Replies to comments provided by Anonymous Referee #1

Title: Quantifying NOx emissions in Egypt using TROPOMI observations
Author(s): Anthony Rey-Pommier et al.
MS No.: acp-2021-1051
MS type: Research article

We would like to thank the reviewers for their careful reading, that led to interesting comments. Some minor points have been addressed in the revised manuscript. For a better readability, reviewer comments are highlighted in grey in this document, while changes in the manuscript are highlighted in orange.

In response to the reviews the authors have submitted a substantially modified manuscript. I thank the authors for the extended replies to my comments. In particular, I am happy to see that the authors repeated the calculations using the vertical tropospheric column instead of the slant column.

Below I repeat the points from my first review (indicated with a >) and comment to the replies provided by the authors to these points (see the author response document). A couple of minor issues remain (see below), and I ask the authors to address these before the paper is published.

One more major point is on the chosen grid (resolution of 0.1 degree) and the interpolation methods used. However, adapting this first basic step in the calculation would imply repeating all calculations, which implies a very major delay. Furthermore, to my opinion the results of the paper, error estimates and discussion of all aspects involved are valuable and sufficient to be published in the form presented in the revised manuscript. I do not expect that improving the gridding approach would lead to fundamentally different results.

To conclude, for me the paper is ready to be published after my other (more minor) points have been addressed.

> In this interesting paper the authors use the continuity or mass closure equation to derive emissions from the TROPOMI NO2 column observations. The authors discuss all the relevant ingredients of the calculation and provide estimates of the uncertainty. I am in favour of publishing, but with substantial revisions in response to a large number of questions provided below.

> Why Egypt? I understand there are many cloud-free days over the desert. Does the method require entire regions to be cloud-free? Or could it be applied to France just as well?

Egypt is motivated by the number of overpasses without cloud cover. This motivation is fine for me. About the chemistry: I appreciate the reformulated discussion of the different reactions and deposition mechanisms in section 3.1.

> Section 2.1: "We use TROPOMI NO2 retrievals from November 2018 to November 2020". Please provide details. Which version (versions) is used?

The version of the product is still not provided. Please add! The TROPOMI data product has changed significantly over time, see for instance v Geffen et al.(https://doi.org/10.5194/amt-15-2037-2022). The TROPOMI measurements are key to the paper, but the dataproduct and quality of the data is discussed only superficially. Instead of the Compernolle [83] reference: why not refer to Verhoelst, https://doi.org/10.5194/amt-14-481-2021?

This article (Verhoelst) should be more convenient than a technical report (Compernolle) indeed. We thank you for the reference.

> Section 2.1: "TROPOMI sounding are gridded for this study at a spatial resolution of 0.1 × 0.1". The authors mention that the resolution of TROPOMI is 3.5 x 5.5 km. So the choice of the grid is a bit disappointing (11x11 km). Why choose this resolution and not a higher one? Please provide details of how the gridding is done. Is this conserving NO_2?

The motivation for the grid chosen ("due to technical constraints") is not satisfactory. It should not be an issue to perform the calculations at higher resolution. Also the averaging procedure is not very satisfactory. Partial overlaps between TROPOMI and the grid cell are not accounted for it seems. The authors claim "The observed plumes remain correctly resolved" but there are clearly plumes to be expected with smaller scales than 0.1 degree. It seems to me the authors are not fully using the information which is provided by the TROPOMI instrument.

One minor advantage of the chosen grid resolution of 0.1 degree is the match in resolution with the emission inventories.

As mentioned above, I do not think that changing (improving) the gridding approach would lead to fundamentally different results so I suggest to move ahead with the results as presented in the revised manuscript.

The original gridding of the data was done in order to work with a decent resolution without having holes in the TROPOMI maps which limit the divergence calculation (a gridding with a better resolution leads to holes in the overlaping with TROPOMI). The resolution of 0.1°×0.1° seems to be a good compromise for this (at least for the calculation of monthly averages).

> Section 2.3: "Therefore, the CAMS OH concentrations are used". The resolution of CAMS is not very high, 0.4 degree. Given non-linearities and dependency on NO_x, would the use of CAMS OH be a good choice? What are typical uncertainties, in particular those linked to the downscaling from 0.4 degree to 0.1 degree?

There are comments on the relatively small variability in OH which "means that CAMS concentrations are able to produce a realistic concentration gradient" I do not understand this argument. I would reason that the resolution is low which implies the model will not be able to resolve gradients in chemical concentrations (e.g. across the Nile delta), and therefore the range in OH may be largely underestimated. Also, it seems that Fig.5 suggest quite a wide range of concentrations/lifetimes.

(Note: One reason why I mentioned France as target area is because of the availability of the CAMS regional air quality forecasts at 0.1 degree, the same resolution as the grid chosen.)

Generally speaking, the concentrations displayed by CAMS (40 km resolution) do not change abruptly from a pixel to another, as explained in the previous answer document. This is the case for all the Egyptian domain that is used here. The question is to know whether these low gradients are realistic or not. The Nile Delta concentrates a large part of the emissions and has many cities with large spatial extents, sometimes similar to CAMS resolution. We conducted WRF-Chem simulations with a resolution of 10 km above the Nile Delta region that also showed weak OH gradients throughout the year, except in Alexandria and, to a lesser extent, in Cairo during summer. The region therefore acts as a diffuse emitter and CAMS can therefore be considered to reproduce the chemistry of the region relatively well (however, we note that wind patterns are sometimes not represented). On the other hand, for the southern part of the domain, where the activity is concentrated along the river, we can expect higher gradients. This is not what CAMS shows. For this region, it is therefore possible that the range in OH is greatly underestimated, with actual concentrations that are veryhigh in the immediate vicinity of the river. Unfortunately, we do not have simulations available for this part of the domain to confirm it or not. In the section dealing with uncertainties, we added :

> "Due to the coarse resolution of CAMS data, OH gradients might also be underestimated, especially in the southern part of the domain, leading to a local under-estimation of the sink term."

> Section 2.4: "It is therefore necessary to remove the natural part of the atmospheric signal " We do not expect a lot of lightning and soil emissions over the desert. How large a signal is expected, why is removal needed, and how is this done?

I appreciate the new discussion on mechanisms responsible for background NOx observed in the TROPOMI data.

The authors write "Removing this natural signal is necessary for two reasons. Firstly, the natural part of the TROPOMI signal has to be removed from the emissions in order to interpret the results in terms of human activities. Secondly, the model uses quantities (wind, [OH], NOx to NO2 ratio, etc.) calculated in the lower troposphere. Applying it to a signal that is not entirely located in the troposphere would not make sense."
It could be useful to the reader to add these sentences to the paper.

In Section 3.4, we replaced :

> "These removed emissions are linked to the NO2 background estimated by TROPOMI, and do not correspond to anthropogenic emissions. They provide the value of what must be substracted from the estimates to obtain emissions related to human activity."

by :

> "These removed emissions are linked to the NO$_2$ background estimated by TROPOMI. This background, which is mostly located in the upper troposphere, is inconsistent with the use of other parameters which are calculated in the lower troposphere. As such, these emissions do not correspond to anthropogenic emissions, but they provide the value of what must be substracted from the estimates to obtain emissions related to human activity."

> Section 2.4: "We conduct this removal by subtracting the mean emissions over desert and rural areas from the mean emissions over urban and industrial areas. " Should "emissions" be "NO2 tropospheric column concentrations" here? Later in the paper there is a background emission term introduced. Why are background corrections not applied to the concentrations?

Thanks for the answer and the revised discussion in the paper, which I find satisfactory.

> Section 2.5: The CAMS emissions also seem to rely on EDGAR and will use similar approaches/assumptions and input datasets. Please comment on how independent or dependent these two datasets are.

Thanks for the extra clarification on the differences.

> Section 3.1, line 184: "Slant column densities are used as vertical densities" This does not make any sence to me, and should be a large and unnecessary source of uncertainty. The simplest approach to the air-mass factor would be a geometric path length of the incoming and outgoing light which depends on the viewing angles and is > 2.0. So, neglecting the air-mass factor can easily lead to 50% errors. Why is this better than using the air-mass factors from the retrieval?? Furthermore, the slant column will include (be dominated by) the stratosphere. Why not use the tropospheric column? As mentioned, the sink is modelled as concentration

divided by lifetime. But this concentration should be the column in the lower troposphere only, otherwise it does not make sense?!

I am happy to learn that the AMF is now used (e.g. the tropospheric columns are used).

> Equation 3: What is the omega_NO2 in this formula. Is it the slant column from TROPOMI?

OK

> Section 3.2. The discussion focusses entirely on electricity consumption, motivaing that 13:30 is representative for the daily mean. However, I would expect that traffic (industry) is also a major source of NOx, and this has a distinct diurnal (seasonal) pattern. So the discussion seems to be over-simplified.

Thanks for the changes in the text in section 3.2, which is now more balanced.

> Line 258: The city of Riyadh has been extensively discussed by Beirle et al., 2019. A reference to this paper in section 3.3 should be added.

Thanks for adding this to the new manuscript.

> Line 263: sqrt(w^2) = w. The notation is a bit unclear.

OK

> Equation 7: I still have a conceptual difficulty with a "rural emission". Over the desert the estimated emission should be close to =0 and negligible compared to urban emissions, otherwise the methodology is flawed.

Thanks for modifying the text and equation. I am satisfied with the response.

> line 324: "limit the high inter-day variability due to changing wind patterns or differences between week days and week-ends". What is the real reason averaging over a month is needed? Winds change, but if the method is correct the emissions should be equal (assuming stationary sources).

I still think that the sentence, which was not modified, is misleading. In their response the authors focus on uncertainties, which makes more sense. I would suggest to rephrase the text of the paper accordingly.

We replaced :

"limit the high inter-day variability due to changing wind patterns or differences between week days and week-ends"

by :

"limit the outliers due to uncertainties in wind and OH representation."

> l 359: "Level B is therefore the one that leads to the best match between the lifetime calculated with Equation (2) and the lifetime calculated from line densities." What does this really prove? Does it really mean Level B is better? Due to the coarse resolution we may expect CAMS is biased in OH since it does not resolve the plumes.

The abstract mentions "It it also provides the location of the most appropriate vertical level to represent typical pollution sources in industrial areas and megacities in the Middle East region." I'm not convinced yet about this conclusion and think it is a rather bold statement. The correlations between CAMS and EMG are low for both A and B, and the slope in both cases is close to 1. It is valuable to see the comparison with the EMG method, but I wonder if the comparison alone can be used as evidence for the selection of an appropriate level.

In the abstract, we replaced :

"It also provides the location of the most appropriate vertical level to represent typical pollution sources in industrial areas and megacities in the Middle East region."

by :

"It also provides some hint on the vertical levels that best represent typical pollution sources in industrial areas and megacities in the Middle East region."

In Section 4.1, we replaced :

"Level B is therefore the one that leads to the best match between the lifetime calculated with Equation (2) and the lifetime calculated from line densities."

By :

"Although both correlations are weak, level $\mathcal{B}$ leads to a better match between the lifetime calculated with Equation \eqref{eq:lifetime} and the lifetime calculated from line densities."

> Figure 6: Before showing this, I would suggest the authors apply the method to Riyadh and compare with Beirle et al. (2019) to test the consistency of the results.

In their response the authors give numbers for the comparisons which are interesting and relevant, because they provide an alternative insight in uncertainties in the method. Beirle is mentioned in Sec 3.3, but the comparisons in terms of total emissions, lifetime, relative importance of the sink term vs transport term is still not provided. I suggest the authors copy some of the numbers from their response to the paper to highlight these substantial differences in the estimates.

In Section 4.2, the following paragraph will be added :

"First, we try to map NO$_{\text{x}}$ emissions in Riyadh using parameters estimated at level $\mathcal{B}$. For the period from December 2017 to October 2018 and using a constant lifetime of 4 h, Beirle et al., 2019 \cite{beirle2019pinpointing} estimated at 6.66 kg.s^{-1} the emissions of the corresponding urban area, and a mean rate density of about 3.7 µg.m^{-2}.s^{-1} for power plants PP9 and PP10/14, the transport term accounting for about 80 to 90\% of this budget. Using the same domain for December 2018 to October 2019 with our method, we found a mean lifetime of 2.94 h and mean emissions of 5.92 kg.s^{-1} for the urban area. We also found smaller rate densities for the power plants (about 3.4 µg.m^{-2}.s^{-1} for PP9 and 3.0 µg.m^{-2}.s^{-1} for PP10/14), with a smaller contribution of the transport term (about 70\%). Despite differences in resolution, AMF calculation, lifetime variability and background removal, the two methods give similar results."

> Table 1: I would suggest to replace "khab/km^2" by "10^3/km²"

I am satisfied with the response.

> l 420: "It is also observed that TROPOMI NO2 column densities above this zone are relatively homogeneous". As demonstrated in several papers, there is a clear shipping signal in the TROPOMI data over oceans and seas, and I would expect TROPOMI to be rather inhomogeneous here?!

I am satisfied with the response.

> Figure 8: The unit is "kt" which I assume is 10^6 kg. But what is the time unit? Per hour, per day, per year? I'm a bit surprised by the big scatter for the weekly (daily) values averaged over the entire country?

I am satisfied with the response (unit is now kt/d). Maybe good to mention once in the text that "kt/d" means kilotons per day, to avoid confusion.

In Section 4.4, we added :

"emissions are expressed as NO and in kilotons per day"

> Section 4.5, Covid-19. There is a nice review paper, https://doi.org/10.1525/elementa.2021.00176, which could be added here.

Thanks for adding this review paper.

> l 488: "no significant changes in OH concentrations ". Does the CAMS system describe the change in emissions and concentrations observed resulting from the lockdown? If not, how would this impact the results (given the non-linearity of the chemistry) ?

I think it would be useful if the response of the authors (no changes in emissions during lockdowns, but possibly some impact through the assimilation of satellite data) is also added to the text of sec. 4.5.

The following text has been added :

"CAMS OH concentrations during the lockdown periods do not show significant variations from previous and subsequent years, although values are slightly lower in 2020 than in 2019 (about 5.5\% lower over the mask cells for the period March/April/May). The near-real-time CAMS system did not take into account the decrease in anthropogenic emissions in the representation of its OH concentrations. However, the satellite constraints inherent in the system may have modulated the lockdown effects locally or globally. Given the non-linearity of the chemistry but also given the large reactivity of OH with other species whose concentrations have varied differently during the lockdown, it is difficult to determine how these observations have impacted OH concentrations."

> l521: "TROPOMI-inferred emissions show an annual variability" I was wondering how much we can believe the seasonality in OH as modelled by CAMS? This seems to directly link to the seasonality of the sink term and, as a consequence, the emission estimate. Please discuss.

I am satisfied with the response.

> l551: "S-5P validation activities" Please add a reference

As above: I suggest Verhoelst et al.

This reference has been added.

> l 558: "For [OH]," The authors showed that OH is strongly height dependent, so it seems that the choice of the vertivcal level is a major uncertainty. Has this been accounted for?

I am satisfied with the response.

> Data availability: TROPOMI data is missing here.

I am satisfied with the response.

Replies to comments provided by Anonymous Referee #2

Title: Quantifying NOx emissions in Egypt using TROPOMI observations
Author(s): Anthony Rey-Pommier et al.
MS No.: acp-2021-1051
MS type: Research article

We would like to thank the reviewers for their careful reading, that led to interesting comments. Some minor points have been addressed in the revised manuscript. For a better readability, reviewer comments are highlighted in grey in this document, while changes in the manuscript are highlighted in orange.

General comment: The proposed method for estimating emissions relies on a series of assumptions regarding the NO_2 sink terms (Section 3.1), which generally hold for the studied region (desertic with large anthropogenic sources). However, the application of this method to other regions might be problematic, as sink terms, e.g. NO_2 deposition on vegetation and NO_2 sink through organic (peroxy)nitrate formation, cannot be neglected. The authors should include a short discussion on the limits of validity of their method.

In the conclusion, a short discussion has been added between the first and the second paragraph :

> "Here, our estimation of NO$_{\text{x}}$ emissions benefited from favorable conditions. Egypt has a desertic climate, allowing to neglect many NO$_{\text{x}}$ loss mechanisms for the sink term calculation, a flat terrain on most of its territory, limitating wind field errors for the transport term calculation, and a large population concentrated in a small number of cities, providing NO$_2$ maps with large signal-to-noise ratios above urban and industrial areas. For other regions of the world that do not have such features, the method presented here must be modified accordingly. However, we expect this method to be applicable to most countries similar to Egypt without substantial changes. For Middle East countries, this study thus demonstrates the potential of TROPOMI data for evaluating NO$_{\text{x}}$ emissions. More generally..."

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l. 230-231, the 2nd channel is not minor because HOONO is rapidly decomposed back to NO_2 and OH. The important point is : the net effect of this channel on NO_2 distributions (and hence on the determination of emissions) is dependent on the rate of decomposition of HOONO, which is unknown. If the process is very fast, then it should not be taken into account in your k_mean. If it is sufficiently slow, it will not affect (much) the NO_2 local horizontal gradients, and therefore, it would be okay to incoporate the second channel in k_mean. I propose that you explain clearly your assumption, namely, that HOONO decomposition is not very fast.

We replaced :

> "Note that although this reaction rate accounts for both reactions with OH, the second channel is minor, because HOONO can be rapidly decomposed back to NO_2 and OH in the lower troposphere. The value of k_{mean} therefore represents the total loss of..."

by :

> "Note that HOONO can be rapidly decomposed back to NO$_2$ and OH in the lower troposphere. We assume here that this decomposition is slow and does not affect the NO$_2$ horizontal gradients. Both pathways are therefore taken into account, and the value of k_{mean} represents the total loss of..."

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l. 237, The range of 26-64% was wide because Stavrakou et al. considered the possibility of a high rate for the reaction $HO_2+NO \rightarrow HNO_3$ based on studies by Butkovskaya and co-authors (this reaction is neglected here).

We do not totally understand the intentions related to this comment. Indeed, Stavrakou et al. considered different sinks with different reaction rates, and the NO2+OH → HNO3 reaction accounts for about 45-65 % of the total NO_x loss, except in one case for which this share is reduced to 26 % while the share of the NO+HO2 → HNO3 reaction reaches 64 %. The way we understand it, this comment invites us to mention this article explaining that NO2+OH → HNO3 is the largest sink but with high uncertainties due to other reactions. Therefore, we replaced :

> "Stavrakou et al., 2013 [47] showed that the reaction between NO2 and OH forming HNO3 accounted for 26 to 64% of total NOx loss at the global scale."

by:

> "Stavrakou et al., 2013 \cite{stavrakou2013key} showed that the reaction between NO2 and OH forming HNO3 accounted for most of total NO$_{\text{x}}$ loss at the global scale, but with high uncertainties associated with other sinks."

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l. 249, "Different models": be more specific, provide emission estimates and references.

We replaced :

> "Different models have estimated low biogenic isoprene emissions in the region. These emissions are concentrated at the level of the Nile and its delta (Guenther et al., 2006 \cite{guenther2006estimates})."

by :

> "Different models have estimated low biogenic isoprene emissions in the region (Wiedinmyer et al., 2006 \cite{wiedinmyer2006futur}, Guenther et al., 2006 \cite{guenther2006estimates}). These emissions are concentrated around the Nile River and its delta, and do not exceed 15 mg.m^{-1}.day^{-1}."

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l. 277, I suggest replacing "suggesting a small yield" by "possibly due to the high temperatures favoring short PAN lifetimes".

The sentence has been modified accordingly.

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l. 284, "minority" --> "minor"

The word has been changed accordingly.