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

Response to Anonymous Referee #3

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We thank the reviewer for their helpful comments, especially the points of clarification in the introduction. 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.

One limitation is the focus on only one area (Atlanta, GA) and one season (Summer). As pointed out by another reviewer, focusing on such a short period leads to a limited dataset, from which it is difficult to obtain estimates of emissions and lifetimes that are significantly different. Since the differences in the estimated emissions are at least a factor of 2, it should be feasible to achieve statistically significant differences by analysing a longer period. This would strengthen the paper considerably.

We are glad to see that the reviewer is interested in the application of daily profiles when constraining emissions and lifetime. It is definitely expected that extending the analysis to a longer time period would allow a more quantitative analysis of emissions and lifetime, but the goal of this paper was primarily to show that there is an important difference in the AMFs and therefore VCDs retrieved using daily vs. monthly profiles, and that these differences do impact the emissions and lifetime inferred from the product. While the uncertainties in the emissions and lifetimes are large, we conclude that the differences due to the a priori are systematic and describe our reasoning why. We hope to extend the time period and geographic area studied to get quantitative estimates of emissions and lifetimes across a number of US cities, and that others will keep these results in mind when considering top-down emissions and lifetime constraints.

The systematic nature of these changes is discussed on P.13 L.20–P.14–L.2. In response to another reviewer's comment, this paragraph has been extended starting from P.13 L.31:

> "In the EMG fit, this manifests as a too short lifetime. As the emissions are inversely proportional to lifetime (Eq. 11), emissions derived using the monthly 12 km a priori profiles will be too great. Therefore, when using a retrieval with a priori profile at fine spatial resolution, daily temporal resolution of the a priori

profiles is necessary to prevent underestimating the lifetime. Further, the spatial resolution of the a priori profiles has a large impact on the magnitude of the derived emissions. To reduce the systematic biases in emissions and lifetime from the choice of a priori profile, it is necessary to simulate these profiles at fine spatial and daily temporal resolution."

We have also added the following sentence on P.4, L.21 to make clear that our goal is not yet to quantitatively constrain emissions and lifetime, but to demonstrate that the systematic error due to the use of monthly average profiles is important and should be addressed:

"...meteorological variables. Our point is not to derive exact answers for the size and frequency of the effects of daily profiles, but rather to illustrate that these effects are large enough that their role should be assessed in any future analysis that does attempt to interpret space-based remote sensing of NO_x. We show that the variability..."

Then there is a serious error in the theoretical framework for BEHR AMFs. According to Eq. (2) and the text in section 2.2, the cloudy AMF is calculated only between the cloud pressure level and the tropopause. In principle this can be done, but then the retrieval needs to account for a so-called ghost column [Burrows et al., 1999]. A ghost column correction however, is not being applied here. The better alternative is to calculate both the clear-sky and cloudy-sky AMFs by integrating the NO2 profile from the surface pressure to the tropopause. This formulation ensures that the AMF value returns a tropospheric column that is representative for all NO2 in the troposphere, and not just for the above-cloud fraction (in the limiting case of cloud fraction 1.0). The authors should revisit their integration limits for Eq. (2), probably also for their current BEHR-product.

The reviewer is correct that this formulation of the AMF yields the visible NO_2 column, however this is the calculation given in the theoretical basis document for the OMI retrievals (Boersma et al., 2002). P. 20 of the TBD indicates after Eq. 2-15 that "z is the altitude of the lower boundary (ground or **cloud top**)," where z is the lower integration bound in the numerator of the AMF calculation. The publicly available BEHR retrieval includes a multiplicative factor which allows users to add the estimated ghost fraction in, if desired. This allows the user to choose how they want to use the product, e.g. (1) as the observed, visible column, (2) for cloud slicing approaches (e.g. Belmonte Rivas et al. 2015; Choi et al. 2014 for NO_2, or Ziemke et al. 2009, 2001 for O_3), or (3) with the ghost column included to get a full column.

We acknowledge the various ways of handling below-cloud NO_2 on P.2 L.25:

"Finally, the tropospheric slant column density (SCD) must be converted to a vertical column density (VCD) by use of an air mass factor (AMF) and Eq. (1). Depending on the specific algorithm, NO_2 obscured by clouds may be ignored (producing a visible-only tropospheric NO_2 column, e.g. Boersma et al. 2002), corrected by use of an assumed ghost column (e.g. Burrows et al. 1999; Koelemeijer and Stammes 1999), or corrected via the AMF (e.g. Martin et al. 2002). In all cases, the AMF must account for..."

We also added information about the available ghost column factor in the BEHR product after Eq. (4):

> "Calculating clear and cloudy AMFs and using the weighted average to compute the final AMF is consistent with the OMI algorithm theoretical basis document (Boersma et al., 2002) and yields only the visible NO_2 column as the final product; the visible column is the value provided in the BEHRColumnAmountNO2Trop field. A scaling factor is provided in the BEHR product for users who wish to include the ghost column. This factor, G, is computed as:
>
> $$G = \frac{V_{\text{surf}}}{(1 - f_{\text{geo}})V_{\text{surf}} + f_{\text{geo}}V_{\text{cld}}} = \frac{\int_{p_{\text{surf}}}^{p_{\text{tp}}} g(p)\, dp}{(1 - f_{\text{geo}}) \int_{p_{\text{surf}}}^{p_{\text{tp}}} g(p)\, dp + f_{\text{geo}} \int_{p_{\text{cld}}}^{p_{\text{tp}}} g(p)\, dp} \quad \text{(R1)}$$
>
> where V_{surf} and V_{cld} are the modeled vertical column densities above the ground surface and cloud, respectively, and which are obtained by integrating the a priori profile above the surface or cloud pressure. f_{geo} is the geometric cloud fraction included in the NASA standard product, which is the OMI O_2-O_2 cloud product (Acarreta et al., 2004). This factor is stored in the BEHRGhostFraction field of the BEHR product. Multiplying the VCDs stored in BEHRColumnAmountNO2Trop by these values will provide the estimated total (visible + ghost) column.
>
> The results obtained in this work use the visible columns only. The ghost column is not added in for any of the following results."

We have clarified one point in Eq. (2) and (3). In (3), the lower bound of integration is always the surface, but in Eq. (2), the lower bound is the cloud or surface pressure, for cloudy and clear sky AMFs respectively:

> "p_0 represents the surface pressure (clear sky AMF) or cloud pressure (cloudy AMF) of the satellite pixel, and p_{tp} the tropopause pressure...p_{surf} in Eq. (R3) is the terrain surface pressure.
>
> $$\text{AMF} = \int_{p_0}^{p_{\text{tp}}} w(p)S(p)\, dp \quad \text{(R2)}$$
>
> where
>
> $$S(p) = \frac{1}{\int_{p_{\text{surf}}}^{p_{\text{tp}}} g(p)\, dp} g(p) \quad \text{(R3)}$$
>
> "

This error is also the reason for the incorrect statement on page 7, under point 3: ... a cloudy pixel will have a much greater AMF than a clear one. The statement would be correct only if the word AMF would be replaced by above-cloud AMF. And, in line with the above

criticism, an above-cloud AMF can only return an above-cloud NO2 columns, which cannot be the purpose of a state-of-science retrieval.

As discussed above, the choice to retrieve the visible column only matches the OMI theoretical basis document (Boersma et al., 2002), and both visible only and full column retrievals have uses. Reviewer #2 also pointed out that AMFs for cloudy pixels are not always larger than clear-sky AMFs, which we confirmed by comparing cloudy and clear AMFs from our product and the NASA standard product. Consequently, this sentence now indicates that changing cloud fraction can lead to a very different AMF, and discusses the reasons for that.

> "Setting cloud fractions to 0 ensures that the AMF for every pixel is calculated with the full a priori profile, rather than just the above cloud part. 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."

There are difficulties with the interpretation of uncertainties in the tropospheric column retrieval. On page 10, the choice for 1.0x1015 molec. cm-2 as a typical number for the uncertainty in the tropospheric NO2 column is rather arbitrary. Detailed error analyses (e.g. Boersma et al. [2004]) have pointed out that the uncertainty in the tropospheric column is highly variable because of AMF (a multiplicative factor indeed) uncertainties, which depend strongly on variable clouds, surface albedo, and NO2 profile shapes. Moreover, the estimate used by the authors is rather optimistic. More realistic uncer- tainties are on the order of 1.0x1015 molec. cm-2 +25% (of the individual column value).

The value of 1.0×10^{15} molec. cm^{-2} was given in Bucsela et al. (2013) as the global average mean clear sky uncertainty based on their error analysis. Since the uncertainty in the AMF is a multiplicative factor, this error may be low for urban signals. We had chosen initially to use this uncertainty, however, because we are introducing new choices for a priori profiles, which may alter the magnitude of each specific element of the uncertainty, and so having a simple, previously determined number to compare to would simplify this for the reader.

To address the reviewer's concern, we have added a new column to what was Table 2 that uses an uncertainty that is the quadrature sum of uncertainty from the slant column fitting, stratospheric subtraction, and AMF, which is $\sqrt{(0.7 \times 10^{15})^2 + (0.2 \times 10^{15})^2 + (20\%)^2}$. The uncertainty in the AMF and stratospheric separation come from Bucsela et al. (2013), which describes the error analysis for the current version of the NASA SP retrieval, and the slant column fitting from Boersma et al. (2011), cited by Bucsela et al. (2013). We add them in quadrature following Boersma et al. (2004) and Bucsela et al. (2013). This leads to the following changes in text:

P.10, L.23–29 changed to:

> "Table 2 describes how frequently significant changes in the retrieved VCD occur for pixels within 50 km of Atlanta, Birmingham, and Montgomery. Changes are considered significant by two different criteria. First, we consider the global mean clear-sky uncertainty from Bucsela et al. (2013). As we are modifying the a priori profiles, and thus potentially the uncertainty associated with the choice

of profiles, this gives us a fixed value to compare against. Second, we use the quadrature sum of uncertainties from spectral fitting (0.7×10^{15} molec. cm^{-2}, Boersma et al. 2007, 2011), stratospheric separation (0.2×10^{15} molec. cm^{-2}, Bucsela et al. 2013), and AMF calculation (20%, Bucsela et al. 2013), assuming that these are independent and so can be added in quadrature (Boersma et al., 2004). We consider the fraction of days with at least one pixel exhibiting a significant change in VCD (rather than the fraction of pixels) because the main NO_2 plume may only fall within a small number of pixels. Up to 54% of days exhibit changes in the VCDs greater than 1×10^{15} molec. cm^{-2}, and up to 43% exhibit changes greater than the quadrature sum of uncertainties. This indicates that when considering individual daily measurements, a considerable fraction of days with any valid pixels would have biases in the retrieved VCDs above the uncertainty due to the temporal resolution of the a priori NO_2 profiles. "

P.11, L.3–5 changed to:

"When considering changes to be significant if they exceed 1×10^{15} molec. cm^{-2}, Montgomery has the least frequent significant changes because it has the smallest VCDs, so a change to the AMF needs to be rather large to produce a significant change in the VCD by this metric, since the AMF is a multiplicative factor. When considering the quadrature sum of errors as the significance criterion, Montgomery and Atlanta both demonstrate significant changes $\sim$ 20% of the time."

And the update to Table 2:

	Percent of days with ΔVCD $> 1 \times 10^{15}$ molec. cm^{-2}	Percent of days with ΔVCD $> [\sum_i \sigma_i]^{1/2}$	Min. change (molec. cm^{-2})	Max. change (molec. cm^{-2})
Atlanta	39%	23%	-2.4×10^{15}	$+2.5 \times 10^{15}$
Birmingham	54%	43%	-3.8×10^{15}	$+3.9 \times 10^{15}$
Montgomery	27%	20%	-2.2×10^{15}	$+1.9 \times 10^{15}$

Table 1: Statistics on the frequency and magnitude of changes in the retrieved VCDs using a daily vs. monthly average profile for pixels with centers within 50 km of Atlanta, GA, USA (84.39° W, 33.775° N), Birmingham, AL, USA (86.80° W, 33.52° N) and Montgomery, AL, USA (86.30° W, 32.37° N). The "percent of days" values are calculated as the number of days with at least one pixel in that subset with a change greater than the given uncertainty divided by the number of days with at least one pixel unobscured by clouds or the row anomaly. The uncertainty represented by $[\sum_i \sigma_i]^{1/2}$ is the quadrature sum of uncertainties from spectral fitting (0.7×10^{15} molec. cm^{-2}, Boersma et al. 2007, 2011), stratospheric separation (0.2×10^{15} molec. cm^{-2}, Bucsela et al. 2013), and AMF calculation (20%, Bucsela et al. 2013).

The specific corrections below have all been addressed. We thank the reviewer for their careful reading, and especially for the additional citations for custom retrievals, as we hope

that others will find this list a useful reference for those interested in custom satellite retrievals.

Abstract, line 6: this paper does not address variations of NO2 in power plant plumes, so this should be removed. The paper is about NO2 variations in urban plumes.
Removed.

Introduction, lines 18 and 30: a reference to the work by Vinken et al., ACP, 2014 on ship emissions estimates using improved-resolution a priori profiles would be appropriate here.
Added Vinken et al. (2014), thank you for the suggestion.

P3, lines 7-8: earlier studies by McLinden et al. [2012] showed that the oil sand signals was detectable also for retrievals using coarse-gridded a priori profiles. The statement should be nuanced in that the spatial signatures are more realistically resolved with higher resolution a priori profiles.
This sentence has been changed to:

> "McLinden et al. (2014) showed that using 15 km resolution profiles increased the NO_2 signal of the Canadian oil sands by $\sim 100\%$ compared to the DOMINO and NASA SP products, which they state corrects a low bias in the retrieved column amounts."

P3, L13: also include here a reference to the DOMINO retrievals using 0.5 x 0.67 profiles from GEOS-Chem over Europe from Vinken et al. [2014]. These retrievals also use diurnal profiles.
Added:

> "Conversely, the DOMINOv2 (Boersma et al., 2011), POMINO (Lin et al., 2015), and DOMINO2_GC (Vinken et al., 2014) retrievals simulate daily profiles at 3° lon × 2° lat (DOMINO) and 0.667° lon × 0.5° lat (POMINO and DOMINO2_GC), respectively, which is insufficient to capture the full spatial variability of NO_2 plumes, but does capture large scale variations in meteorology."

P3, L17: the NASA retrieval is usually indicated as the Standard Product v2 or SP v2. OMNO2 (actually OMNO2A v1) refers to the software for DOAS spectral fitting.
Corrected, for all instances as well as this one.

P4, L30: 13 x 24 km2
Corrected.

P4, L32: 'theoretical daily global coverage is a strange term. Coverage was nearly global every day until the row anomaly, and after that, coverage is more or less global every 2 days.
Rephrased to:

"It has a continuous data record since 1 Oct 2004, with global daily coverage for the first $\sim$ 3 years of operation. Since 25 June 2007..."

P5, L6: please provide the name and appropriate version of the NO2 SCD product used. I think it is OMNO2A v1. This product has recently been evaluated in Marchenko et al. [2015] and van Geffen et al. [2015]. It would be appropriate to cite those papers here.

Thank you for pointing us to the proper identification. This section now reads:

"Briefly, the BEHR retrieval is based on the NASA Standard Product v2 (SP v2) retrieval (Bucsela et al., 2013). The total slant column densities (SCDs) are from OMNO2A v1.2.3 (Boersma et al., 2002; Bucsela et al., 2006, 2013), and have been recently evaluated by van Geffen et al. (2015) and Marchenko et al. (2015). The stratospheric subtraction and destriping used is that of the NASA SP v2 retrieval. The tropospheric AMF is then recalculated..."

P5, L17: apart from albedo, please also provide details on the cloud information (effective cloud fraction, pressure) used in the BEHR-approach.

We have added information about the cloud pressure and cloud radiance fraction used in the retrieval. For cloud pressure, the sentence "The cloud pressure is that provided in the NASA SP v2 product, and is retrieved using the OMI O_2-O_2 cloud algorithm (Acarreta et al., 2004; Sneep et al., 2008; Bucsela et al., 2013)," was added before Eq. 2. For cloud radiance fraction, the sentence "The cloud radiance fraction is taken from the SP v2 data product (Bucsela et al., 2013)," was added before Eq. 4.

P5, L27: which version of the WRF-Chem model is used?

Version 3.5.1. The first sentence of sect. 2.3 now reads:

"Modeled NO_2 a priori profiles are simulated using the WRF-Chem model v3.5.1 (Grell et al., 2005)."

P6, section 2.3: please provide some more details on the WRF-Chem model such as what is the NOx emission total over the US in the period of interest, and on the meteorological and chemical boundary conditions used. It is unclear how realistic WRF-Chem simulations of NO2 profiles are. Have these been validated against surface and aircraft measurements of NO2?

The chemical mechanism used has been validated against surface data from the SOAS campaign. However, the purpose of this paper is to illustrate that using daily profiles at high spatial resolution can and does alter the retrieved NO_2 columns. We feel that it is important to demonstrate that the temporal resolution of the profiles is important first, and because the monthly and daily 12 km *a priori* profiles are taken from the same model run (and the 108 km profiles from a run using the same inputs at different resolution), any error in the model is in all three sets of *a priori* profiles. In this way, our goal is to show that daily, high spatial resolution profiles are important in general.

We have added details of the meteorology, boundary conditions, and emissions to the paragraph on P.5 L.26–P.6 L.4. It now reads:

"Modeled NO_2 a priori profiles are simulated using the WRF-Chem model v3.5.1 (Grell et al., 2005). The domain is 81 (east-west) by 73 (north-south) grid cells centered on 84.35° W, 34.15° N on a Lambert Conformal map projection (approximate edges of the domain are 89.5° W–79.2° W and 30.3° N–38° N). Meteorological initial and boundary conditions are driven by the North American Regional Reanalysis (NARR) dataset. Anthropogenic emissions are taken from the National Emissions Inventory 2011 (NEI11) and scaled to 88.9% to account for 2011–2013 NO_x reductions (EPA, 2016); total emissions of NO for the domain are approximately 3.1×10^6 kg NO day^{-1}. The MEGAN model (Guenther et al., 2006) is used to determine biogenic emissions. Chemical initial and boundary conditions for the domain are obtained from the MOZART chemical model (Emmons et al., 2010). The RACM2 (Goliff et al., 2013) and MADE-SORGAM schemes are used to simulate gas-phase and aerosol chemistry respectively; the RACM2 scheme is customized to reflect recent advancements in understanding of alkyl nitrate chemistry using Browne et al. (2014) and Schwantes et al. (2015) as a basis. Lightning NO_x emissions were inactive."

P7, L19: please clarify what the vertical range is of the 'first five layers of WRF-Chem.

Five model layers is approximately 500 m high, which is the same height used in Lu et al. (2015). The sentence now reads:

"The surface wind direction and speed are calculated as the average of the first five layers ($\sim$ 500 m) of the 9 WRF 12 km grid cells closest to Atlanta at 1400 local standard time for each day."

P8, L6: is it really necessary or relevant to refer to a Matlab function, when explaining how you do the fit? If it is, please provide some more information on why you chose this particular fit approach over other alternatives. As a non-Matlab user, the sentence does not mean much to me. Later on the same for fmincon.

Other reviewers had similar comments; in the main paper, we've replaced references to Matlab functions with the mathematical algorithm behind it, which is an interior-point algorithm.

P8, 12-17: this part is very technical and should be moved to an appendix or supplement.

Again, other reviewers had similar comments. We have moved Table 1 and P8, L12–29 to the supplement. Section 2.5 now ends with "Technical details of the EMG fitting and uncertainty calculation are given in the supplement."

P10, L13: that lightning is not included in this WRF-Chem set-up should not be mentioned only here, but already in the model description section 2.3.

We have added the following sentence on P. 6, L. 4: "Lightning NO_x emissions were inactive," (see end of change for previous comment on WRF-Chem details).

P12, L18: sigma_x represents the width of the Gaussian plume, but also the spatial smearing of the signal caused by the satellite pixel extent, and the fact that cities are covered by different satellite footprints from day to day.

Originally we had focused on its representation of the plume width because that is the most physically intuitive quantity, but we have extended the first sentence of P12, L18 to read:

> "σ_x is the Gaussian smoothing length scale, representing both the width of the upwind Gaussian plume and smoothing of the NO_2 signal due to the physical extent of the source, the averaging of NO_2 within one OMI pixel, and daily variability in the overpass track (Beirle et al., 2011)."

P13, L14-19: this whole section presumably discusses Table 4, but that is not obvious from the text. I'm confused by the statement that the choice of a priori leads to statistically different emissions for all five cases, whereas Table 4 shows emission values that all overlap within the quoted uncertainty estimates.

When comparing two measured values, t-tests designed for either comparing replicate measurements or paired tests must be used (Harris, 2010). This paragraph is discussing results from using t-tests for comparing replicate measurements, i.e. two sample t-tests (Harris 2010, sect. 4-3, case 2). We chose this method because the emissions and lifetime estimates are the result of averaging VCDs over 3 months, so the fitting parameters are effectively means. Additionally, unlike the paired t-tests, this method takes into account the uncertainty in each value. Using these tests, any pair of emissions derived using different a priori for the same city and wind division are statistically different, even though the confidence intervals overlap.

To clarify which t-tests were used, we have modified P.13 L.14 to read ("2-sample" added):

> "We also use 2-sample t-tests at the 95% confidence level (Harris, 2010) to determine if differences in emissions and lifetimes given in Table 4 are significantly different..."

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