Responses to Reviewer Comments on acp-2015-237

Emissions of nitrogen oxides from US urban areas: estimation from Ozone Monitoring Instrument retrievals for 2005–2014

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We thank all referees for their positive and constructive suggestions and comments, which have helped us improve the manuscript. All comments have been carefully dealt with, as detailed below, and we have highlighted all the changes in the revised manuscript (green for referee #1 and yellow for referee #2).

To Referee #1

This paper presents an interesting and valuable assessment NO_x emissions based on OMI observations. The strategy for using wind observations building on Valin et al.'s initial insight is nicely explained and results in emissions estimate that are significantly different from prior estimates. The paper should be published.

The one larger concern I have is with the absence of a discussion of possible remaining systematic errors. The authors have identified a substantial systematic error in prior emissions estimates (and trends) and explained the error arises by using OMI observations under all wind conditions. I wonder whether they think other significant systematic errors remain. Examples: Weekend vs. weekday emissions, a priori correlated with winds, chemical feedbacks . . .

<u>Response</u> Obtaining a sufficient number of samples is the major obstacle in this work to studying the weekend/weekday emission impact. Because all OMI measurements are divided on the basis of the wind speed, we have made our filter criteria as relaxed as possible and even combined all the valid data in three consecutive years to increase the amount of valid OMI data. If we further divided the OMI measurements by weekend/weekday, we would not have sufficient samples in the dataset, particularly for the weekend.

The potential systematic errors in wind field selected and the treatment of the wind field seem to be minor. First, the choice of wind field datasets was reported to be insensitive to the EMG results (Valin et al., 2013). This was further confirmed by de Foy et al. (2015) who tested wind fields of both the ERA-interim reanalysis (the one used in the current work) and the North American Regional Reanalysis (NARR) and obtained similar results in the EMG analysis. Second, although we choose fixed 12:00 LT as the time of the wind field, based on the work of Beirle et al. (2011), if the average wind over the last 6 hours is considered, the results only change slightly (~10%). Overall, Beirle et al. (2011) estimate the uncertainties due to the choice for the wind data as 30%, and we have taken that into account in the current analysis.

The chemical feedback of NO_x chemistry is somewhat related to the wind speed (i.e., slow or fast), because if there was no non-linearity of NO_x chemistry, there should be no significant differences between NO_2 trends derived from the all-wind and the weak wind situations. Please refer to our responses to the second and the third comments of Reviewer #2. In the revised manuscript, we have added a discussion of the possible relationship between the wind speed and the non-linear NO_x chemistry at the end of the last paragraph of Sect. 3.1.

Other minor issues: >On p. 14976, lines 10-15, the paper describes the procedure for filtering by wind speed and ensuring sufficient OMI observations. de Foy et al. (2014) does show that the EMG method generally gives the same answer for emission rate once the wind speed is greater than 3 m/s. Given this, why not take all winds >3 m/s rather than vary the cut off? Was this to be more consistent with Valin et al. (2013) where the number of OMI observations made it possible? Would always using 3 m/s change the results very much?

Response We will first briefly summarize the findings of de Foy et al. (2014) and Valin et al. (2013), and then give our consideration about the cut-off wind speed in this work. In de Foy et al. (2014), we evaluated the performance of the EMG method using simulated column densities over a point source with known emissions under three chemical lifetime cases (i.e., ∞, 12-h, and 1-h). Generally, the EMG method provides reliable emission estimates at wind speeds >3 m/s in all lifetime cases. However, the EMG-obtained emissions seem to be more accurate when wind speeds are higher, especially for the 1-h chemical lifetime case (see Table 2 of de Foy et al., 2014).

In Valin et al. (2013), although they inferred NO_x emissions in the city of Riyadh from the OMI measurements with wind speeds >6.4 m/s only, their NO_2 burden and lifetime estimates at wind speeds >6.4 m/s were same as those at wind speeds >5 m/s (see bottom panel of Figure 4 of Valin et al., 2013). It implies that, instead of 6.4 m/s, using the cut-off wind speed of 5 m/s in their analysis would infer the same NO_x emission estimates of Riyadh. However, their mass balance calculations further showed that the NO_2 lifetime increased with decrease of the wind speed, implying the inaccuracy of NO_x emission estimates when using wind speeds slower than 5 m/s.

Taking into account the results of both of these studies, we choose to use 5 m/s (if possible) as the cut-off wind speed in this work because it should provide us with the most accurate emission estimates. For urban areas that don't have enough (i.e., at least 30 in three consecutive years) valid OMI samples due to this wind speed criterion, we relaxed the cut-off to 4 or 3 m/s. In fact, we did examine the results of using a fixed cut-off wind speed of 3 m/s and the differences are minor. In the revised manuscript, we have added two sentences here (i.e., the first paragraph of Sect. 3.2) to make the expressions clearer.

>Is the "fitting interval" (p. 14978, line 21) x_0, sigma, or something else? If it is x_0, then what is the difference between the effective and dispersion lifetimes? Is it that the effective lifetime is obtained at high wind speeds and the dispersion lifetime at low wind speeds?

Response The "fitting interval" is neither x_0 nor sigma. It is the dimension of the fitting domain downwind from the urban center, and it is used to calculate $\tau_{dispersion}$, which is further used to explain the differences between $\tau_{residence}$ obtained at low wind speeds and $\tau_{effective}$ obtained at high wind speeds (de Foy et al., 2014, 2015). We have made changes in the revised manuscript to make this clear.

>The discussion of NO_x lifetimes on pp. 14977-14978 is secondary to the main discussion of the correlation between the various emissions and burden trends, so it seems out of place in the middle of a section on emissions. Maybe it should be moved to the end of that section, or given its own (albeit short) section?

Response We agree with the reviewer that the discussion of the NO_x lifetimes seems out of place in the middle of Sect. 3.2. In the revised manuscript, we have moved this part to the end of Sect. 3.2.

>p. 14975 – lines 12-15. when discussing trends from weak wind data, is it meant to isolate the trends in the NO_2 column/burdens specifically? This sentence could clarified as "we utilize OMI data under weak wind conditions to calculate the satellite-observed NO_2 columns, burdens, and trends in these quantities in this work." This would just make clear that weak wind data is not being used to calculate trends in OMI-derived emissions.

Response The manuscript has been changed as suggested.

>p. 14977 – lines 9-10. Consider rewording as "and trends in NO_2 columns obtained at slow winds may better reflect the real bottom-up NO_x emissions trends." This helps make clear that that column measurements at weak winds are being used as an indication of trends in surface emissions, rather than the strong-wind method of deriving emissions directly.

Response The manuscript has been changed as suggested.

>p. 14977 – lines 20-21. What exactly is meant by statistically significant in this context? Is it just that the columns are observable over background given the precision of the OMI measurements?

Response The term "statistically significant" here means that the uncertainty of the OMI burden estimated based on the method described in this work is less than 100% (including the impact of the precision of the OMI measurements).

To Referee #2

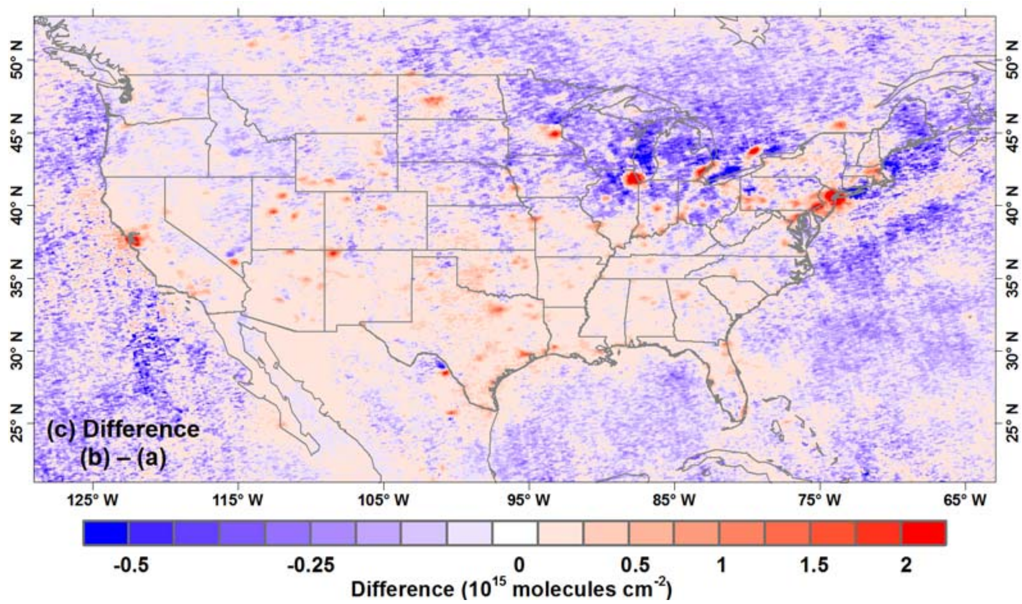
In their paper "Emissions of nitrogen oxides from US urban areas: estimation from Ozone Monitoring Instrument retrievals for 2005–2014", Lu et al. report on an analysis of OMI satellite observations of tropospheric NO_2 column amounts over the US yielding detailed emission estimates for 35 major urban areas. This study builds on earlier work by Beirle et al. and Valin et al. but extends on it by using low wind speed situations to better estimate the absolute NO_2 burden and applying the effective life time derived from high wind speed scenarios after rotation by wind direction. The results show very good correlation with the absolute emissions from bottom-up estimates and also with their temporal evolution. The paper is well written, reports on an interesting and thorough study of satellite derived emission estimates and fits well into the scope of ACP. I therefore recommend it for publication after minor revisions.

Comments

- *One of the interesting aspects of this paper is the comparison of NO_2 columns taken at different wind speeds. As expected, NO_2 columns are larger in urban areas at low wind speed which is relevant for emission estimates and interpretation of satellite maps. However, I'm surprised to see that in Figure*

1c, there is not the expected ring of low (blue) values around the hot-spots. On the contrary, NO_2 levels appear to be higher nearly everywhere at low wind speed with the exception of the Great Lakes area. Do you have any explanation for this?

Response The differences between OMI NO_2 maps of an urban area at the all-wind-speed condition and at the slow-wind-speed condition depend on the local average wind speed. The higher the average wind speed, the larger the differences. In the US, the annual average wind speeds are relatively high in the northeast region (i.e., the Great Lakes area the reviewer mentioned), and therefore, the low NO_2 regions around the hotspots are more obvious in the northeast areas of the US. The figure below is a reproduction of Figure 1c with a different color bar scale from -0.5 to 2×10^{15} molecules/cm^2. The low-value regions can be seen around nearly all hotspots.



- *As pointed out in the manuscript, not only the NO_2 columns over urban areas are larger at low wind speed, but also their relative changes over time. This is interesting but not explained in the paper. In my opinion, one explanation could be that at high NO_2 levels, the non-linearity in NO_2 lifetime increases the observed trends as under polluted conditions, the same reduction in NO_x emissions leads to larger reductions in NO_2 columns as it would under cleaner conditions. If this is the case, I would argue that the larger trends reported in this study are not necessarily an improvement over values derived from all wind conditions.*

Response We agree with the reviewer that larger NO_2 column reductions observed over urban areas at low wind-speed condition may be related to the non-linearity of the NO_x chemistry. At fast speed winds, the decreased NO_2 level over polluted urban areas may increase the NO_x lifetime so that the same reduction in NO_x emissions would lead to a smaller reduction in NO_2 columns compared to the slow wind-speed conditions. In the revised manuscript, we have added this possible explanation/discussion at the end of the last paragraph of Sect. 3.1.

However, we did not assert in the original manuscript that "*the larger trends reported in this study* *are … an improvement over values derived from all wind conditions*". Our points are: (1) the presence of strong winds changes the observed NO_2 trends over a number of US urban areas (i.e., greater NO_2 reductions are observed under the weak-wind condition than under the all-wind condition) (e.g., the last paragraph of Sect. 3.1 in the original manuscript); and (2) trends in NO_2 columns obtained at slow winds may better reflect the real bottom-up NO_x emission trends (e.g., the second paragraph of Sect. 3.2 in the original manuscript). Therefore, in terms of constraining emission trends, based on our results and discussion in Sect. 3.1 and 3.2 of the original manuscript, we suggest using OMI NO_2 data at slow wind speeds when comparing to the bottom-up emission trends.

- *Also with respect to the difference in emission estimates at different wind speeds I would assume that in the absence of non-linearities in NO_x chemistry, there should not be a difference in NO_x emissions or trends derived from all wind situations if the averaging areas are large enough (as usually was the case in previous studies).*

Response In the absence of non-linearity of NO_x chemistry, if there is no significant interannual variation in wind fields and if the averaging areas are large enough, we agree with the reviewer that there should not be significant differences in NO_x emission trends and NO_2 trends derived from all-wind situations or NO_2 trends derived from weak-wind situations. However, NO_x chemistry is nonlinear and there are interannual variations in the meteorological field. Both factors make the NO_2 trends derived from the all-wind condition differ from the NO_x emission trends. Our trends comparison in the second paragraph of Sect. 3.2 and the third paragraph of Sect. 3.3 of the original manuscript clearly shows that NO_2 decreasing trends in US urban areas obtained at slow winds are greater than the previously reported values obtained in all-wind conditions, and, more importantly, are closer to both the "top-down" EMG-derived NO_x emission trends and the "bottom-up" NEI NO_x emission trends, as well as surface NO_2 trends.

- *The discussion of uncertainties is in my opinion somewhat misleading as the effect of cancellation of some systematic errors in trend analysis is not taken into account. As a result, all changes in OMI derived quantities over time shown in Figure 5 are smaller than the error bars which would make them non-significant. I think this should be improved.*

Response In this work, we use exactly the same uncertainty analysis method as previous EMG-related studies such as Beirle et al. (2011) and Ialongo et al. (2014), and the terms "uncertainty" or "error" refer to one standard deviation (SD) or the coefficient of variation (SD divided by the mean) of the estimated results throughout the manuscript. From this point of view, we prefer to keep the current meaning of error bars in Figure 5. For clarification, in the revised manuscript, we have emphasized in the caption of Figure 5 that "error bars express the ±1 SD of the annually estimated results".

- *I do not see the point of Figure 8 and recommend to remove it.*

Response We still prefer to keep this figure in the current work. Figure 8 shows the sum of OMI NO_2 columns for all urban areas, and it represents the direct OMI NO_2 measurements. From this figure, we can clearly see the decrease and the pace of OMI NO_2 observations over selected US urban areas, and it supports two aspects of our discussion in Sect. 3.3. Though similar, "OMI NO_2 column" is different from another quantity used in this work "OMI NO_2 burden", because the latter is not a direct measurement but a fitted result from the EMG method.

Technical Comments

- *p14963, l19: inventories of NO_x -> inventories of NO_x emissions*

Response The manuscript has been revised correspondingly.

- *p14963, l20: bottom up inventories are uncertain but I would guess that both fuel type and technology are rather well constrained*

Response We respectfully disagree with the reviewer at this point. Even in the US, fuel type and technology are not well constrained in some sectors: the fleet of US vehicles and residential biomass burning, for example.

- *p14963, l25: not sure if current satellites really have "high temporal and spatial resolution" for NO2*

Response Take the OMI onboard the NASA-Aura satellite as an example. It has been continuously providing us NO_2 measurements at daily global coverage with the smallest pixel size of 13 km by 24 km in the past ten years. We understand that the reviewer might be unsure about whether this could be called "high temporal and spatial resolution". Therefore, we have removed this expression in the revised manuscript.

- *p14965, l24: (also elsewhere) as there are several OMI NO2 retrievals, I would replace "the OMI NO2 retrievals" by "OMI NO2 retrievals" or "TEMIS OMI NO2 retrievals"*

Response The manuscript has been revised as suggested.

- *p14966, l10: the multi-annual -> a multi-annual*

Response The manuscript has been revised correspondingly.

- *p14967, l19: make -> makes*

Response The manuscript has been revised correspondingly.

- *p14967, l23: "to smooth" – I don't think that a high sampling rate smooths the data – better sampling will lead to smoother looking averages but in fact, the level of details is higher, not lower as after smoothing.*

Response In the revised manuscript, this sentence has been revised to "all the valid pixels were oversampled on a 2 km × 2 km grid to obtain detailed spatial distributions of NO_2 over hotspots". Averaging a large number of OMI pixel data (the smallest pixel size 13 km × 24 km) to fine grids (e.g., 2 km × 2 km in this work) does help us see the detailed spatial distribution of NO_2 plumes over hot spots (see Russell et al., 2010; Fioletov et al., 2011, 2013; de Foy et al., 2009; Lu et al., 2013).

- *p14968, l16: Please add spatial resolution of ERA-interim data in km for comparison with your 2 km sampling grid*

Response We use the gridded ERA-interim data at the resolution of 0.5 degree in this work and this information has been added in the revised manuscript.

- *p14968, l25: I think this point deserves a little bit more discussion – if the NO2 plume of a point source depends on the evolution of wind speed over the last hours, why is it OK to just take the wind field at one (interpolated) time, arbitrarily selected to be 12:00 LT?*

Response First, the choice of 12:00 LT has been proven to successfully reproduce the observed spatial transport pattern of the OMI NO_2 at the daily level (Valin et al., 2013). Second, based on the work of Beirle et al. (2011), if, instead, the average wind over the last 6 hours is considered, the results only change slightly (~10%). Therefore, for simplicity, we choose 12:00 LT as the time of the wind fields. Please also note that we assigned 30% uncertainty to the wind data. In the revised manuscript, we have added some discussion in the last paragraph of Sect. 2.3 to reflect this point.

- *p14969, l10: and the longitudes -> and longitudes*

Response The manuscript has been revised correspondingly.

- *p14969, l14: one-dimension -> one-dimensional*

Response The manuscript has been revised correspondingly.

- *p14970, l16: parameter -> parameters*

Response The manuscript has been revised correspondingly.

- *p14970, l20: "we made additional treatments in processing" sounds odd to me*

Response In the revised manuscript, this sentence has been revised to "We made additional treatments to the OMI NO_2 data when using the EMG method".

- *p14971, l1: the north-westerly -> north-westerly*

Response The manuscript has been revised correspondingly.

- *p14972, l12: countries -> counties*

Response The manuscript has been revised correspondingly.

- *p14973, l2: inclusive -> included*

Response The manuscript has been revised correspondingly.

- *p14974, l20: in sum -> in summary*

Response The manuscript has been revised correspondingly.

- *p14985, l10: While I agree that "a comprehensive and integrated analysis of satellite observations, ground-based measurements, and bottom-up emissions can overcome shortcomings of the individual datasets", I don't think this has been done in the manuscript at hand.*

Response This sentence has been revised to "a comprehensive and integrated analysis of satellite observations, ground measurements, and bottom-up emissions can provide a better understanding of the true NO_x situation in a given area" in the revised manuscript.

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