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

Response to Anonymous Referee #1

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October 20, 2016

We thank the reviewer for their positive response and very careful reading of both the main article and the supplement. Below we respond to the individual comments. 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.

Figure 1 is a very nice example of how the AMF is influenced by monthly and daily a prioris. From the manuscript, the authors suggest that this is based on the prior vertical information of NO2 from the model and the scattering weighting function. However, in the introduction there was no reference to why the AMFs are smaller/larger in the scenarios in Figure 1. I think adding a short explanation of what causes the AMFs to be different would be useful. Also, as far as I can tell, the manuscript doesnt quantify which of the two processes in Eqn 2 & 3 influencing the AMFs are most important. Which is it?

The paragraph on P4, L7–18 discusses each of the scenarios in Fig. 1. We have added a sentence near the beginning of this paragraph to remind the reader of the physical cause for the dependence on the NO_2 profile illustrated in each panel, so the paragraph now begins as:

> "In this paper we explore how day-to-day changes in the a priori NO_2 profiles affect satellite retrievals of urban NO_2. Several scenarios are illustrated in Fig. 1. In each case the change in the AMF results because, over low albedo surfaces, a UV/visible satellite spectrometer is less sensitive to near surface trace gases, necessitating a smaller AMF to account for the reduced sensitivity. In Fig. 1a..."

Regarding which process is more important, and taking this to mean the calculation of scattering weights vs. the a priori profile, there is no one answer, as this varies with the amount of NO_2 present. We have expanded the part at P.1 L.25–27 to explain this and noted that previous work has found a priori profiles to be very significant near urban areas:

"...the AMF must account for the varying sensitivity of the satellite to NO_2 at different altitudes, and therefore a priori knowledge of that sensitivity and the vertical profile of NO_2 is required. Over low-reflectivity surfaces, light scattered in the atmosphere is the primary source of radiance at the detector. The probability of back-scattered light penetrating to a given altitude is greater for higher altitudes; thus there is greater interaction with, and therefore greater sensitivity to, NO_2 at higher altitudes (Richter and Wagner, 2011; Hudson et al., 1995). Because of this, the correct AMF is smaller in locations influenced by surface NO_x sources. The relative contribution of errors in the calculated sensitivity and in the a priori profiles of NO_2 to error in the final VCD varies between polluted and clean pixels (Boersma et al., 2004). Previous work (e.g. Russell et al. 2011) has sought to reduce errors in both, and highlighted the importance of accurate a priori profiles in urban areas."

In section 2.5 the EMG is discussed in detail (and in the supplementary information), but often with reference to how Matlab functions are used to calculate the required equations. For someone who has not used Matlab before, this might be difficult to follow (e.g. fmincon, nansum etc). Could the authors just discuss the mathematical and statistical methods used and leave discussion of Matlab functions in the supplementary material? I also think that there should be discussion on what a, x0, μx, σx and B are in the text of section 2.5, instead of just referring to Table 1.

This comment is well taken. Similar comments were made by other reviewers. We have moved the technical elements (P.8 L.9–29) to the supplement and included a description of the fitting parameters here instead, after Eq. (9):

"Eq. (9) is minimized using an interior-point algorithm, finding the values of a, x_0, μ_x, σ_x, and B that best fit the line densities. The values of a, x_0, μ_x, σ_x, and B have physical significance and so their optimum values yield information about the NO_x emission and chemistry occurring within the plume (Beirle et al., 2011; de Foy et al., 2014; Lu et al., 2015). Specifically:

- a describes the total amount of NO_2 in the plume (referred to as the burden)
- x_0 is the distance the plume travels in one lifetime, τ. It relates to τ by $x_0 = \tau \times w$, where w is wind speed.
- u_x describes the effective center of the emission source. In the supplement to Beirle et al. (2011), it is represented by X which is the point at which exponential decay of the NO_2 plume begins.
- σ_x is the standard deviation of the Gaussian component of the EMG function. Lu et al. (2015) terms this a "smoothing length scale," which describes smoothing of the data due to the spatial resolution and overlap of OMI pixels (Boersma et al., 2011). It can also be thought of as capturing effects of both the spatial extent of emissions and the turbulent wind field.
- B is the background line density.

”

In section 2.3, there is discussion on weighting schemes (i.e. Eqn 5). Unfortunately, I do not understand how and why this is used. If you are trying to calculate the model monthly mean relative to the OMI sampling, could you not just subsample the model to the individual satellite overpass times (e.g. within 1 hour of 14.00LT)? This text (Page 6, Lines 5-10) needs to be improved to make the motivation for Eqn 5 clearer.

We have added a paragraph after Eq. (5) explaining our reasoning for these weights:

> “The weighting scheme in Eq. (5) was chosen over simply using the model output for 1400 local standard time for each longitude 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:
>
> $$t_{\text{apriori, local}} = \frac{l}{15} + t_{\text{apriori, utc}} \tag{R1}$$
>
> where t_{local} is the local standard time in hours past midnight, t_{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:
>
> $$w = 1 - |t_{\text{overpass}} - t_{\text{apriori, local}}| = 1 - \left|13.5 - \frac{l}{15} - h\right| \tag{R2}$$

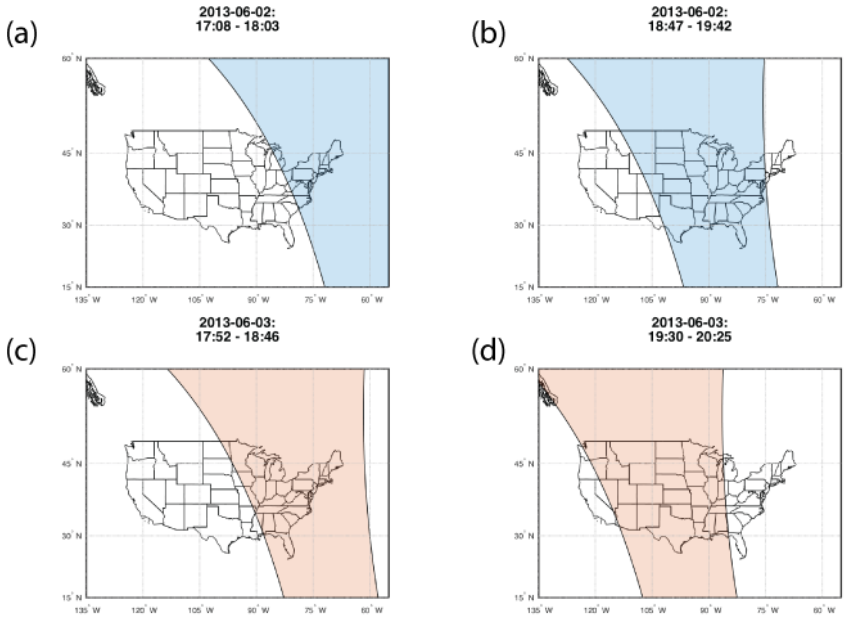

Figure R1: Swaths covering the east coast of the US for 2 June 2013 (a,b) and 3 June 2013 (c,d). The times given are the start and end times of the daytime half of the orbit in UTC. As shown, on different days, the time of the OMI swath that covers Atlanta can vary by up to an hour.

where t_{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 abstract and introduction any reference to “Atlanta, GA should be “Atlanta, GA, USA as not everyone will know Atlanta is in the USA.

We have added “USA” to any instance where the city is given as “Atlanta, GA,” and did the same for “Birmingham, AL” and “Montgomery, AL.” Cases of the city name alone were left as such (i.e. just “Atlanta” not “Atlanta, GA”).

On Page 2, Lines 26-27, “Because the satellite is less sensitive to NO2 near the surface, the AMF should be smaller in locations influenced by surface NOx sources this is discussed in detail throughout the manuscript, but it would be good to add a short sentence here explaining why this is the case.

We have added two sentences before Eq. 1 explaining the physical basis for the lower near-surface sensitivity:

“Over low-reflectivity surfaces, light scattered in the atmosphere is the primary source of radiance at the detector. The probability of back-scattered light penetrating to a given altitude is greater for higher altitudes; thus there is greater

interaction with, and therefore greater sensitivity to, NO_2 at higher altitudes (Richter and Wagner, 2011; Hudson et al., 1995). Because of this, the correct AMF is smaller in locations influenced by surface NO_x sources."

Page 3, Line 11: BEHR needs to be defined here, not later in the section.
Definition of BEHR added.

"The current generation Berkeley High Resolution (BEHR) (Russell et al., 2011, 2012) and OMI-EC (McLinden et al., 2014) retrievals simulate monthly average NO_2 profiles at 12 and 15 km, respectively."

Page 3, Lines 13-14: "2° x 3° and 0.5° x 0.667°. Im assuming this is lons then lats? These are actually lat x lons. We have reversed the order and clearly defined it:

"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..."

For the statement "Valin et al. (2013) showed that the concentration of NO2 downwind of a city increases significantly with wind speed. on Page 3 Lines 24-25, can a range be provided to which this statement is true.
We have added specific numbers based on Fig. 4 in Valin et al. 2013:

"Valin et al. (2013) showed that the concentration of NO_2 downwind of a city increases significantly with wind speed, observing that NO_2 100–200 km downwind from Riyadh, Saudi Arabia was approximately 130–250% greater for wind speeds between 6.4–8.3 m s^{-1} than wind speeds < 1.9 m s^{-1}."

Page 3, Lines 27-29: The authors should explain why OMI is less sensitive at lower altitudes or provide a reference which explains this.
In response to the fifth comment, we added this before Eq. 1. We have included a phrase directing the reader to that explanation here:

"As discussed before Eq. (1), UV/visible satellite observations of NO_2 are less sensitive to NO_2 at low altitudes..."

In the Introduction, we are informed that this study focuses on Atlanta. The reasoning for this is explained later on in the manuscript, but needs to be mentioned here as well to make it clear why this region is the focus of the study.
We have added a sentence to page 4, around lines 21–22 that explains this reasoning:

"...to demonstrate the impact of day-to-day variations in the modeled NO_2 profiles on the calculated AMFs surrounding a major urban area such as Atlanta, GA, USA. Atlanta provides an example of a strong NO_x area source relatively isolated from other sources, with straightforward response of the day-to-day a priori profiles to meteorological variables."

Please reword Lines 30 (P4) 2 (P5) to make the text clearer. i.e. These have been classified as the row anomaly and as of 5 July 2011 affect one-third of the pixels http://projects.knmi.nl/omi/research/product/rowanomaly-background.php), reducing coverage from global daily to global every two days.

We have reworded this 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, anomalous radiances have been observed in several of the pixel rows. These have been classified as the "row anomaly"..."

In the title of section 2.2 please expand out what BEHR represents. Also, P5, L5, BEHR doesnt need to be expanded again as it is done so in the Introduction.

These changes have been made.

In Eqn 2,3 please specifically state what p represents. Also, how is ptp defined (e.g. dynamical, chemical tropopause)?

We have added this to lines 16–21 on p. 5 (p is the vertical coordinate pressure, the tropopause is defined as a static value of 200 hPa). This paragraph has been expanded to address other reviewers' comments as well:

"...p represents the vertical coordinate as pressure. $w(p)$ represents scattering weights derived from the NASA SP v2 look up table. $g(p)$ represents the mixing ratio NO_2 a priori profile taken from WRF-Chem, simulated at 12 km resolution in the published BEHR product. p_0 represents the surface pressure (clear sky AMF) or cloud pressure (cloudy AMF) of the satellite pixel, and p_{tp} the tropopause pressure. 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). A static tropopause pressure of 200 hPa is used. p_{surf} in Eq. (3) is the terrain surface pressure. The integration is carried out using the scheme described in Ziemke et al. (2001) which allows integration of mixing ratio over pressure."

Please provide a reference or explanation why "an assumed cloud albedo of 0.8" is used, on P5 , L 21.

Citations for Stammes et al. (2008) and Bucsela et al. (2013) have been added, this paragraph has been expanded for other comments as well:

"The scattering weights, $w(p)$, depend on the viewing geometry, surface albedo, and terrain pressure altitude. The BEHR algorithm uses the $0.05° \times 0.05°$ combined MODIS MCD43C3 black-sky albedo product and a surface pressure derived from the Global Land One-km Base Elevation project database (`http://www.ngdc.noaa.gov/mgg/topo/globe.html`; Hastings and Dunbar 1999) with a 7.4 km scale height as inputs to the clear sky scattering weights. Cloudy scattering weights treat the cloud pressure as the surface pressure and use an assumed cloud albedo of 0.8 (Stammes et al., 2008; Bucsela et al., 2013). The final AMF is computed as the cloud radiance fraction (f_{rad}) weighted average of the clear and cloudy AMFs (Eq. 4). The cloud radiance fraction is taken from the SP v2 data product (Bucsela et al., 2013)."

P5 Line 29: What does NE1 11 stand for/represent?
This stands for National Emissions Inventory 2011; the abbreviation has been expanded.

The manuscript should not reference papers in prep such as Zare, (in prep) on P6 Line 4.
We have removed this citation; this now reads:

"...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."

P5 L5: Please provide justification for using a spin up period of just 5 days.
This is similar to Browne et al. (2014). Qualitatively, after about 24 hours, the initial conditions of NO_2 appear to have little effect on the modeled concentrations significantly due to the short lifetime of NO_2. The initial concentrations are removed chemically, and the modeled concentrations are driven by the emissions and meteorology. Other species affecting NO_x chemistry, such as ozone and peroxy acetylnitrates (PANs) appear to require about 3 days before the initial concentration no longer has a large impact on the modeled concentrations. We have added a reference to Browne et al. (2014).

"The model is run from 27 May to 30 August, 2013. Similar to Browne et al. (2014), the five day period 27–31 May is treated as a spin up period, thus we use 1 June to 30 August as our study time period."

P6, L20: Please rephrase " 'pseudo-retrieval' that is much simplified compared to a full operational NO2 retrieval" and outline how it is much simpler. Also on L21, please state some examples of co-founding variables.
Both the simplification and confounding variables are addressed in the numbered points below this paragraph (P7, L15–24). We have removed the sentence mentioned in this comment, as we see how it can be confusing, and believe that by doing so, we direct the reader to these points further on in the paragraph where they are addressed in more detail. The paragraph beginning on P.6, L.20 now begins with:

"Two retrievals are used to study the effects of incorporating daily a priori profiles in the BEHR algorithm. The first is what we term a "pseudo-retrieval." To create this retrieval, an 11 × 19 (across × along track) subset of pixels..."

Section 2.5: Firstly, I cannot find an example of where EMG is expanded (i.e. what does it stand for)? Secondly it should be expanded in the introduction where EMG is first mentioned. It should also be written in full for the title of this section.

We have expanded it in both the section title and introduction.

P7 L20: An explanation on why the WRF winds are transformed to earth-relative from grid-relative would be much appreciated.

We have added a concise explanation to P.7, L.16–20:

"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. WRF wind fields are given relative to the model grid; however, the x and y coordinates of the grid do not correspond directly to longitude and latitude. Therefore, the wind fields must be transformed from grid-relative to earth-relative..."

Eqn 8: Please define specifically what F(x. . ...) represents. Also stating what "erfc" stands for would be useful as well. Im assuming it is "error function"?

We have expanded the paragraph between Eq. 8 and 9 to address both of these concerns, as well as better connect these two equations for the reader:

"where erfc is the error function complement, i.e. $\mathrm{erfc}(x) = 1 - \mathrm{erf}(x)$. $F(x|a, x_0, \mu_x, \sigma_x, B)$ serves as an analytical function that can be fitted to the observed line densities. We find the values of a, x_0, μ_x, σ_x, and B that minimize the sum of squared residuals between $F(x|a, x_0, \mu_x, \sigma_x, B)$ and the line densities, $\mathrm{NO_2}(x)$:"

P9, L8-9: As stated above, the reasoning for choosing Atlanta needs to be outlined earlier on in the manuscript.

This was addressed at the previous comment.

In Figure 2b, it would be useful to add a scale for the wind speed or add some text to the caption stating what the min, mean and max winds in the domain are for that period.

We have added these statistics to the caption, which now reads:

"Average conditions for June 2013. (a) The red box indicates the part of the SE US being considered. (b) Surface wind directions from the WRF model; average wind speed is 5.0 m $\mathrm{s^{-1}}$ (min 1.7 m $\mathrm{s^{-1}}$, max 12.7 m $\mathrm{s^{-1}}$). (c) WRF-Chem tropospheric $\mathrm{NO_2}$ columns. (d) AMFs for the pseudo-retrieval calculated using the average monthly $\mathrm{NO_2}$ a priori. The direction of the colorbar is reversed in (d), as small AMFs correspond to high modeled VCDs. In all panels, the star (★) indicates the position of Atlanta. Longitude and latitude are marked on the x- and y- axes, respectively."

P9-10, L30-1: Please expand on "All pixels show a positive change." Is this correct. Should there not be negative changes somewhere in the domain?

We were also surprised initially that all pixels showed a positive change. The remainder of that paragraph (P10, L1–5 in the original paper) explains the cause of this positive everywhere change. We have added a connective clause that makes it clearer that the rest of the paragraph addresses this point:

> "All pixels show a positive change. This occurs because 77% of the daily profiles have less NO_2 than the corresponding monthly average profile..."

Please expand VCDs in the section 3.2 title

We have expanded the definition of VCDs in the section 3.2 title.

P11, L9-11, the authors state that the uncertainty value of 1015 molecules per cm2 can be reduced by a factor of the SQRT(n). However, this assumes that all errors in this uncertainty value are random. Surely, some of the error will be systematic or smoothing errors? Therefore, the authors show take this into account or explain why it can be done like this.

Another reviewer also made this point. Boersma et al. (2004) notes that an explicit separation of the random and systematic components is very difficult, so the errors are usually treated as entirely random. Nevertheless, while we retain the 1×10^{15} molec. cm^{-2} criteria for the discussion because of its simplicity, we also added a second criterion that separates error due to spectral fitting, stratospheric separation, and AMF calculation. We assume that the spectral fitting and stratospheric subtraction errors are random, and half of the AMF error. The expanded paragraph is:

> "Implementing the daily profiles also changes the average VCDs, in addition to the day-to-day changes in VCDs discussed above. Figure 4b shows the changes in VCDs averaged over the period studied. The largest decrease around Atlanta is to the northeast, along the direction that the monthly average model results placed the NO_2 plume, but clear decreases can also be seen to the northwest and southwest. In these directions, a systematic decrease of up to 8% (4×10^{14} molec. cm^{-2}) is observed. Although this change is small, it is expected to be systematic. Statistically, a pixel's a priori profile is more likely to have less surface NO_2 when different wind directions are no longer averaged in, thus decreases in the VCD when using a daily a priori profile are more common.
>
> Greater relative changes are observed around the smaller cities of Birmingham (down to -12.5%, 5×10^{-14} molec. cm^{-2}) and Montgomery (down to -13%, 4×10^{-14} molec. cm^{-2}). This appears to be due primarily because the areas of emissions are smaller which makes shifts in wind direction have a greater average relative effect on the plume shape.
>
> We also compare this average change to the measurement uncertainty. The uncertainty due to random errors in the retrieval should reduce as the square root of the number of observations, but delineating random and systematic errors

in the retrieval is challenging (Boersma et al., 2004). The most optimistic approach assumes that the global average uncertainty of 1×10^{15} molec. cm^{-2} (Bucsela et al., 2013) can be treated entirely as random error, and can be reduced by $\sqrt{40}$ for the number of observations (not impacted by clouds or the row anomaly), to a lower bound of $\sim 1.6 \times 10^{14}$ molec. cm^{-2}. Most of the changes near the three cities exceed this lower limit. More realistically, the spectral fitting and stratospheric uncertainty may be considered largely random, but only part of the error in the AMF calculation is random, due to spatial or temporal autocorrelation in the models or ancillary products (Boersma et al., 2004). For simplicity, we assume that the spectral fitting and stratospheric subtraction errors are entirely random, while only half of the error in the AMF is random. This reduces the error from $\sqrt{(0.7 \times 10^{15})^2 + (0.2 \times 10^{15})^2 + (20\%)^2}$ to $\sqrt{(0.11 \times 10^{15})^2 + (0.03 \times 10^{15})^2 + (11.6\%)^2}$. Only the largest changes near Birmingham and Montgomery exceed this threshold. This more conservative estimate suggests that the changes in averages are primarily important for smaller or very geographically concentrated cities, where wind direction can have a large effect. Nevertheless, larger cities may exhibit important changes as well."

The two sentences on P11, L7-9 "The main decrease around Atlanta is to the northeast, along the direction that the monthly average model results placed the NO2 plume. A systematic decrease of 5-10% to the northeast of Atlanta is observed; this is the plume direction in the monthly average profiles." need to be reworded as discussing "northeast" twice is repetitive.

This has been done; we've also better indicated that decreases are seen in other directions as well (see first paragraph of revision for previous comment).

Just double checking on P11, L17, this should definitely "southeast"?

Yes, the wind blows to the southeast most frequently. The average wind direction for June 2013 is to the northeast, but this is because the wind direction usually falls within a 180° arc centered on the northeast.

P12, L12. Should be "a x0" and not "an x0".

According to the Chicago Manual of Style, "an" before the "ex" sound is acceptable, and we prefer that. (http://www.chicagomanualofstyle.org/qanda/data/faq/topics/Usage/faq0068.html)

On P13, L14-19, the t-test is discussed to determine if "differences in emissions and lifetimes are significantly different among the results derived from using the three different a priori profiles...." The t-test assumes that data within the sample population are independent. However, I imagine there will be lots of temporal autocorrelations in the samples. Do the authors account for this and if not, why?

We agree with the reviewer that it is likely that there would be some degree of autocorrelation in the temporal evolution of VCDs over the study period, and so in the day-by-day line densities as well. However, the emissions and lifetime are derived from fitting parameters that fit the dependence of the temporal average line densities as a function of space. Of

course, there will naturally be spatial autocorrelations as well, given the physical processes governing the evolution of the NO_2 plume. The EMG function used to fit these line densities should account for these physical processes (Beirle et al., 2011), and according to Chatterjee and Hadi (2012), autocorrelation often appears when the fitting model does not include dependence on a key variable. Since a Durbin-Watson test indicates that there is still unaccounted for spatial autocorrelation, we now acknowledge in the paper that the t-tests may underestimate the uncertainty and focus on the fact that these changes will be systematic:

> "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 3 are significantly different among the results derived from using the three different a priori profile sets for a given city and wind speed bin (i.e. we compare the three values of emissions derived using different a priori profiles for Atlanta and wind speeds ≥ 3 m s^{-1}). This found that, for emissions, the choice of a priori leads to statistically different emissions for all five cases. For the derived lifetimes, in all cases the monthly 108 km and daily 12 km a priori are statistically indistinguishable, but the monthly 12 km a priori is statistically different. 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."

On P14, Lines 3-15, comparisons to the NEI 11 emissions are discussed. From the text, the NE1 11 emissions are overestimated by 50%. If the NEI 11 emissions took this into account, which results (e.g. monthly or daily apriori) which have best agreement with them. Also, on L7, the authors state that the "daily 12 km a priori are within 5- 24%". Are they lower or higher than the NEI 11 emissions or both? The authors have stated that the "coarse monthly a priori" are lower by 43-62%.

We have reorganized this paragraph to accommodate this comment; the sentence indicating which emissions agree with the current and 50% reduced NEI emissions has been brought closer to the statement about the uncertainty in the NEI inventory. That emissions derived from daily 12 km profiles are both greater and less than the NEI inventory has been explicitly stated:

> "We also compare the derived emissions rates to the emissions in a 12 km WRF-Chem model driven by the NEI 11 emission inventory with NO_x emissions scaled to 88.9% of the 2011 values to account for the decrease between 2011 and 2013 (EPA, 2016). WRF-Chem emissions are calculated as the sum of all grid cells within a 50 km radius of the city. 50 km was chosen as the line densities were integrated for $\sim$ 50 km to either side perpendicular to the wind direction. The coarse monthly a priori are 43–61% lower than the NEI-driven emissions, while

emissions derived using daily 12 km a priori are within 5–24% (both greater and less than the NEI emissions). Recent work (e.g. Travis et al. 2016 and references within) suggests that the NEI inventory is overestimated by $\sim$ 50% using both satellite and in situ observations. Emissions derived using daily 12 km show the best agreement to the current NEI inventory, and emissions derived using monthly 108 km a priori profile agree with the NEI inventory reduced by 50%. Therefore, we cannot say which a priori profiles provide the best measurement of emissions by comparing to NEI...."

Supplement

P1, L16: Why would we expect the mean and median differences to be 0?

Since the upper troposphere is generally removed from NO_x sources, we expect NO_2 concentrations there to be fairly stable. Deviations from the average should mostly be due to larger scale motions of air masses, also separated from direct influence from NO_x sources, so the deviations should be essentially random and normal. Only at the surface with direct influence of NO_x sources do we expect a non-normal distribution of NO_2 concentrations, and so a non-zero mean difference in the average of AMF calculated from the monthly average profile vs. the daily profiles.

P1, L20-22: "This, combined with the greater scattering weights at these altitudes, explains why the effect on the AMF is as large as it is, although day-to-day changes in the boundary layer still dominate the effect using daily profiles has on the AMFs,". The "," at the end of the sentence should be a full stop. Secondly, can the impact of the scattering weights at this altitude be quantified, say in comparison to the impact of the apriori on the AMFs?

Thank you again for careful proofreading. As far as quantifying the effect of the scattering weights, the response of the upper tropospheric scattering weights to perturbations in the inputs to the radiative transfer model that computes them could be calculated, but is not particularly relevant to this part of the work. More important here is that scattering weights in the UT are 2–8x higher than near the surface, so small perturbations in the NO_2 profile have larger effects on the AMF than the same perturbations would have near the surface. We have included this factor of 2–8x:

"Day-to-day changes in the free tropospheric a priori profile are smaller in magnitude than those in the boundary layer, but usually occur over a much greater vertical extent. Further, the scattering weights are greater at these altitudes ($\sim$2–8 times those near the surface), amplifying the effect of small changes in the profile shape at these altitudes. This explains why the effect on the AMF is as large as it is, although day-to-day changes in the boundary layer still dominate the effect using daily profiles has on the AMFs."

P1, L30: Is this period of 91 days long enough for the averaging to have no net impact?

Yes, as evidenced by the fact that the average difference between the hybrid and full profiles is 0 (Fig. S3).

P2, L 9: Should be "Atlanta, nevertheless" or "Atlanta. Nevertheless".
Corrected, thank you.

P4, L4-5: Should be these "]" brackets and not ")"?
No, in a range, infinity should be accompanied by a round parenthesis since no finite range can truly include infinity.

P5, L20: "as the errors contributing to it should be random in nature". Why is this the case? No systematic errors?
This is similar to a previous comment on the uncertainty in VCDs; while there is definitely some systematic component, the errors are usually treated as random in practice (Boersma et al., 2004). Removing the factor of $\sqrt{n}$ does not alter our conclusions however, because we calculated n conservatively, so we will use the flat 25% from Lu et al. (2015). Table 4 and this section have both been updated accordingly (P.5 L.19–24 of supplement removed).
Updated Table 4:

		Atlanta			Birmingham		
	Wind speed bin	Monthly 108 km	Monthly 12 km	Daily 12 km	Monthly 108 km	Monthly 12 km	Daily 12 km
E (Mg NO_x h^{-1})	WRF-Chem NEI		**13.74**			**10.49**	
	≥ 3.0	$6. \pm 4$	$16. \pm 9$	$11. \pm 7$	$4. \pm 2$	$10. \pm 6$	$8. \pm 5$
	≥ 4.0	$6. \pm 3$	17 ± 11	$11. \pm 6$	$4. \pm 2$	$13. \pm 7$	$9. \pm 5$
	≥ 5.0	-	-	-	$6. \pm 3$	$15. \pm 9$	$11. \pm 6$
τ (h)	≥ 3.0	1.6 ± 0.7	1.3 ± 0.5	1.7 ± 0.7	2.5 ± 1.0	1.8 ± 0.7	2.6 ± 1.0
	≥ 4.0	1.8 ± 0.7	1.2 ± 0.5	1.8 ± 0.7	2.1 ± 0.9	1.5 ± 0.6	2.2 ± 0.9
	≥ 5.0	-	-	-	1.8 ± 0.7	1.3 ± 0.5	1.8 ± 0.7

Table R1: Values of the emission rates (E) and effective lifetime (τ) obtained when the separation between slow and fast winds is set at 3, 4, and 5 m s^{-1}. For comparison, the total NO_x emission for all 12 km WRF-Chem grid cells within 50 km of each city is given. These emissions are derived from NEI 11 and scaled to 88.9% to account for 2011–2013 reductions. Uncertainties calculated as described in the supplement.

P6, L15: "). nfit"?
The period should go inside the parenthesis (as it is) when the full sentence in parenthetical.

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