Effects of daily meteorology on the interpretation of space-based remote sensing of NO_2

Response to Anonymous Referee #2

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We thank the reviewer for their positive response and very careful reading of the manuscript. The reviewer's comments will be shown in red, our response in blue, and changes made to the paper are shown in black block quotes. Unless otherwise indicated, page and line numbers correspond to the original paper. Figures, tables, or equations referenced as "Rn" are numbered within this response; if these are used in the changes to the paper, they will be replaced with the proper number in the final paper. Figures, tables, and equations numbered normally refer to the numbers in the original discussion paper.

While I see the nice qualitative discussion of the effects as a strong point of this manuscript, the quantitative results are much less convincing in my opinion. The reason for my skepticism is the large change in emissions and lifetimes the authors find when changing their a priori spatial or temporal resolution in combination with the large uncertainties given in the tables. Most of the results are in agreement within uncertainties when changing from monthly to daily a priori in spite of the large changes seen. To me this indicates that the time period used for averaging is too short to really separate the effect of a priori changes from noise in the data, and as the authors acknowledge, all previous studies used much longer averaging times. Please comment on the magnitude of uncertainties and the significance of differences seen.

We do understand the reviewer's skepticism, given the magnitude of the uncertainties. In the original paper, we had already included statistical tests of the significance of these differences using t-tests (P14, L18–23) which showed that, in most cases, the differences due to the choice of a priori profiles are statistically significant at the 95% confidence level, except for lifetimes derived using the monthly 108 km and daily 12 km profiles. Reviewer #1 raised the possibility of autocorrelations in the data, which would indicate that the uncertainties were underestimated. While we do not believe that temporal autocorrelations are significant in this method, some spatial autocorrelation seems to be unaccounted for by the EMG fit. In response to Reviewer #1, we have acknowledged this possibility in the revised paper, but note that the differences are systematic, and therefore are not expected to be reduced by additional averaging, while the uncertainties will be. Added at P.13, L.19:

> "We note that a Durbin-Watson test indicates some spatial autocorrelation remains, and so the uncertainty may be underestimated and the t-tests may be

> incorrectly identifying the differences as significant in this case (Chatterjee and Hadi, 2012). Even if this is true, with a longer averaging period such as those in Beirle et al. (2011), Valin et al. (2013), and Lu et al. (2015), we would expect the random uncertainties to reduce while the systematic difference from the choice of a priori profile remains. Therefore, the choice of a priori profiles does have an important effect on derived emissions and lifetimes."

In the discussion of the results both in the text and in the abstract, I'm confused by statements such as "Comparing an optimized retrieval to a more standard one, we find that NOx emissions estimated from space-based remote sensing can increase by 100% when daily variations in plume location and shape are accounted for in the retrieval." If I'm not misinterpreting Table 4, the change in emissions when moving from monthly to daily a priori is closer to 45% and actually is a decrease, not an increase of estimated emissions. This also makes more sense considering the qualitative discussion given in the first part of the paper. The factor of 2 increase is relative to a low spatial resolution a priori which is also interesting but not the focus of the study and also not what is suggested by the formulations in the text. I think these statements need to be rephrased.

Thank you for clearly explaining what you found confusing about this statement; we agree now that its present form is unclear. We have reworded it in both the abstract and conclusions to clearly indicate that there are different effects of spatial and temporal resolution on the emissions. While the effect of temporal resolution is indeed the main focus of this paper, it is important to separate the effects of spatial and temporal resolution since no application of the EMG fitting method has yet used a retrieval with high spatial resolution profiles (Beirle et al., 2011; Valin et al., 2013; Lu et al., 2015; Liu et al., 2016).

In the abstract, P.2 L.8–9 changed to:

> "Additionally, we show that NO_x emissions estimated from space-based remote sensing using daily, high spatial resolution a priori profiles are $\sim$ 100% greater compared to a retrieval using spatially coarse a priori profiles, and 20–45% less compared to a retrieval using monthly averaged high spatial resolution profiles."

And in the conclusion, P.15 L.3–5 changed to:

> "When the methods of Lu et al. (2015) are applied to these prototype retrievals, significant changes in derived NO_x emissions are found, increasing by as much as 100% for Atlanta compared to emissions derived from a retrieval using coarse a priori profiles."

The discussion of standard mathematical methods for fitting a function to the decay curve is a bit out of place in such an article, in particular as the method used for emission estimation is not the topic of the paper. I would suggest to shorten this part and to remove the discussion of Matlab internals which are of little interest to the reader.

Both other reviewers also requested that references to Matlab functions be made more general, which we have done. The technical considerations have also been moved to the supplement (P.8, L.9–29).

The approach taken to averaging the model data in time (Equation 5) appears overly complex and not transparent. The obvious way to treat this problem is simple interpolation in time to the OMI overpass.

Effectively this approach is an interpolation, as an interpolation can be recast as a weighed average. If we are interpolating to a point (x_q, y_q) between (x_1, y_1) and (x_2, y_2):

$$\begin{aligned} y_q &= y_1 + m(x_q - x_1) \\ &= y_1 + \left(\frac{y_2 - y_1}{x_2 - x_1}\right)(x_q - x_1) \\ &= \frac{x_q - x_1}{x_2 - x_1} y_2 + \left(1 - \frac{x_q - x_1}{x_2 - x_1}\right) y_1 \\ &\equiv w y_2 + (1 - w) y_1 \text{ where } w = \frac{x_q - x_1}{x_2 - x_1} \end{aligned}$$

This approach assumes that the average OMI overpass time varies linearly with longitude. We have added a paragraph after Eq. (5) and a section to the supplement explaining this reasoning; essentially, as shown in Fig. R1, there is the possibility that an area (such as Atlanta) may be covered by the west edge of an earlier swath or the east edge of a later swath, so these weights are an *ad hoc* attempt to choose profiles that approximate the average overpass time.

After Eq. (5):

> "The weighting scheme in Eq. (5) was chosen over simply using the model output for 1400 local standard time for each latitude to create smooth transitions between adjoining time zones. This attempts to account for the day-to-day variability in OMI overpass tracks as well as the fact that pixels on the edge of a swath can be observed in two consecutive overpasses at different local times. More detail is given in the supplement."

And in the supplement:

> "When computing the monthly average profiles, it is necessary to use profiles that represent OMI's overpass time, typically quoted as 13:30 to 13:45 local standard time (e.g. McLinden et al. 2014; Levelt et al. 2006). To average the profiles output from WRF-Chem, weights were calculated that fulfilled two requirements:
>
> 1. The weights should be 1 at OMI overpass time and 0 when more than 1 hour away from overpass time.
> 2. The transition between profiles from different hours should be smooth.
>
> For #1, we assume that the average overpass time is 1330 local standard time. We compute local standard time as:
>
> $$t_{\text{apriori, local}} = \frac{l}{15} + t_{\text{apriori, utc}} \tag{R1}$$

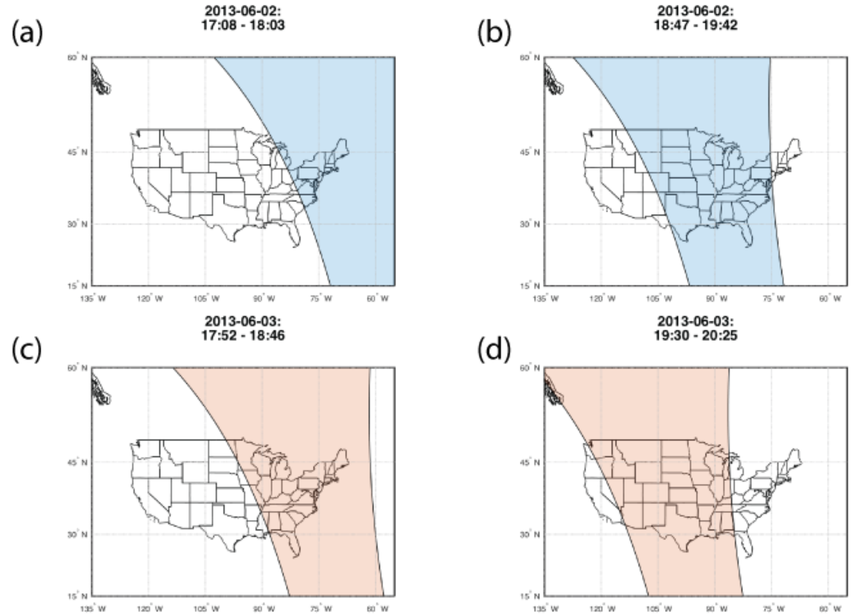

Figure R1: Swaths covering the east coast of the US for 2 June 2013 (a,b) and 3 June 2013 (c,d). The times given are the start and end times of the daytime half of the orbit in UTC. As shown, on different days, the time of the OMI swath that covers Atlanta can vary by up to an hour.

where $t_{\text{apriori, local}}$ is the local standard time in hours past midnight, $t_{\text{apriori, utc}}$ the UTC time in hours past midnight, and l the longitude (west is negative). To meet the second requirement, this is a continuous function, rather than a step function (where each 15° longitudinal segment/time zone has a single local time). Areas further west in a time zone are more likely to be observed on the east edge of a later OMI swath, and vice versa for areas further east. This weighting includes some influence from later profiles to account for this.

The weights from Eq. (5) are derived from:

$$w = 1 - |t_{\text{overpass}} - t_{\text{apriori, local}}| = 1 - \left|13.5 - \frac{l}{15} - h\right| \qquad \text{(R2)}$$

where t_{overpass} is the assumed overpass time for OMI and $h \equiv t_{\text{apriori, utc}}$. If $w < 0$, w is set to 0. This gives us the desired form where the weights smoothly vary in time.

"

In the course of revisiting this weighting method, we redid the key runs using an assumed OMI overpass time of 1330 LST (which is more consistent with previous papers, e.g. McLinden et al. 2014, compared to 1400 LST used originally). The difference is very small and does not affect the conclusions of the paper, but we have updated the appropriate figures and parts of the text.

On a more general note I think that the paper would benefit from a short discussion of the impact model errors could have on the results. It is obvious that in theory, using daily a priori data is better than using monthly averages as the process of NO2 retrieval is not linear. However, in real data this might not necessarily be true. At the high spatial resolution of the model used here, even a small uncertainty in wind direction, emission height or emission source location can move the NO2 plume into different model grid cells, potentially leading to poor matching of plume position in measurement and model and thus wrong air mass factors and NO2 columns. I find this an interesting topic in particular in view of future instruments having improved spatial resolution.

Agreed, model uncertainty is an important (and deep) topic highly relevant to this paper, so we have added a section to the discussion with our thoughts on this topic. There has been some interesting work comparing WRF-Chem simulations to measurements both directly (Tie et al., 2007; Zhang et al., 2009) and in terms of spatial variability (Follette-Cook et al., 2015) which suggest WRF does capture daily wind fields and NO_2 distribution reasonably accurately. We would also expect the overall uncertainty to change source, but not necessarily magnitude between monthly and daily profiles, as monthly profiles effectively have the true day-to-day variability as a source of uncertainty that daily profiles do not; but in exchange, the random error in the monthly profiles is reduced through averaging. We have added a new section at the beginning of the discussion that covers this point:

> "WRF-Chem has generally been found to reproduce wind fields, especially above 2 m s^{-1} (Tie et al., 2007; Zhang et al., 2009), and spatial variability of trace gases (Follette-Cook et al., 2015) well. Nevertheless, a natural concern when modeling daily NO_2 profiles for satellite retrievals is the accuracy of the plume location. We, however, note that the transition from monthly average to daily profiles does not necessarily result in increased model uncertainty, but rather a change in the type of uncertainty.
>
> When using monthly average profiles, the uncertainty in the modeled NO_2 concentrations compared to the true mean will be reduced (assuming at least some component of the error is random in nature), but the true day-to-day variability not captured by the monthly average effectively becomes a new error term. In contrast, when using daily profiles, the random model error is not reduced, but the day-to-day variability is also not averaged out. Ideally, the error in a set of daily profiles will manifest as deviation from the true set of profiles for that day, rather than the monthly profiles' smaller deviation from a mean set of profiles that itself may not represent any single day.
>
> An important step in managing the uncertainty in the daily profiles is to constrain the modeled meteorology with observations or reanalysis datasets. By default, meteorology in WRF is constrained via initial and boundary conditions only. With larger domains and longer runs, further constraints using four-dimensional data assimilation (FDDA, Liu et al. 2006) and/or objective analysis (Follette-Cook et al., 2015; Wang et al., 2014; Yegorova et al., 2011), possibly combined with periodic model reinitialization (Otte, 2008) are strongly recommended."

Page 7, line 5: Cloudy AMF is smaller, not lager [larger] for boundary layer NO2 profiles

Thank you for bringing this up; we double checked this statement by comparing clear sky and cloudy AMFs from both the NASA standard product and our own, and found that in both products, the regression slope of cloudy vs. clear sky AMFs is > 1, but that there is sufficient scatter that it is hard to say definitively that one is larger or smaller. Thus, we've reworded this point to simply identify that clear and cloudy AMFs can be significantly *different*, but not that one is necessarily greater than the other. P.7, L.5 changed to:

> "Day-to-day variations in cloud fraction also lead to large changes in AMF because the presence of clouds changes both the scattering weights (due to high assumed reflectivity of clouds and smaller effective surface pressure compared to ground) while also obscuring the NO_2 profile below the cloud."

Page [15] line 6: I do not understand the sentence Further work is needed to understand the impact of this change on top down constraints of NOx emissions, given the recent work showing that bottom up estimates are high by 50%. I think the relevance is obvious if you believe your own results all previous estimates based on this technique and using monthly high resolution a priori profiles give too high emission estimates. Whether or not the emission inventory is off is another topic (which could of course also impact on your AMF values and thus emission estimates) and should not be mixed here.

Indeed, any previous work using monthly high resolution profiles would be biased high, which would fit in nicely with Travis et al. (2016) indicating that the bottom up inventory is too high. However, the previous application of the EMG method to US emissions used a retrieval with coarse a priori profiles (Lu et al. 2015); our results indicate that emissions found using a retrieval with spatially coarse set of a priori profiles will be lower than when using daily, fine spatial resolution profiles. Hence the need for more work–Travis et al. (2016) found that the bottom up inventory should be reduced by $\sim 50\%$ using GEOS-Chem as a transfer standard between aircraft and satellite measurements, while we find that, with better a priori, the bottom up inventory is about 25% high.

We have edited this sentence to make this clearer, it now reads:

> "Emissions derived using the fine daily a priori are within 25% of the bottom up number from the NEI inventory, a smaller reduction than that suggested by Travis et al. (2016). Future work will aim to resolve this difference."

Finally, each of the following technical issues has been addressed

Equation 2: $w_s(p)$ is later written as $w(p)$

Equation 4: Please add how cloud radiance fraction was computed

Added just before Eq.(4):

> "The cloud radiance fraction is taken from the SP v2 data product (Bucsela et al., 2013)."

Page 6: Add reference for MOZART model
Added Emmons et al. (2010).

Page 14, line 17: something missing here?
The "and" after "*a*" was unnecessary.

Conclusions, line 28: Please add again that you count days even if only a single pixel shows a change larger than the noise (which I personally find a strange way of counting)

> "Up to 59% of days with valid observations exhibit changes in VCDs $> 1 \times 10^{15}$ molec. cm^{-2} in at least one pixel."

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