec/cm²/h, which is a factor 10 higher than the threshold for the "high emission densities" classification in the paper. There are many more such pixel, and also many examples for negative values of the same order of magnitude. Monthly mean maps of NO₂ column densities from TROPOMI are usually quite smooth, and noise of individual pixels is about 1Pmolec/cm². If divided by lifetime, the sink term over ocean should thus be well below 1Pmolec/cm²/h. Topography is not existing over the ocean. Thus only the derivative terms could cause these high numbers. But with low column, and thus low fluxes, how can the derivative be that large?

One even more extreme case occurs at (225, 419) with emissions of -173 Pmolec/cm²/h. This corresponds to 153.8°W, 75.9°S, and might be over Antarctica, i.e. the topographic term might have issues as well.

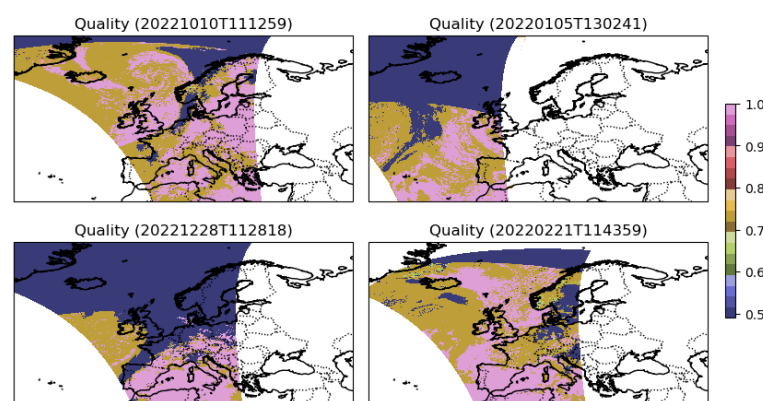
I suspect that the derivatives and/or gridding algorithms, in combination with gaps, causes these extreme outliers. The authors should check these examples and investigate the values in the "orbit" reference frame:

- if the values for individual TROPOMI pixels show such high values, check from which term they come from

- if the orbit data looks reasonable, check the gridding routine.

These extreme and unexplainable outliers devalue the whole dataset. They should be identified and fixed before publication of this product on ESSD.

→ All the analyses carried out in the article are based on pixels between latitudes 65°S and 65°N, i.e. the domain shown in Figure 2. We forgot to specify this in Section 3. This choice is motivated by the systematic low-quality flag applied by TROPOMI above a certain latitude that depends on the satellite orbit and the season, as shown on the different examples below ($q_a < 0.5$ under a given along-track line).



With such low number of observations, mean inferred emissions rates at these latitude are very noisy. Knowing the absence of any major emitter/city above these latitudes (the only cities with more than 100,000 inhabitants above these latitudes are Murmansk and Norilsk, Russia), we expect this restriction not to influence our results.

The outlier identified by this comment is another issue. We thank the reviewer for identifying this outlier, which was not detected, and lead to the detection of an error in our routine. This error stems from an approximation in the estimation of distances between pixels, which is no longer valid in the polar regions (the code was initially developed for estimating emissions in the Middle East). This error underestimated the distance between two pixels that is used to calculate the derivatives, artificially inflating the transport term. For moderate latitudes, the inflation is negligible. However, it grows rapidly with latitude (~6% around 50° but ~94% at 75°). This error does not question the results and the trends obtained here, due to the low underestimation between 65°S and 65°N (where we carried out the analyses), but it remains problematic. We have therefore corrected the distance routine and recalculated all the emissions. All results and product data have been adjusted accordingly. The effects of this modification are the following:

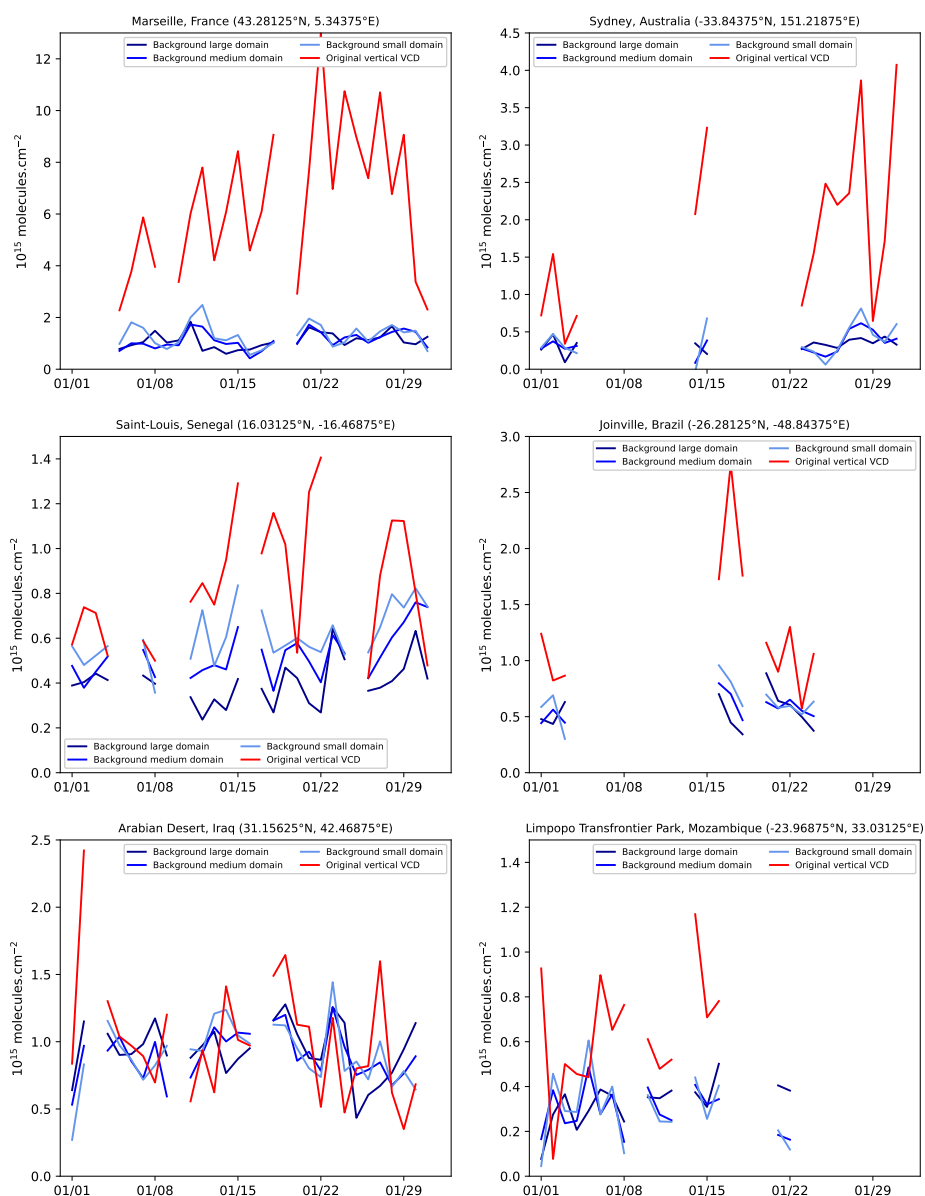
- The typical noise at high latitudes is reduced (visible on Figure 2).
- Some extreme outliers disappear. The detected outlier at (225, 419) is clearly affected by the correction: its January emissions are lowered from -172.88 to -0.02 Pmolecules.cm⁻².h⁻¹ (annual emissions: -0.07 Pmolecules.cm⁻².h⁻¹). The other outlier at (2348, 582) is also affected but to a lower extent: its January emissions are lowered from 19.86 to 15.19 Pmolecules.cm⁻².h⁻¹ (annual emissions: 1.19 Pmolecules.cm⁻².h⁻¹). This second outlier is obtained because this point is observed only one time in January (out of 431 orbits). During this day (23/01), abnormally high VCD values are observed (~4 Pmolecules.cm⁻²) with fast winds, leading to the calculated value.
- Depending on the sign of the transport term, many pixels have emissions that are now above or under the thresholds of 2 Pmolecules.cm⁻².h⁻¹ used to detect point and diffuse sources. As a consequence, the list of detected sources is modified. The number of point sources considered as outliers is reduced from 61 to 48. The average absolute emission change is 4.8% for diffuse and point sources, with a median at 2.7%.
- The effect above is amplified when estimating country emissions, for which a threshold of 0.2 Pmolecules.cm⁻².h⁻¹ is used. Depending on the sign of the transport term, many pixels have emissions that are now above or under this threshold. The total output of countries with very low emissions (generally in sub-Saharan Africa) is therefore changed not by the amplitude of the correction, but by the number of pixels that fall above or below the threshold. However, for high-latitude countries, the correction allows a better estimate of emissions (for instance, Russia has now emissions closer to the EDGAR estimate).

We thank the reviewer for noticing these outliers that allowed to correct and improve our NO_x estimates.

Background correction

The choice for background correction is quite extreme: a full swath width of TROPOMI can cover a wide range of conditions and rather presents a "mesoscale" than a "local" background. I would expect that changing the settings for the background correction can have strong impact on the presented results. This effect has to be investigated and quantified.

→ We chose a full swath width to ensure the background is calculated on a high number of pixels, which would not be the case in cloudy regions. The comment made here remains valid: using such a large region covers several climates. However, as the across-track direction is closer to the east-west axis than the north-south axis, the variations are less marked there than in the along-track direction, where climatic variations can be strong. To quantify the optimal trade-off between those two effects, below is displayed the background calculated at six different pixels in January 2022 when using: 1/ the total satellite swath ($\sim 700 \text{ km} \times 2400 \text{ km}$); 2/ a third of it ($\sim 700 \text{ km} \times 800 \text{ km}$); 3/ a tenth of it ($\sim 250 \text{ km} \times 700 \text{ km}$), centered around the pixel of interest. The pixels have been chosen in order to have examples of rural/empty areas (in Iraq and Mozambique), small cities (Saint-Louis, Senegal and Joinville, Brazil) and the core of large cities (Marseille, France and Sydney, Australia) in the two hemispheres.



Small differences appear between the three estimates. They generally remain below $\sim 0.4 \text{ Pmolecules.cm}^{-2}$, and the use of one particular background has therefore a negligible impact on emissions estimated for powerful emitters (first row), for which VCDs are higher than the background by an order of magnitude. The effect is also low for rural/empty areas (third row), where the three backgrounds roughly equal the value of the point of interest, leading to a zero or near-zero VCD in all three cases. However, the choice of the background has an impact for medium-size emitters (second row): for such emitters, the background calculated on the $\sim 250 \text{ km}$

× 700 km zone can be similar to the VCDs around the emitter. This happens when a lot of low-quality observations are located within the background calculation domain, but not near the point of interest. In this case, some pixels with intermediate pollution levels can be below the 30th percentile or the remaining observed pixels, therefore increasing the background to levels close to the medium-size emitter. This effect is less pronounced when the total swath is used. We thus prefer to use the total satellite swath for the background calculation, despite the associated drawbacks highlighted by this comment. In the revised version of the manuscript, we briefly precise this reasoning.

What is the reason for including a far larger distance across-track than along track?

→ When selecting pixels in the along-track (roughly north-south) direction, one must consider the different origins of background NO_x due to different climatic and geographical conditions. For instance, in a region of interest like Cyprus, extending too far in the north-south direction could result in mixing pixels from arid areas (e.g. Egypt) with more temperate or humid regions (e.g. Ukraine). These areas have distinct NO_x backgrounds originating from different sources, such as soil emissions and human activities, which could skew the analysis if mixed. Conversely, variations in NO_x levels are generally less pronounced in the across-track (roughly east-west) direction, making it more suitable to extend the pixel selection in this direction. However, selecting a large extent across the satellite track also carries the risk of including measurements taken at different local times, which could be problematic as NO_x levels vary with OH which has a strong diurnal cycle. In this specific case, the risk of temporal inconsistency is minimal because the satellite swath has a relatively small longitudinal span, and the overpass occurs around midday when diurnal variations are less pronounced. Therefore, extending the selection in the across-track direction provides a better representation of the background NO_x levels. In the revised version of the manuscript, we briefly precise this reasoning.

The choice of the 1st tercile for defining the background implies that 1/3 of all considered TROPOMI pixels have corrected columns < 0. This should be mentioned, and this is also one of the reasons for negative emissions.

→ There might be a misunderstanding here. When a TROPOMI pixel has a vertical column lower than the calculated background, the corresponding corrected column is reduced to zero: the field of corrected columns is never negative. To make sure this aspect is understood correctly, we detail this in the revised version of the manuscript.

Negative emissions

The dataset includes negative values for numerous pixels. This is shortly discussed in the manuscript, but this discussion should be extended. In particular, the authors should provide a recommendation to the user how to treat negative emissions in potential applications. In my point of view, it makes sense to keep the negative results in the data product, even if unphysical, as the alternative (skipping or setting them to 0) would bias high the overall mean. But this is exactly what happened in the spatial integration of country/regional emissions, which only considers pixels above a (positive) threshold. This might be one reason for the results being systematically higher than those from previous studies (even though no air mass factor correction was applied). The authors should discuss this aspect, and should provide information about how the derived

emissions depend on the selection of pixels, and how they would look like if also the negative emissions would be considered in the spatial integral.

→ This point is particularly relevant for countries with no powerful sources or with low observation densities, leading to a majority of noisy pixels. For other countries however (industrialized countries with typical or high observation densities), the impact is relatively low. We discuss this aspect in the revised version of the manuscript and provide comparisons of country emissions when the positive threshold of $0.2 \text{ Pmolecules.cm}^{-2}.\text{h}^{-1}$ is used (excluding noise but also high negative values), and when no threshold is applied. In the revised version of the manuscript, a complete paragraph is dedicated to this comparison after Figure 8 and Table 3.

Air mass factors

The authors just took the operational product without any correction of the air mass factor (AMF). This is problematic, as the AMF, and also its stated uncertainty, refer to the full tropospheric column, while the authors performed a background correction with the aim to only consider near-surface pollution. For this, the operational AMFs are not appropriate, and are systematically too high (and thus Omega' is biased low). This systematic effect cannot be described by a 30% uncertainty (in both directions).

→ Verhoelst et al., 2021 indicate that the effect of the AMF leads to tropospheric NO_2 columns that are systematically biased low by about 30%–50%, but only over cities. This percentage is lower elsewhere. We used a 30% uncertainty in our estimates of emissions for entire countries, where emissions do not take place only in cities. However, as the reviewer describes here, a higher uncertainty must be used for estimates of cities only (and, to a lesser extent, large industrial facilities that have similar albedo values). We now make this distinction in the revised version of the manuscript.

Uncertainties

The section on uncertainties needs to be extended with discussions of the aspects listed above. The presented agreement to EDGAR is quite good, but due to the issues discussed above, there might be some of the systematic effects compensate each other.

→ In the revised version of the manuscript, we discuss the possibility of offsetting biases that can cancel out following a comment from *Anonymous Reviewer #1*. Goldberg et al. (2019) have shown that emissions at 13:30 LT are higher than the daily average. In these cities, emissions are mainly transport emissions. However, in our study, we also estimate emissions from industrial facilities (power plants, cement kilns, etc). Such facilities account for a large part of the global NO_x budget. In addition, they are generally located outside cities, where the TROPOMI bias is weaker. Finally, their emissions at 13:30 LT are not necessarily higher than average emissions (this depends very much on the use of the industry in question: some power plants are used for baseload, while others are used primarily to meet peak demand). In conclusion, the effect of the two offsetting biases mentioned by the reviewer is indeed present for cities, but is probably less significant outside cities, in a way that depends very much on the location under consideration. This discussion is detailed in Section 3.3 of the revised version of the manuscript.

Purpose

In the abstract, the authors claim that "this dataset is designed to be updated with a low latency to help policymakers monitor emissions". I think that this is an important aspect, as monitoring of changing emissions in timeseries might even work with the remaining high uncertainties. However, this aspect is not discussed any further in the manuscript. It would help a lot to support this argument if the authors could find e.g. a power plant that has been switched off in 2022 and show the corresponding time series of monthly mean emissions.

→ This review is interesting as it suggests to use a precise example to support the scope of our article. It is highly difficult to find information about a power plant that:

- has high emissions to illustrate the method properly,
- has a high density of observations,
- is isolated from other industrial activities and cities,
- whose activity was drastically altered in 2022 (in most cases, it is only a few turbines that are turned off for maintenance), and
- whose activity was precisely reported in global databases or covered in the press.

In the revised version of the manuscript, we illustrate this with the example of the Zaporizhia thermal power plant in Ukraine (not to be confused with the neighbouring nuclear power station), whose activity was altered following the ongoing conflict in the country. This power plant ticks most of the boxes above, but the interpretation of the monthly time series remains uncertain due to low observation densities in January and a lack of knowledge of the power plant activity in the fall. For this reason, we prefer to detail this example in the Supplementary Materials and briefly mention it in the revised version of the manuscript.

Dataset

- Unit

I understand the origin of the unit used ($\text{Pmolecules.cm}^{-2}.\text{h}^{-1}$) as this is just the commonly used unit for NO_2 column densities ($\text{Pmolecules.cm}^{-2}$) divided by a lifetime. However, the presented dataset provides global emissions, which is of high interest for communities beyond those familiar with satellite NO_2 products. Thus, the unit should be modified to an (SI) unit commonly used, like $\text{kg/m}^2/\text{s}$. At least, a conversion factor needs to be provided in manuscript and data product metadata.

→ To our knowledge, there is no global norm on which NO_x must be expressed. Some studies count NO_x in μg , kg or t (tons), but as if all NO_x molecules were NO_2 , some others use NO , and some only count nitrogen atoms. The confusion of such units is the source of many mistakes in articles when this is not clearly specified. Here, we wanted to avoid confusion for future users by using the *Pmolecules* unit, and only convert to tons per hour when presenting emissions of power plants, cities and countries in Sections 3.2 to 3.4. It is also a quite convenient unit since it allows to express pixel emissions usually between 0.5 and 10 $\text{Pmolecules.cm}^{-2}.\text{h}^{-1}$, which are easier ranges to work with. However, we acknowledge that the interest of this article goes beyond the scope of Earth Observation. In the revised data product, we specify the unit and provide six conversion factors (in $\text{kg.km}^{-2}.\text{h}^{-1}$ and $\mu\text{g.m}^{-2}.\text{s}^{-1}$, counted as NO_2 , NO and N).

- NC files

The coordinates should directly be lat and lon. There is no need for a "lat_grid" index. The unit "degrees_north" for the grid *index* makes no sense. When having lat and lon as coordinates, there is no need for additional "latitude_data". I propose to add the grid pixel area (1d, latitude dimension only) such that the user can simply convert the emission densities to total emissions for each grid pixel, which simplifies spatial integration.

→ We modified the data product accordingly, following the advice in this comment: each monthly file of the dataset has now three variables: longitude (1D, 5760 elements), latitude (1D, 2880 elements), and NOx_emissions (2D, 2880×5760 elements).

- Annual mean

As the figures in the paper often display annual means, also an annual mean data file should be provided next to the monthly means.

→ A mean data file has been added in the new version of the data product to account for this comment (see comment above).

- Negative values

The negative values should not be skipped in the dataset. But I would propose to add a disclaimer to the metadata of each file explaining the reason for the occurrence of negative data.

→ Negative values are included in the dataset. A "readme" file is added to the new version of the data product, explaining briefly how the emissions are estimated, the reason for negative data, and a reference to the article for more information.

Minor comments

Line 64: A reference to TROPOMI (e.g. Veefkind) should be added.

→ This is done in the revised version of the manuscript.

Line 68: This high spatial resolution only holds for nadir pixels.

→ We changed "3.5×5.5 km² since 6 August 2019" to "up to 3.5×5.5 km² since 6 August 2019" in the revised version of the manuscript.

Line 92: Provide information about the spatial resolution of GMTED2010. How exactly is this data "regridded on the TROPOMI grid"? I assume that one TROPOMI pixel covers many GMTED2010 pixels, and simple interpolation of GMTED2010 data to the TROPOMI center pixel coordinates would not be appropriate. Instead, all covered GMTED2010 pixels should be averaged.

→ The resolution of GMTED2010 (0.0625°, <https://temis.nl/data/gmted2010/>) is now being indicated in the Section 2.2.2 of the revised version of the manuscript. A TROPOMI pixel generally covers only one GMTED2010 pixel. It was also corrected on Figure 1.

Line 110: "resulting in a purely horizontal calculation of emissions": The considered transport is "purely horizontal". Emissions are calculated as sum of divergence and sink term (proportional to the column).

→ It was meant here that the model was using 2D arrays in its calculation of emissions. The words "purely horizontal" have thus been changed to "purely 2D" in the revised version of the manuscript.

Line 113: Of course the approach requires input data like wind fields which have uncertainties. But on top, the assumptions (stationary state and a pollution concentration close to the ground) are probably just wrong afar from strong local emission sources.

→ This precision has been added in the revised version of the article manuscript.

Line 130: Note that even without NO_x from lightning and soil emissions, there would be a free tropospheric background from long range transport of NO_y & PAN.

→ This precision has been added in the revised version of the article manuscript, with a reference (Zhai et al., 2024).

Line 139: This reads as if the enhanced NO₂ over ship tracks is caused by lightning NO_x, but it is primarily caused by direct ship emissions.

→ Shipping emissions enhance the NO₂ column but also influence lightning activity that creates NO_x. To avoid any conclusion, we changed the sentence highlighted by this comment.

Line 141: Sink term: $k[\text{OH}]$ needs to be multiplied with Ω .

→ This typo has been corrected in the revised version of the manuscript.

Line 143: Please provide more specific information, e.g. global maps of NO_x lifetimes from CAMS for Jan and Jul, which might be added to the Supplement.

→ The sentence relative to this comment has been made more specific. Two global maps of NO_x lifetimes (DJF and JJA) have also been added in the revised version of the Supplement.

Line 150: In particular, CAMS cannot resolve the extreme conditions within power plant plumes. This should at least be mentioned.

→ This text has been added in the revised version of the article manuscript to account for this comment.

Equation 4: I think that the topographic correction must be determined from Ω , not Ω' , as it describes the change of the *background* column when blown over a inhomogenous terrain. With Ω' , the effect will be underestimated.

Equation 5:

- is the divergence calculated for Ω or for Ω' ?

- $k[\text{OH}]$ needs to be multiplied with Ω .

→ These typos have been corrected in the revised version of the manuscript.

Line 239: Tehran was discussed in Beirle et al., 2023 (Fig. A1 therein) as example for the benefit of the topographic correction: without correction, emissions were negative, but with topographic correction, maps were far more plausible. I suspect that the topographic correction does not work properly here, which might be due to the way of interpolation of the GMTED2010 data to TROPOMI grid and/or the choice of Ω' instead of Ω in Eq. 4. Please check.

→ The reasons why Beirle et al. (2023) are more plausible is that they studied Los Angeles, USA, Tehran, Iran, Seoul, South Korea, and the Shanxi province in China with three different empirical scale heights: $H = 0.5$ km, 0.666 km and 1 km. This differs from our study, which uses a scale height of $H = 3.333$ km (X_e factor = $1/H = 0.3 \text{ km}^{-1}$). Our value of X_e (or H) is based on Sun, 2022 (Supplement of <https://doi.org/10.1029/2022GL101102>), for which $H = 1/X_e = 3$ km on average over the year, based on the US mountainous cells above the US. Here, we have preferred to use this last order of magnitude as it represents the average value of a times series deduced by fits, rather than the empirical values of Beirle et al. (2023), while admitting that using smaller values of H seems to solve the problem better for the cities mentioned above. Note that the choice of Ω instead of Ω' would not solve the issue, as the difference between the two is very low in polluted areas (generally $\Omega - \Omega' \ll \Omega$). We will however mention the work of Beirle et al. (2023) for this aspect in the dedicated section in the revised version of the manuscript.

Fig. 4: The extreme outlier of $-173 \text{ Pmolec/cm}^2/\text{h}$ occurs in January, while Fig. 4 refers to annual means. But even if values would be close to 0 for the other months, the annual mean at that pixel would be about $-14 \text{ Pmolec/cm}^2/\text{h}$, i.e. there should be at least one "negative" pixel with absolute value > 10 . Why does this not show up in Fig. 4?

→ This is because this pixel is outside the interval $[65^\circ\text{S}, 65^\circ\text{N}]$, precisely $(-75.90625, -153.78125)$ in Antarctica. As explained above, all the analyses in this article were carried out for this domain (corresponding to the area shown in Figure 2). Note: for this pixel, the only months for which there is at least one observation are January, February, March and December, with estimated emissions of -172.88 , 2.21 , 0.01 and $-63.89 \text{ Pmolecules.cm}^{-2}.\text{h}^{-1}$, i.e. a final average value of $-58.89 \text{ Pmolecules.cm}^{-2}.\text{h}^{-1}$ for 2022.