Replies to referee comments

Title: Quantifying NOx emissions in Egypt using TROPOMI observations
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We would like to thank the reviewers for their careful reading, that led to interesting comments and subsequent improvements of the article. Several additional results (which will be discussed in this document) will be added in the revised manuscript. Figures presented in this document will be indicated with letters. When they correspond to a figure that appear in the original manuscript, it will be indicated in the corresponding caption.

Questions provided by *Anonymous Referee #1*

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?

→ If the question is adresses on the statistical level, the method can be applied to any region of the world, but the average cloud cover would severely limit the relevance of the monthly averages presented here. Approximately 90% of the TROPOMI observations in Egyptian territory are characterised by a quality assurance (q_a) value greater than 0.75, with low seasonal and spatial variations. The calculated monthly emissions are therefore obtained by averaging 22 to 30 days of observations. The averages would be less robust if the method were applied to France, whose territory shows q_a values lower than 0.75 about 50% of the time, with large seasonal and spatial variations (the northern part of France is characterised by low-quality values 20% of the time during winter months). This limitation cannot be compensated by calculating an annual average, as this would lead to an over-representation of summer days in the calculation, but an intermediate sampling period (e.g. 2 or 3 months) could be relevant. However, if the question is adressed on the theoretical level, the limitations come from the chemistry: many NO_x loss mechanisms (other than the reaction with OH) can be of significant importance in other regions, and their moddeling can be very complex. The method can be applied to these other regions only if the sink term is modified accordingly.

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

→ The only version of TROPOMI data that have been used is the version S5P_OFFL_L2. This will be precised in the revised manuscript.

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 NO2?

→ The gridding is performed by averaging the corresponding TROPOMI observations for each cell. When a cell correspond to several measurements (in general 2), then the average column value is given. The choice of the resolution (11×11 km) corresponds to technical constraints. Moreover, gridding the data with a lower resolution leads to several areas without data, which complicates the estimation of the derivatives in the transport term. Conducting a separate sensivity test of the method above Qatar for several months in 2020-2021 with a higher resolution (6.5×6.5 km), it seems like the change of resolution has no impact on monthly estimates.

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 NOx, 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?

→ Here, we do not have data on OH concentrations with a better resolution, hence the downscaling to match the resolution of TROPOMI columns. Nevertheless, spatial variations from one cell to another are not abrupt. In the Nile delta area (boundaries: 30°E to 33°E; 28°N to 31°N), the ratio between 90th and 10th percentiles does not exceed 1.9, while the maximum/minimum ratio does not exceed 4, which means that CAMS concentrations are able to produce a realistic concentration gradient. However, it is difficult to evaluate the associated uncertainty: OH is very challenging to model due to its fast cycling and dependence on many sources and sinks, which makes the quantification of its uncertainties even more complicated at all scales.

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?

→ TROPOMI observes a NO_2 tropospheric background (i.e. a non-anthropogenic signal) of ~0.5×10^{15} molecules/cm². In the lower troposphere, natural NO_x emissions are due to fires and soil emissions. In the upper troposphere however, sources include lightning, aircraft, convective injection, and downwelling from the stratosphere (Ehhalt et al., 1992; Jaeglé et al., 1998; Bertram et al., 2007), but the factors controlling the resulting concentrations are poorly understood. According to state-of-art estimates, anthropogenic NO_x accounts for 60-70% of the annual global NO_x emissions, whereas natural emissions from fires (biomass burning), soils and lightning are less significant at the global scale and are estimated at about 9%, 17% and 6% respectively (Jaeglé et al., 2005; Müller and Stavrakou, 2005), although associated errors can be very high. In eastern China, Lin, 2012 has shown that the non-anthropogenic share of total NO_x emissions ranked to 8% with a maximum of 18% in July. Egypt being a desertic region and not very conductive to lightning (Albrecht et al., 2016), we expect the share of those non-anthropogenic emissions to be much smaller than 10%, especially in dense areas, with a corresponding signal located in the upper troposphere. 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], NO_x to NO_2 ratio, etc.) calculated in the lower troposphere. Applying it to a signal that is not entirely located in the troposphere would not make sense. We will clarify this point in the revised manuscript. In Section 3.4, we calculate an emissions-equivalent background, defined as the mean value of emissions obtained above rural cells. This value does not correspond to natural emissions because the quantities involved are vertically inconsistent. However, it provides an equivalent of what needs to be removed from total estimates to obtain anthropogenic emissions.

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?

→ This comment is linked to the remark that has been made above. Indeed, to avoid calculating unrealistic emissions in the desert, it is possible to make a concentration correction by removing the observed upper tropospheric background before calculating the emissions rather than calculating the emissions with this upper tropospheric background and then obtaining the anthropogenic emissions by removing the "equivalent tropospheric background emissions". The two methods are not mathematically equivalent, and both present practical issues. For instance, defining a background with the same mask but based on concentrations instead of emissions could lead to errors in emissions, because high concentrations can be spotted outside "urban" cells due to wind transport ; they would be responsible for a over-estimation of the background (an example of this is illustrated on Figure 1 (right panel), where very high concentrations cover "rural" cells). To avoid such over-estimations, it is possible to define a background concentration based on the lowest values for TROPOMI NO_2 retrievals (typically by taking the 10th or 20th percentile value of of all columns in the domain) rather than using a mask. This is perfectly feasible for our large domain in Egypt, for

which the majority of cells have low NO_2 concentrations. However, it would be less feasible for a domain of smaller size centered on a city centre or a power plant (such as the domain centered on Riyadh as shown on Figure 3).

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.

→ CAMS-GLOB-ANT_v4.2 use EDGARv4.3.2 (not EDGARv5.0) as a basis for 2010 emissions and the emissions between 2011-2014 are obtained using this basis and the trends in CEDSv3 (Hoesly et al., 2018). Those trends are then used to project NO_x emissions for 2018 (Granier et al., 2019). This is discussed in Section 2.5.

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?!

→ Concerning the first question, we took this comment into account and used the air mass factor provided by TROPOMI. The effect of this correction on our method is to decrease NO_x emissions estimates (by about 17% on average), except for winter 2019/2020 where the it leads to a small increase. Its use will be presented in details in this response to a referee comment: [p.10]. Concerning the second question, it is indeed the tropospheric column that is used. This will be clarified in the revised manuscript.

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

→ Ω_{NO2} corresponds to the slant column indeed ; it is introduced at Line 172 in the original manuscript. In the revised version, it will correspond to the vertical column.

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.

→ According to CAMS_GLOB_ANT_v4.2, the power sector accounts for 50 to 60% of total NO_x emissions in Egypt (this number increases up to 55 to 70% if the residental sector, which has a smiliar pattern to electricity, is added). EDGARv5.0 presents a lower share (40 to 45% of total emissions for the power sector, 45 to 50% if the residential sector is added). For both inventories, the transport sector accounts for the majority of the remaining emissions. Moreover, the daily pattern for traffic emissions seems to indicate that emissions at 13:30 are close to the daily mean emissions : congestion index in Cairo (https://www.tomtom.com/en_gb/traffic-index/cairo-traffic/) at this moment of the day seems to be slightly higher than during the morning peak, but lower than the during night peak. Emissions around 13:30 would therefore remain quite representative of the average emissions in the country. All these indications will be added in the revised version of the manuscript.

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.

→ The manuscript has been modified to cite Beirle et al., 2019, but also Valin et al., 2013, which conducted another study about Riyadh’s emissions.

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

→ Line 263 and Eq. 5 have been modified in the manuscript to make the notation more clear.

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.

→ Maps in Figure 6 show total emissions, including anthropogenic emissions. To get the anthropogenic emissions, it is necessary to substract the inferred emissions outside the mask from total emissions, as in Equation 7. Figure 6 illustrates how emissions are calculated. In the revised manuscript, Figure 6 will consist in four maps instead of three, with the addition of a map showing anthropogenic emissions (see example below, Figure A). Furthermore, Equation 7 will be re-written and clarified in the revised manuscript to avoid any confusion.

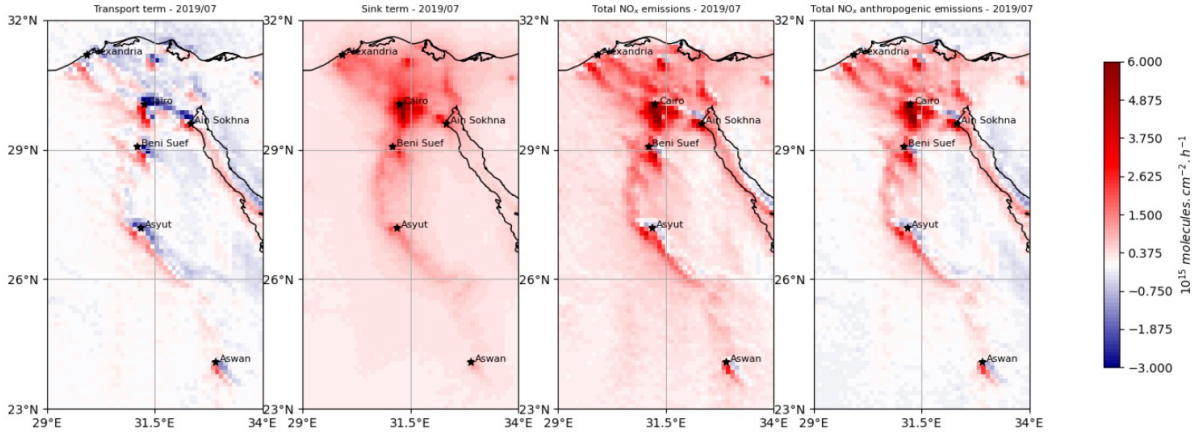

Figure A: Correction of Figure 6 after correction with air mass factor (transport term, sink term, total emissions on the left), for which the last step in estimating nitrogen oxides emissions by background removal is displayed (right).

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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).

→ The calculation of the divergence term is very sensitive to the wind direction, and an uncertain representation of the wind in ERA5 would lead to errors in the emission calculation, especially near large emitters (large industrial plants) for which the divergence term can be much higher than the sink term. Averaging over a month limits the effects of this sensitivity. This assumes that errors in the ERA product are not systematic.

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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.

→ A high NO_2 resolution is needed to derive the lifetime (Valin et al., 2011). However, the lifetime calculated by $\tau = (k_{mean}[OH])^{-1}$, is computed using the mean OH value above Riyadh. Riyadh's size is about 30×50 km², which is close to the CAMS resolution. It is therefore difficult to know the impact of the resolution on the estimation of the mean value in OH. On one hand, provided that this effect is uniform across all the samples presented in Figure 5, then the value of the correlation coefficient would not be affected but such a change, and it would still be higher for level B than for level A. On the other hand, this effect could have an effect on the slope presented: if this effect leads to an underestimate, then slope A would be further away from 1 than slope B and level B would remain the most adequate. If not, then level A will be the most adequate and it will be more difficult to gauge which level is the most representative.

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.

→ Indeed, this has been done to test the method. Using the method in the domain centered around Riyadh (boundaries: 46.0°E to 47.5°E; 24.0°N to 25.5°N) for 2018/12 to 2019/10, we obtain a value of 5.10 kg/s for total emissions in Riyadh's urban area, which is close to the estimate of 6.66 kg/s found by Beirle et al., 2019 (for 2017/12 to 2018/10). Using a meridional cross section at 47.05°E, we observe emissions from the two power plants PP9 and PP10. While Beirle et al., 2019 estimated a rate density of ~3.5 µg/m²/s (with ~80% of this value being due to the transport term), we estimate a rate of ~1.7 µg/m²/s with ~60% due to the transport term (see below, Figure B). Note that Beirle et al., 2019 have used a constant lifetime of 4.0 h, while our analysis indicates a mean value of 2.94 h according to Equation 2. If they had used this value, they would have found 7.86 kg/s for total emissions. However, the emissions of PP9 and PP10 would not differ much since they are mostly represented by the transport term. Our model is therefore consistent with the results of Beirle et al., 2019 in the sense that spatial variations and total budgets are reproduced in a similar way. We tend to infer lower emissions at both small and large scales. This comparison has its limits, due to our coarser resolution, our different modelling of the AMF and the "background" value, and the fact that we study two different years)

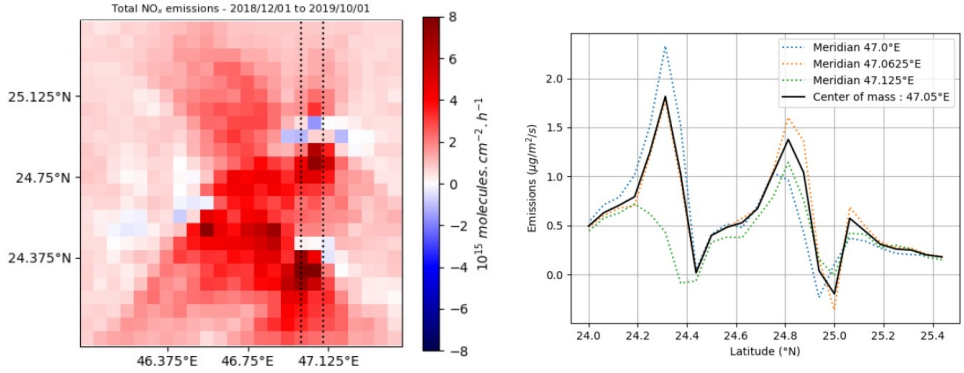

Figure B : NO_x emissions above Riyadh's urban area (left) according to our model and meridional cross-section (right) around 47.05°E with identifications of power plants PP9 and PP10. This figure can be compared to Figure 2 in Beirle et al., 2019.

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

→ For readability puroposes, we will replace "khab/km²" by "people per square kilometer" in the revised manuscript, to specify the nature of quantities without units.

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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?!

→ We agree with this review and have revised our interpretation. The reagion acts as a very thin line of emitters, which leads to a very large value for the transport term. The TROPOMI signal in this area is clear and contrasts with the NO_2 background, but it remains quite low compared to the emissions in the main cities).

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?

→ The y-axis corresponds to kilotons per day (kt/d). Points that constitute grey lines corresponds to daily emissions. Low values in this figure illustrate the need for monthly averages in the general method we are using throughout the article. For instance, the calculation of emissions for a particular day with no observations or low-quality data ($q_a < 0.75$) over the strongest emitters generally leads to an underestimation of real emissions. To illustrate this point, we can use the lowest estimate for Fridays, which corresponds to the 06/12/2020 with 0.036 kt (framed in red on Figure 8 that has been reproduced and updated below, Figure

C). On this day, there is a significant part of the urban domain without reliable TROPOMI data, in particular around the urban areas of Cairo and Alexandria which are strong hotspots of the country. These situations happen frequently, but most of the time they are not taken into account in the annual mean (and consequently not displayed on Figure 8), because more than 20% of the domain is covered with low-quality data. Such cases could be removed from the average by lowering the latter limit to 15 or 10%. However, because in this case the annual mean would be over-estimated, this would alter our weekly estimates.

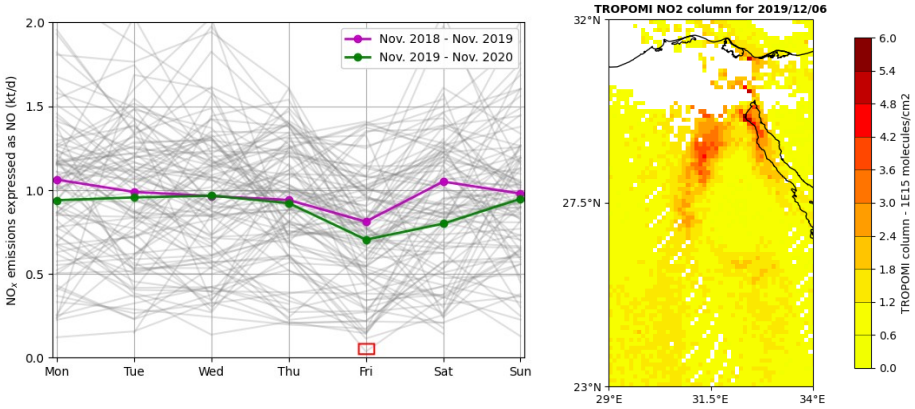

Figure C: Correction of Figure 8 after correction with air mass factor (left) and map of TROPOMI columns during the day corresponding to the lowest NO_x estimates (right). This day (2019/12/06) corresponds to the point framed in red on the left.

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

→ This citation will be added in the revised manuscript.

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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)?

→ CAMS OH concentrations during the lockdown periods do not show significant variations from previous and subsequent years (Figure D), although values are slightly less elevated in 2020 and 2021. 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 with different ways during the lockdown, it is difficult to *a priori* determine how these observations would impact OH concentrations.

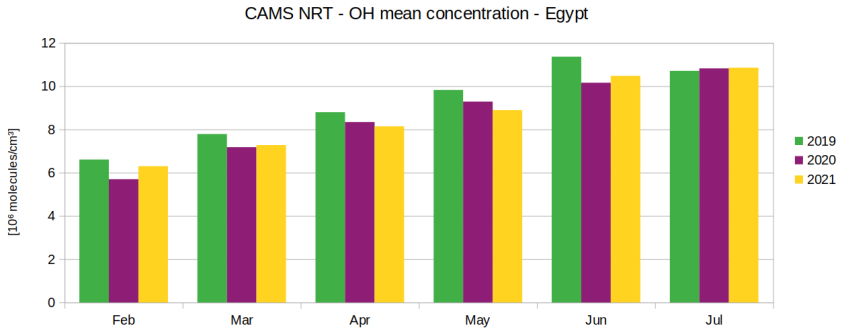

Figure D: Mean OH concentrations above Egypt for different months.

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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.

→ Indeed, OH concentration, which intervenes linearly in the expression of the sink term, has a major role on the monthly emission estimates in the same way as TROPOMI columns. Thus, the ability of

CAMS data to reflect human activity should be evaluated. We do not have precise information on how CAMS represents this activity in Egypt, and the non-linearity of the chemistry involving OH does not allow us to anticipate the corresponding seasonality. The chemistry-transport models that we have do not allow us to estimate it correctly because their resolution is not sufficiently high to assess what is happening in polluted regions. Note that a referee comment addressing the seasonal cycle was also addressed and led us to calculate monthly emissions on smaller domains. These emissions are more pronounced in areas with large polluters, with a seasonal cycle which is consistent with emissions at the scale of the country, indicating that our estimates are not directly linked to the variability of OH. We invite you to read the corresponding answer here: [p.10]

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

→ The reference added in the revised manuscript is "Compernolle, S. (2018). S5P MPC VDAF Validation Web Article: Nitrogen dioxide. S5P-MPC-VDAF-WVA-L2_NO2_20180904".

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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?

→ The effect of the vertical was taken into account in the relative uncertainty on OH. We will detail the composition of this uncertainty in the revised version of the manuscript.

Data availability: TROPOMI data is missing here.

→ The following link will be added in the revised manuscript: http://www.tropomi.eu/data-products/data-access.

Questions provided by *Anonymous Referee #2*

- l. 142: and following: the "urban" pixels (>100 hab. km-2) are not all truly urban. Croplands in Egypt are located almost exclusively within the "urban" cells of Figure 1, whereas the non-urban pixels are mostly (semi-)desertic. Therefore I doubt that the removal of the non-anthropogenic part of the NOx emissions discussed here makes any sense. Soil NOx emissions are primarily located within the Nile delta and Nile Valley. Same holds for agricultural residue burning, a substantial source of pollution in Egypt (https://egyptindependent.com/agricultural-burning-clouding-sky-sickness/).

→ The largest sites of anthropogenic emissions outside the Nile Valley and delta are counted as "urban" cells. We acknowledge the words "urban cells" and "rural cells" can be misleading. Instead, we will drop this separation and use the terms "inside mask" and "outside mask" in the revised manuscript.

- l. 174-184: Equation (1) does not make much sense if slant column densities (SCD) are used as vertical column densities (VCD). The air mass factor (AMF) is generally different from unity, and furthermore, it varies in time and space. Contrary to the assumptions made here (Section 4.7), the albedo over the region is not uniform since we have areas covered by deserts, by crops, by water and by cities. The vertical profiles of NO2 can also be expected to vary according to the landscape. Those variations will impact the divergence term in Equation (1). The authors state that the AMF is taken "into account in the final uncertainty estimates". However, Section 4.7 only discusses the VCD uncertainty due to the AMF, (~30% following Boersma et al., 2004). This does not say anything about the impact of using SCD instead of VCD in Equation (1). We are worried that the real impact of this substitution is unknown.

→ This question deals with a subject that was addressed in a previous referee comment. We invite you to read the corresponding answer here: [p.3]

- l. 193: I don't understand "using a temperature-dependent analytical formula for different pressure ranges". Burkholder et al. provides a general expression of the rate as function of both T and [M]. Please clarify.

→ We used the formula in Burkholder et al., 2020 in which the temperature is taken from CAMS and [M] is calculated as $[M] = P/k_BT$ where P is the pressure level on which the calculations are conducted. The term "temperature-dependent analytical formula" will be been changed in the revised manuscript.

- l. 196-197: I don't understand "The value of k_{mean} therefore represents the total loss of NO2 due to OH and cannot be used to infer HNO3 and HOONO production". This is not clear. Only the first channel is a true NOx sink, therefore the other channel should be ignored entirely.

→ Bukholder et al., 2020 indicates that "The fate of HOONO has to be included in atmospheric models. If this fate involves rapid loss due to reaction or photolysis, the effect of the second pathway is the diminution of the HONO2 forming rate constant. Evaluation of data, taking into account both pathways, indicates that the contribution of the HOONO forming reaction can be from 5 to 15% under atmospheric conditions at 298 K". It is therefore difficult to conclude about the importance of this second channel without having more information about the fate of HOONO, which we do not have in models.

- l. 211-212: "Losses due to deposition and the formation of (...) nitrates are thus considered insignificant in Egypt where the forest cover is totally negligible": this is not correct. Forest cover might indeed by very low, but vegetation (mostly croplands) is present in the so-called "urban cells" of Figure 1 (leaf area index typically between 1 and 2 according to MODIS). Furthermore, TROPOMI HCHO maps show HCHO vertical columns over the Nile Valley and the Delta (>1 Pmolec cm-2) in summer, which are similar to values found in Southern Europe. This suggests significant NMVOC emissions, of biogenic and/or anthropogenic origin. Organic (peroxy)nitrate formation cannot be assumed to be negligible. There is very likely a significant net export of RONO2 and PAN compounds from the Nile area to the surrounding regions. A comprehensive model might be needed to evaluate its importance. Quite importantly, this export might be seasonally dependent, since organic nitrate formation is strongest in summer.

→ The HCHO columns that can be observed around the Nile in Egypt are indeed distinct from the background that can be observed in the rest of the region, which seems to indicate BVOC emissions. These compounds remove NO_x from the atmosphere through the formation of organic nitrates ($RONO_2$), competing with depletion through the formation of HNO_3. This competition between the two processes is strong in forested areas, but to our knowledge there are no studies focusing on croplands in the Middle East / Eastern Mediterranean region. Different models have estimated low, but not zero, biosphere isoprene emissions in the region. These emissions are concentrated at the level of the Nile and its delta (Guenther et al., 2006). They are certainly noticeable and higher in summer than in winter, and contrast with the rest of the country, but they remain low compared to most other regions in the world. They are, for instance, about an order of magnitude lower than in the forested areas of the eastern United States. In this region of the US, the $RONO_2$ sink accounts for between 30% and 60% of the total sink (Romer Present et al., 2020). Furthermore, at large NO_2 concentrations (compared to VOC concentrations), the share of $RONO_2$ sink in the total NO_x loss is weakened compared to that of HNO_3 (Romer Present et al., 2020). In a dense and highly polluted environment such as the Nile and its delta, and where isoprene emissions are low, it can therefore be hypothesised that the NO_x sink by $RONO_2$ chemistry is minor in comparison with that forming HNO_3 and in view of the uncertainties of the model, and thus that it has not been taken into account. However, NMVOC emissions can also be of anthropogenic origin, particularly in cities where they are difficult to estimate. To our knowledge, there is no study evaluating the competition of the two sinks in Egypt or in a region with similar features. This may be the subject of a future study. In the revised version of the manuscript, we will highlight this limitation in sections 3.1 and 4.7. The term "totally negligible" will be removed and replaced by a description detailing these minor sinks. Finally, concerning the NO_x sink by PAN formation, it appears that PAN concentrations in the lower troposphere (0-2 km) vary between 50 and 200 pptv around the Nile delta, while in southern Europe they are between 100 and 400 pptv (Fischer et al., 2014). We will precise this in the revised manuscript.

- l. 213: Regarding the HNO3-forming channel of the NO+HO2 reaction, note that field studies (e.g. Nault et al. 2015, doi:10.1021/acs.jpca.5b07824) indicated that this path is very minor.

→ This study will be added to the references in the revised manuscript.

- l. 216-217: Production of PAN might peak in the late afternoon, but it might still be significant earlier in the day.

→ This question deals with a subject that was addressed in a previous referee comment. We invite you to read the corresponding answer here: [p.8]

- l. 221-228: Why is electricity consumption assumed to be the best proxy for NOx emissions? Traffic and industry follow different patterns. According to current inventories, what are the respective relative contributions of the main sectors (traffic, industry, power generation) in Egypt? Some discussion is needed.

→ This question deals with a subject that was addressed in a previous referee comment. We invite you to read the corresponding answer here: [p.3]

- l. 258: "We therefore use the nearby city of Riyadh (...) to perform the comparison between the CAMS-induced lifetime and the fit-induced lifetime": despite some similarities, Riyadh and the Nile valley are quite different environments, with much more vegetation and NMVOC emissions in Egypt than around Riyadh, possibly impacting e.g. the wind profile, the OH fields and the NO2 profile. Therefore, the OH validation for Riyadh might be of limited value for Egypt. I recommend comparing the TROPOMI HCHO columns over Egypt and Riyadh.

→ TROPOMI HCHO columns can be spotted over Egypt and Riyadh. Between May and October, they are significantly higher than the HCHO background in the Eastern Mediterranean / Middle East region (see below, Figure E). The Nile region has slightly higher HCHO columns than Riyadh. (4-12 Pmolec/cm² for Riyadh and 4-16 Pmolec/cm² for the Nile region).

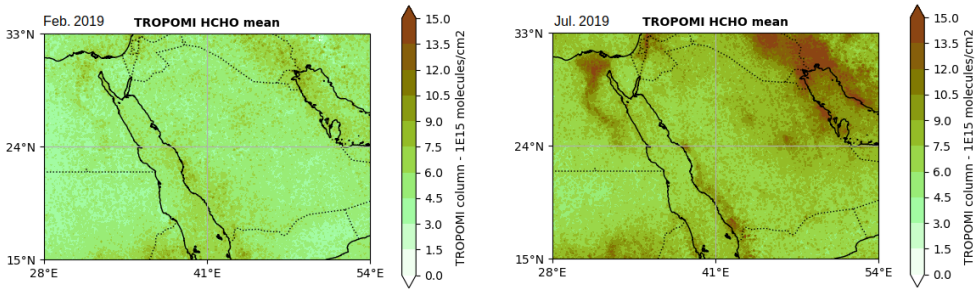

Figure E: Average TROPOMI HCHO columns above Egypt and Saudi Arabia for February and July 2018.

- Equation (7): As explained above, the "urban cells" do contain natural emissions. Moreover, the non-urban cells contain anthropogenic emissions. Those are less intense than in the Nile Valley and delta, but the natural emissions follow the same pattern. I recommend to drop this separation.

→ This question deals with a subject that was addressed in a previous referee comment. We invite you to read the corresponding answer here: [p.7]

- l. 389-406: What is the location and capacity of the main power plants in Egypt? Are the industries mentioned in the text (e.g. cement plants) really strong NOx emitters?

→ Cement plants are usually strong sources of NO_x. In Egypt, most of them are of average capacity (between 1 and 7 Mt/yr). Oil and gas power plants are also strong emitters, whose capacities rank between

20 and 2200 MW (median: 612 MW). Finally, depending on the processes used (which are not known here), iron smelters can be considered as large NO_x emitters as well.

- Section 4.6: The temporal variation of TROPOMI-based emissions seems very unrealistic. The 2019/2020 winter minimum would be explained by reduced electricity consumption due to reduced usage of air conditioning. But then why not in the previous winter? In any case, it should be possible to check whether the biggest power plants are the places where the seasonal behavior is most pronounced. And regions without any significant power plants should not exhibit this phenomenon at all. I doubt very much that air conditioning would increase so much the traffic-related NOx emissions. I think much more plausible that the temporal variations are due to errors in the methodology, in particular regarding NOx sinks and the air mass factors. I recommend to check whether the TROPOMI AMF (or an AMF recomputed using CAMS profiles) presents significant temporal variations. This requires a more in-depth analysis than is currently provided.

→ This comment raises the question of the reliability and the origins of the seasonal cycle observed in Egypt's NO_x emissions. Our response encompasses three considerations: we analyse the impact of taking into account the AMF on our estimates, we compare the inferred emissions of winter 2019/2020 with previous and subsequent winters, and we observe the dynamics of NO_x emissions near and far from fossil fuel-fired power plants, which will lead to major additions in the revised manuscript.

The TROPOMI AMF shows spatial and temporal variations. Overall, it is higher for rural cells than for urban cells, probably due to an albedo difference. Concerning seasonal variations, the AMF reaches a maximum in December/January and a minimum in May/June. These variations are of the order of 0.1-0.2, while average values inside and outside mask cells are about 1.6 and 1.8 respectively. The figure below (Figure F) summarises these variations.

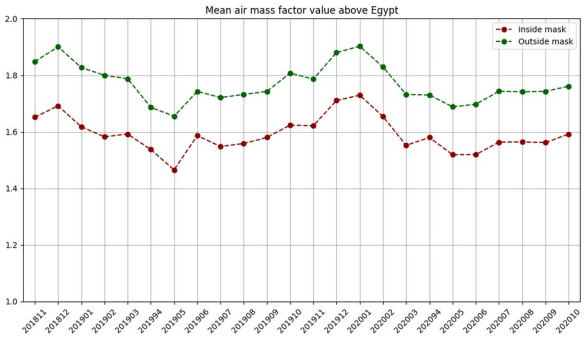

Figure F: Monthly TROPOMI AMF above Egypt inside and outside mask.

It should be noted that within the two types of cells, the spatial variations are of the same order as the temporal variations: standard deviations are about 0.13 outside mask cells and 0.18 inside mask cells. Finally, the AMF is rarely below 1.4 and rarely above 2.0. For the majority of months in the studied period, the effect of taking into account AMF is to decrease the monthly emission estimate. Except for the period 2019/12-2020/03, this happens for all months, and leads to a decrease between 6 and 27% (average: 16.8%). For the months of December 2019 to March 2020, the use of AMF leads to an increase (of 3.1%, 19.3%, 6.9% and 14.0% respectively). For these months, for which the estimates were very low, the correction decreased emissions inside our mask more than the background emissions. The figure below (Figure G) provides a comparison between previous estimates (with SCD) and new estimates (with VCD).

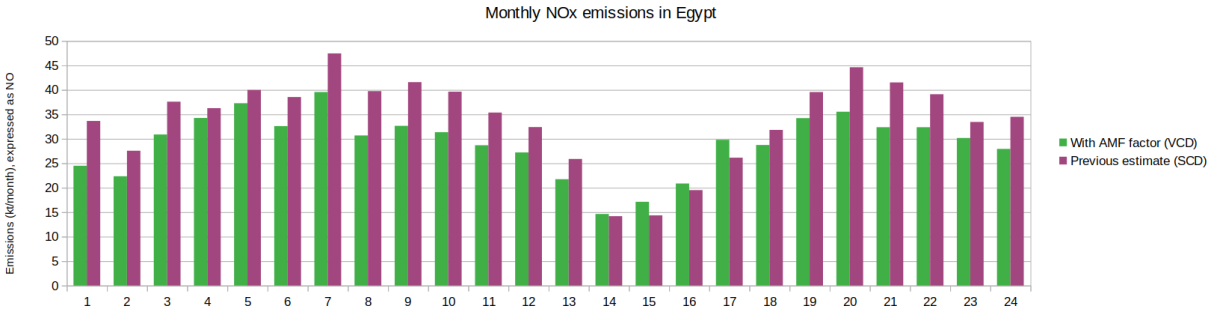

Figure G: Correction of Figure 9 after correction with AMF factor.

Accounting for the AMF does not therefore explain the temporal variations observed, even if it reduces their amplitude.

Emissions in winter 2019/2020 are very low compared to other months. Section 4.6 states that this decrease is due to the OH concentration of CAMS, which is significantly lower than in previous years. If we calculate the emissions for 2020-12/2021-01, or 2021-12/2022-01, we obtain monthly emissions of 38.8 kt/h and 32.7 kt/h respectively, which are similar to values found for 2018-12/2019-01 (35.8 kt/h) than to those for 2019-12/2020-01 (21.3 kt/h). It can therefore be assumed that the inferred emissions for winter 2019/2020 are not realistic. Nevertheless, winters remain the period with lowest emissions, while summer emissions generally shows a maximum around 50 kt/h.

Finally, we were deeply interested in the comment regarding the correspondence of a pronounced seasonal behavior where large power plants are. This can be assessed. In the revised manuscript, we will compare the cities of Aswan and Asyut. Both have similar features in terms of human presence : their respective populations were 315 khab and 467 khab in 2021, while their human densities were 1.6 khab/km² and 3.0 khab/km². However, their industrial features largely differ. There are no large fossil fuel fired power plants in Aswan, where most of the electricity is produced by the hydroelectric dam, whereas Asyut counts three oil and gas power plants of various capacities (90, 650 and 1500 MW) in its urban area. Both cities have a cement plant, but the one in Asyut has a larger production capacity (5.7 Mt/yr in Asyut, 0.8 Mt/yr in Aswan). The maps below shows the location of those large emitters for the two cities and the corresponding domains (Figure H).

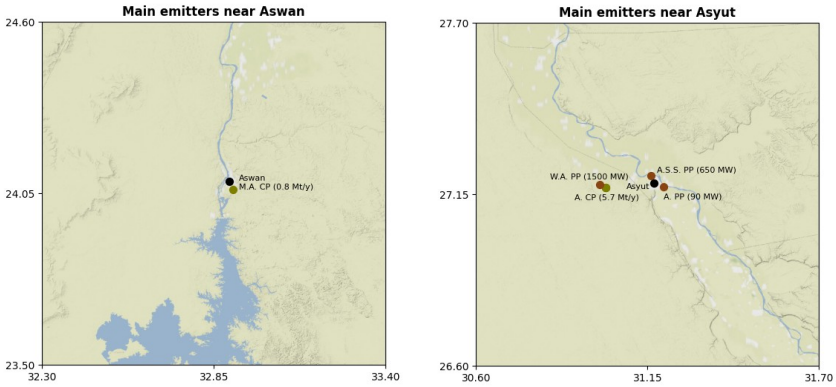

Figure H: Domains for Aswan and Asyut and location of city centres and large industrial facilities.

The method can be applied to those domains the same way it has been applied to the entire country. For both domains, power plant emissions and emissions from the city centre overlap and it is difficult to distinguish those emitters as point sources with our resolution. According to the seasonnal behaviour we highlighted and our assumption of a dominance of electricity production in the source of NO_x emissions throughout the country, the emissions of the Asyut-centered domain should display a seasonnal behaviour more pronounced than the rest of the country. Conversely, the Aswan-centered domain should have much smaller emissions, with a different (or less pronounced) seasonnality. The following figure (Figure I) presents NO_x emissions for both domains:

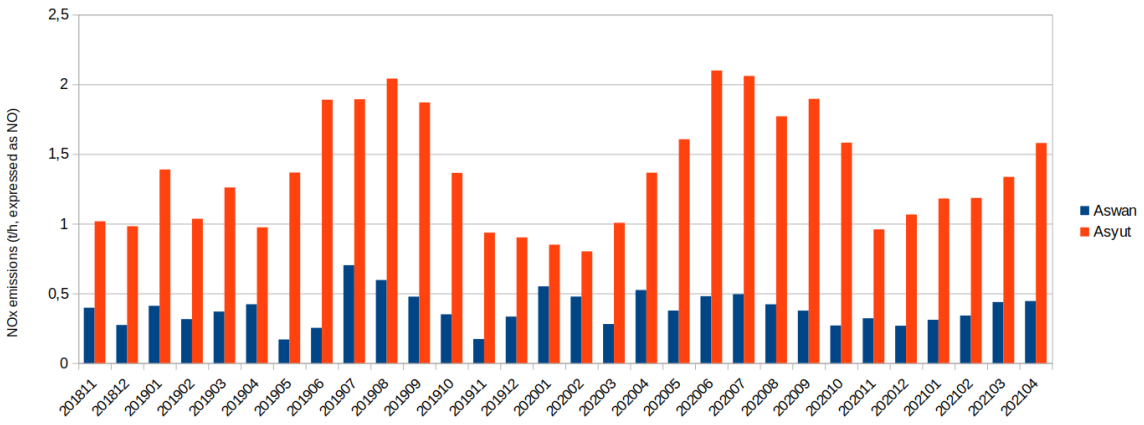

Figure I: NO_x emissions for Aswan and Asyut.

A seasonal cycle appears for Asyut, with a minimum in the winter months and a maximum around summertime (June/July/August/September). This cycle seems slightly shifted from the one observed for the entire country, for which May have emissions as large as summer months. We also note that the decrease in emissions for the winter of 2019/2020 is less marked than for the emissions of the whole country and consistent with the previous and following winter. All this suggests that there is indeed a decline in winter emissions, but that the winter of 2019/2020 is poorly represented at the national level. The seasonality of the emissions is more pronounced for the Asyut area than for the country as a whole. If we consider winter 2018/2019 as representative of winter emissions (i.e. excluding the results of the winter 2019/2020), then the ratio between the maximum of the cycle (May-June-July) and its minimum (December-January) is around 1.3 for Egypt but 2.2 for Asyut. The case of Aswan is different. Emissions are significantly lower there than in Asyut. It is difficult to characterise an annual cycle because the signal-to-noise ratio is higher than in the previous case but the results do not seem to indicate low emissions in winter and high emissions in summer. This seems to confirm the fact that power plants play a major role in the NO_x emission cycle of the country.

- Section 4.7: Regarding uncerainties, as noted above, I doubt that NO2+OH is really the only relevant NOx sink in the area. Furthermore, the impact of ignoring the AMF in Equation (3) should be assessed. The uncertainty of only 1 m/s for the wind components seems optimistic since the precise altitude at which the wind is interpolated is arbitrary, and the Coburn et al. study concerns the U.S. which is likely better characterized in the CAMS model.

→ We will increase the wind uncertainty from 1 m/s to 3 m/s in the revised manuscript. However, as the transport term is not significant in the total emissions calculation, this increase will be of small impact on total uncertainty.

- l. 239: Burkholer → Burkholder

→ Corrected in the revised manuscript.

- l. 378: "towards in the northeast and southeast quadrants": unclear. Do you mean towards the northeast in summer and southeast in winter?

→ This sentence means that the zonal component of the wind is positive most of the time. We acknowledge that the formulation can be misleading and we will change it in the revised manuscript, refering explicitely to this zonal component.

- l. 666: I could not find Huijnen et al. 2016, please provide URL

→ Added: https://pure.mpg.de/rest/items/item_2441827/component/file_2441834/content

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References:

Ehhalt et al., 1992	doi:10.1029/91JD03081
Jaeglé et al., 1998	doi:10.1029/97GL03591
Bertram et al., 2007	doi:10.1126/science.1134548
Jaeglé et al., 2005	doi:10.1039/B502128F
Müller and Stavrakou, 2005	doi:10.5194/acp-5-1157-2005
Lin, 2012	doi:10.5194/acp-12-2881-2012
Albrecht et al., 2016	doi:10.1175/BAMS-D-14-00193.1
Hoesly et al., 2018	doi:10.5194/gmd-11-369-2018
Granier et al., 2019	https://hal.archives-ouvertes.fr/hal-02322431
Beirle et al., 2019	doi:10.1126/sciadv.aax9800
Valin et al., 2013	doi:10.1002/grl.50267
Burkholder et al., 2020	http://hdl.handle.net/2014/49199
Guenther et al., 2006	doi:10.5194/acp-6-3181-2006
Romer Present et al., 2020	doi:10.5194/acp-20-267-2020
Fischer et al., 2014	doi:10.5194/acp-14-2679-2014