

Interactive  
Comment

## ***Interactive comment on “NO<sub>x</sub> lifetimes and emissions of hotspots in polluted background estimated by satellite observations” by F. Liu et al.***

**Anonymous Referee #1**

Received and published: 6 October 2015

This manuscript, titled “NO<sub>x</sub> lifetimes and emissions of hotspots in polluted background estimated by satellite observations” by Liu et al. is an interesting addition to continuing line of research, making use of an innovative approach to make lifetime and emission estimates for sources in strong source regions. The paper is clearly written, except for a few noted word- or phase choices, and is well-suited for publication to ACP. However, there are several concerns that should be addressed or considered before being accepted for publication.

Major comments:

Wind effect: Please make note on the possible importance of comments below, or confirm or deny.

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The authors need to make it clear in the main text that uncertainty in wind speeds biases lifetime measurements low and thus biases emissions high, as shown by de Foy et al. The authors make this point clearly in the supplementary information but can make it more strongly in the main section.

Evaluation of ERA with sonde data should be moved to supplementary information. It is not the main point of the paper. However, the main findings of the wind analysis should be clearly summarized, particularly the finding that wind speeds in ERA are biased low by more than  $\sim 20\%$  at all sites, and by  $\sim 40\%$  at mountainous sites (Table S3 percent bias and  $r^2$ ). I would also expect that this bias in wind speed should be independent of the bias caused by uncertainty in wind direction (see above comment).

There is a strong diurnal increase in wind speed over land from morning to afternoon (e.g., Dai et al., 1999; 10.1029/1999JD900927). I expect that this will also bias inferred lifetimes low.

Many large sources are coastal. Sharp temperature gradients will also induce local circulation biases that may affect wind analysis in a similar manner as suggested by comments above.

The authors should briefly discuss their results in the context of de Foy et al who simulated a tracer with a well-behaved lifetime and realistic wind patterns.

Chemical effects: We do not expect the authors to fully account for all effects, but rather hope they clarify their potential impact on the results in the text.

L194 24 “we could not unambiguously relate the variability of NO<sub>x</sub> to a driving parameter like surface elevations, mean wind or latitude.” What about VOC? Could any links be made? It would seem that there should be some systematic dependence, especially with latitude. SO<sub>2</sub> has a much longer lifetime, does it have any different spatial pattern? Sources of SO<sub>2</sub> in China should be large enough to perform the analysis. If so, does that suggest that mixing processes and instrumental resolution are putting an

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upper limit on inferred lifetimes?

There are very large gradients in VOC in the regions of interest. We would expect some influence of VOC on the lifetime (reduces OH sink, but increases RO<sub>2</sub> sinks).

The authors suggest that any uncertainty in the NO<sub>2</sub>:NO ratio will only affect emission estimates. However, there are two ways in which this can interfere with inference of the lifetime. In cities where incoming O<sub>3</sub> is very low (e.g., as low as 20 ppb. Houston, Gulf air), O<sub>3</sub> production in the plume up to 100 ppb. will have a five fold effect on the NO<sub>2</sub>:NO ratio downwind (1:1 vs 5:1 -> a 60% increase in NO<sub>2</sub>:NO<sub>x</sub>), an apparent increase in NO<sub>2</sub> where the true NO<sub>x</sub> lifetime should decrease (more NO<sub>2</sub> available to react with OH as well as more RO<sub>2</sub> and OH from O<sub>3</sub> photolysis). A second effect is related to mixing and the NO:NO<sub>2</sub> ratio. The lifetime inferred by this study is very similar to values for the timescales of dilution with the free troposphere used in field studies (Zaveri et al., 2002 - doi:10.1029/2002JD003144 ; Wang et al., 2006 - 0.1029/2006GL027689 ). In the FT, winds are often faster and from a different direction than at the surface and the NO:NO<sub>2</sub> ratio favors NO due to much faster photolysis (e.g., Dickerson et al., 1997 10.1126/science.278.5339.827) and lower number densities (i.e.,  $J[\text{NO}_2]/k[\text{NO}][\text{O}_3]$ ). These effects are in addition to latitudinal and altitude impacts which are nominally mentioned in the text.

Retrieval effects: NO<sub>2</sub> products using coarse resolution inputs for converting slant columns to vertical columns have a very different urban to regional gradients than those using higher resolution inputs (e.g., Russell et al., 2011 - doi:10.5194/acp-11-8543-2011). It is unclear which is best for this purpose, as one would bias the background high whereas the other would bias urban plumes extending in to the background low, but this difference is likely worth noting.

Miscellaneous : The seasonal patterns of inferred NO<sub>x</sub> lifetime and emissions in Figure S4 indicate that there is far more uncertainty in this method than alluded to in the text. The method infers large seasonal variations of emissions (log scale) and relatively

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small seasonal variability of lifetime (linear scale). Most would expect the opposite pattern. Please make this result more clear in the text.

For Table S2 Please include more fit statistics for the summertime analysis, including number of fits that meet the criteria out of the 8 directions, and add the +/- 1-sigma lifetime inferred from different directions.

Specific comments:

Title: Consider different word use than “hotspots” in title and throughout.

180 L13-14: The last sentence in the abstract is confusing and should be clarified. In regards to the finding, can you address this at a larger scale by using the average lifetime from valid analyses over a region (e.g, E China or NE China)? Is the result the same?

183 Paragraph 1: any impact of new spectral fit? (van Geffen et al., 2015 - doi:10.5194/amt-8-1685-2015)

1183 L5 - Please, if available, cite and state numbers of any source that quantifies difference of this version of DOMINO with other products.

186 L4: Please list instead of  $r^2$  the range of inferred lifetimes and other important parameters. The model may be over-determined .

186 Footnote: Does this mean that calm winds are only 2-3% of faster winds?

189 - see major comment on NO<sub>2</sub>:NO<sub>x</sub> - a few sentences or a paragraph here should be sufficient.

191 L8-27 - This paragraph was a bit confusing. It was unclear to me whether the large decreases in the US or large increases in China would effect results by only using 2005-2008. Also, the decrease that is reported seems smaller than reported elsewhere. Does this agree with the rate of decrease observed elsewhere?

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192 I think that there should be some justification as to why European sources were not analyzed.

L198 22 - see major comments on wind effects. Please clarify here that the wind speeds are biased high by ~20% and that any additional uncertainty in direction, and potentially diurnal oscillations (i.e., sea breeze, mountain breeze ), will lead to biased lifetimes.

L198 25 - Where do these numbers come from? There are definitely conditions where the choice of NO<sub>2</sub>:NO<sub>x</sub> ratio used here is off by more than 10%. Please add reference and value for analysis of different products / validation papers.

210 and wind analyses - I would expect that the sonde data have a large influence on the ECMWF re-analysis? I would expect that the comparison at the site and sonde time (0 and 12 UTC) would be good but that might not extend to other locations and times.

214 - See major comment - If lifetime from all individual sources is averaged in some way and emissions are inverted by mass balance, is there still a large EDGAR underestimate?

Sup 9 - see major comment. Please make the results of Table S3 much more clear in the text. “Percent change” heading should be “percent difference” and please include (+) or (-) to indicate that all are biased in the same way. Also, I assume that r<sub>2</sub> is wind speed. Is there some way to indicate agreement of direction, or u and v components?

Technical comments: 181 L10: “Emissions . . . ” sentence should be re-phrased.

181 L22: “. . .allow. . .” consider re-wording.

182 L4 “hotspots” re-word

183 L10 “by” different word choice

184 L3: More descriptive section heading “Outflow model”?

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184 L8 “recap” -> “summarize”

184 L16. This source is actually reasonably isolated relative to the others. Please identify Harbin on Figure 5.

184 L1 New label? “Isolated point source outflow model: Lifetime and ENOx”

185 L1 New label? “Mixed source outflow model: Lifetime”

186 L12 New label? “Mixed source outflow model: Emissions”

186 L9 What is the the typical number of fits that meet the criteria of the 8 possible?

192 L9 - It seems like a global database of urban areas or population density would be a better classification for future reference.

211 - As mentioned elsewhere. Please label and emphasize Harbin. If possible label al locations.

Supp 5 L5 direction -> direct

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 24179, 2015.

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