

Anonymous Referee #1

*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.*

**Response:** We thank Referee #1 for the encouraging comments. All comments and suggestions have been considered carefully and well addressed.

*Major comments:*

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

**Response:** We agree that the impact of wind uncertainties on the total estimated emission uncertainty is an important issue which has to be discussed more comprehensively in the paper. We thus extended the paper in this respect by (a) reviewing the error estimate performed by Beirle et al. (2011) within Sect.2.2.1, (b) discussing the effects of uncertainty in wind speeds and directions on estimated lifetimes and emissions within the main paper, and (c) extending the discussion of sophisticated uncertainties related to winds (and other effects) in the supplement. In addition, we have adjusted the final uncertainty estimate associated with wind fields from 20% to 30%.

*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.*

**Response:** We agree that the discussion about wind comparison is very important (as stated in the last response), but the effect of the difference between ECMWF and sonde projected wind speeds on estimated lifetimes and emissions is not as drastic as it sounds (in fact, their mean unprojected winds agree within 10%), and can be understood by the sorting procedure, which is explained in detail in the supplement (see also the response for comment 3). We have added one sentence to point out this in Sect.2.6 of the revised manuscript as well.

*2. Evaluation of ERA with sonde data should be moved to supplementary information. It is not the main point of the paper.*

**Response:** We agree that the evaluation of ERA with sonde data is not the main point of the paper, but it clearly shows that mountainous wind data is highly uncertain, which we consider as an important aspect for the general applicability of our approach, and thus should be part of the main paper.

3. 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 ~20% at all sites, and by ~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).

**Response:** We summarize main findings of the wind analysis in Sect.3 of the supplement, as follows:

“We carried out a comparison of wind information between ECMWF and sounding measurements (Table S3). Here we focus on the comparison of the quantity used for the lifetime estimate, i.e. the projected wind components for each wind direction sector. We firstly sorted ECMWF wind fields for the years 2005–2013 into 8 wind direction sectors and classified the simultaneous sonde data into the same wind direction sector, and then calculate the mean of the projected wind speeds from both datasets to compare. While *total* wind speeds from ECMWF and sonde measurements agree quite well (~5% on average for wind speeds >2 m/s), the *projected* wind components are systematically higher for ECMWF. This can be expected, as ECMWF wind fields are the basis for the wind direction classification. If, for instance, the true wind would be 5 m s<sup>-1</sup> from north, but the model wind is 5 m s<sup>-1</sup> from east, the case is classified as easterly, while the actual easterly wind component is 0. That is, deviations of the wind direction (even if 0 on average) cause a systematic bias due to this projection procedure. Thus, the deviation of the projected wind speeds reflects uncertainties of the sorting procedure caused by deviations of the wind direction, and allows for an estimate of the overall uncertainty due to wind fields. The deviations for non-mountainous sites are, on average, acceptable (26%). Note also that de Foy et al. (2015) report on ERA-Interim winds yielding a better lifetime estimate compared to the North American Regional Reanalysis project (NARR). For mountainous sites, however, significantly higher deviations are found (37% on average) due to insufficient spatial resolution of ECMWF (see also Sect. 2.6 of the manuscript). ”

4. 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.

**Response:** We have discussed this in Sect.3 of the supplement, as follows:

“Wind fields often reveal systematic spatio-temporal patterns, such as diurnal cycles or land-sea transitions, which could have systematic effects on our results. As the underlying physical processes are included in the models, these effects should, in first order, be accounted for by ECMWF. However, the spatial resolution might be too coarse to capture these effects completely.

Beirle et al. (2011) varied the time of the wind data used for the fit and found changes below 10%. In addition, from the comparison with sonde data, we see no indication that ECMWF data are particularly biased for coastal cities (Miami, Xiamen). We thus

consider the uncertainties caused by diurnal cycles of wind speeds or land-sea transitions to be covered by the estimated overall uncertainty related to wind fields. Overall, we estimate the uncertainties associated with the wind data as 30% for non-mountainous sites.”

5. *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.*

**Response:** Thanks. We have added the discussion in the introduction of the revised manuscript, as follows:

“de Foy et al. (2014) further analyzed the performance of the method using model simulations with fixed a-priori lifetimes and realistic wind data, which proved that the fitted results were accurate in general and showed best performance for strong wind cases.”

6. *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  $\text{NO}_x$  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.  $\text{SO}_2$  has a much longer lifetime, does it have any different spatial pattern? Sources of  $\text{SO}_2$  in China should be large enough to perform the analysis. If so, does that suggest that mixing processes and instrumental resolution are putting an 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 RO2 sinks).*

**Response:** We thank the reviewer for these suggestions.

VOC: In order to investigate a potential link between VOCs and the estimated  $\text{NO}_x$  lifetimes, we used the tropospheric  $\text{H}_2\text{CO}$  columns as provided by BIRA (De Smedt et al., 2015) from OMI observations. We averaged the  $\text{H}_2\text{CO}$  columns for the ozone season during 2005–2013, and explore their relationship with  $\text{NO}_x$  lifetime. We observed systematic spatial patterns for the  $\text{H}_2\text{CO}$  columns, e.g., the concentration of  $\text{H}_2\text{CO}$  is higher in the eastern US than the western US, which is similar to the spatial distribution of  $\text{NO}_x$  lifetime. However, the overall correlation between  $\text{H}_2\text{CO}$  TVCDs and  $\text{NO}_x$  lifetime is rather low ( $r^2 = 0.13$ ). Thus, we see no indication that VOCs are the main driver for the spatial variability of  $\text{NO}_x$  lifetime. We have discussed this in Sect. 3.1 of the revised manuscript.

$\text{SO}_2$ : We have added the text to the end of Sect. 3.2, as follows:

“Satellite observations also enable the study of spatial and temporal distributions of  $\text{SO}_2$  emissions (e.g., Fioletov et al. (2011)) and even to obtain estimates of  $\text{SO}_2$  lifetimes and emissions under special circumstances (e.g., Beirle et al. (2014)). However, if the method developed in this study would be applied to  $\text{SO}_2$  directly, higher uncertainties have to be expected due to the longer lifetime of  $\text{SO}_2$  (see Sect. 5 of the supplement for a detailed discussion).”

We have also added a new section (Sect.5) to the supplement, as follows:

## “5. Potential applications for SO<sub>2</sub>

We have presented a method for the estimation of NO<sub>x</sub> lifetimes and emissions from space for strong sources on top of a generally polluted background.

Satellite observations of SO<sub>2</sub> have been used before for top-down estimates of emissions (e.g., Fioletov et al., 2011) and even to obtain estimates of SO<sub>2</sub> lifetimes under special circumstances. Beirle et al. (2014) analyzed downwind plume evolution of SO<sub>2</sub> from the Kilauea volcano on Hawaii and estimated the respective SO<sub>2</sub> lifetime and emissions by a method similar to that proposed in Beirle et al. (2011) for NO<sub>2</sub>. In this special case, however, wind conditions were pretty stable, and only one main wind direction had to be considered, without any sorting, due to the prevailing trade winds.

For multiple sources in polluted background and variable wind conditions, however, the situation for SO<sub>2</sub> is much more complex than for NO<sub>2</sub>: The NO<sub>2</sub> observations are sorted according to the wind direction at the time of the measurement, while the “history” (i.e. the potential impact of NO<sub>x</sub> emissions from the previous day, transported under possibly different wind conditions) is not considered. While this is appropriate for NO<sub>2</sub> due to the lifetime of a few hours, this is fundamentally different for SO<sub>2</sub> with longer lifetimes, which causes considerably higher uncertainties due to changes of wind directions. In addition, also the across-wind integration (needed to compensate for spatial dilution) as well as the fit would have to be performed on larger intervals for longer lifetimes, such that nearby sources cannot be separated from each other anymore and the quantification of SO<sub>2</sub> emissions from an individual source would be more difficult.

Thus, it might be worth testing a similar method for SO<sub>2</sub>, but one has to be aware of the potential drawbacks, and we expect a higher uncertainty of resulting emissions as a consequence of the generally longer lifetime of SO<sub>2</sub>.”

*7. 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[NO<sub>2</sub>]/k[NO][O<sub>3</sub>]). These effects are in addition to latitudinal and altitude impacts which are nominally mentioned in the text.*

**Response:** Concerning the first point, we generally agree that changes of the

NO<sub>2</sub>/NO<sub>x</sub> ratio could influence the NO<sub>x</sub> lifetime, in particular if the difference in O<sub>3</sub> concentrations between upwind and downwind plumes is significant.

We have discussed this in Sect.3 of the supplement, as follows:

“However, the NO/NO<sub>2</sub> ratio of course might differ locally, in particular when the difference in O<sub>3</sub> concentrations between upwind and downwind plumes is significant. But the influence is not dramatic on the scales of the OMI footprint (at least 13 km×24 km). In addition, the influence has been included in the overall uncertainty estimates by averaging the fit results for different wind direction sectors that usually represent different levels of incoming O<sub>3</sub>. We consider the applied correction (with an assumed uncertainty of 10%), to be adequately represented by the CTM, reflecting the mean conditions over spatial scales of ~100–200 km.”

With respect to vertical profiles, we have checked the impact of different altitudes used for the extraction of horizontal wind fields (compare also Beirle et al., 2011), and found the dependencies to be low (~10%) and covered by the overall uncertainty due to wind fields. However, we could not find the statement that fresh NO<sub>x</sub> emissions mix with the free troposphere within a few hours in the cited references: (Zaveri et al.(2002) explained the relationship between ozone production and NO<sub>x</sub> by model simulations, but the set of model seems to only consider the vertical mixing within the PBL (Sect. 4.1). The work in Wang et al. (2006) seems not to deal with the free troposphere as well.)

*8. 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.*

**Response:** Thanks. We have added the discussion in the Sect.3 of the supplementary information of the revised manuscript, as follows:

“Though the recent update of the DOMINO algorithm (Boersma et al., 2011) has improved some issues related to the spatial resolution of external databases, retrievals are still based on relatively coarsely resolved terrain height, ground albedo, and a-priori NO<sub>2</sub> vertical profile shape, probably causing low-biased VCDs over strong emission sources (e.g., Russell et al., 2011). These effects are, however, covered by the assumed uncertainty of TVCDs of 30%.”

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

**Response:** The seasonal lifetimes reveal higher uncertainties due to the smaller number of available satellite observations and thus reduced number of wind direction

sectors that yielding a valid fit, compared to the ozone season. The uncertainty is sometimes too large to get reasonable seasonal patterns for a specific location. On top of that, the emission estimate is affected by poorer statistics, in particular in case of spatial gaps, probably causing the large seasonal fluctuations found for some sites. We have clarified this in the Sect. 3.1 of the revised manuscript, as follows:

“The seasonal lifetimes reveal higher uncertainties due to a smaller number of available satellite observations compared to the ozone season and thus reduced number of wind direction sectors that yielding a valid fit. The uncertainty is sometimes too large to get reasonable seasonal patterns for a specific location. But still a systematic seasonal variability can be observed for most non-mountainous cases: mean lifetimes are found to be shorter in summer (3.2 hours) compared to spring (4.2 hours) and autumn (4.5 hours), as expected.

For some locations, the resulting emissions vary considerably over season, which again can be attributed to the poor statistics; in particular spatial gaps cause high uncertainties of the determined total NO<sub>2</sub> mass based on Eq. (5).”

*10. 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.*

**Response:** Thanks. We have added it in Table S2 of the revised manuscript

Specific comments:

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

**Response:** We have replaced “hotspots” by “Cities and power plants” in title and throughout the paper.

*12. 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?*

**Response:** The different performance between regional inventory MEIC and global inventory EDGAR could not be attributed to the difference in the total budget as the comments concerned, because the deviation in national total NO<sub>x</sub> emissions is far less (20.7 and 24.9 Tg-NO<sub>2</sub> for year 2008 in EDGAR and MEIC respectively). In addition, the extant inverse estimate at regional level has suggested that top-down national budget is close to the bottom-up emission estimate for East China (Lin et al., 2010). We have revised the last sentence in the abstract in the revised manuscript as follows: “Regional inventory shows better agreement with top-down estimates for Chinese cities compared to global inventory, most likely due to different downscaling approaches adopted in the two inventories.”

*13. 183 Paragraph 1: any impact of new spectral fit? (van Geffen et al., 2015 -doi:10.5194/amt-8-1685-2015)*

**Response:** We are aware of the recent improvements of the spectral analysis for OMI

and add the discussion about these references to the Sect.3 of the supplement, as follows:

“Recently, an overall bias of the OMI NO<sub>2</sub> column density has been reported, which turns out to be related to an imperfect spectral analysis and could be removed by improved spectral fitting procedures (van Geffen et al., 2015; Marchenkov et al., 2015). Unfortunately, the updated datasets are not available yet.

However, as an overall bias in total columns is mostly removed by the stratospheric correction procedures, we do not expect a large effect on the tropospheric NO<sub>2</sub> column densities over polluted sites, and thus no impact on our emission estimates.”

*14. 183 L5 - Please, if available, cite and state numbers of any source that quantifies difference of this version of DOMINO with other products.*

**Response:** Besides the DOMINO v2 NO<sub>2</sub> product, also NASA provides an NO<sub>2</sub> “standard product” (Bucsela et al., 2013). Both products differ in the retrieval details, in particular in the stratospheric correction and in the a-priori used for the calculation of AMFs (in particular the a-priori NO<sub>2</sub> profiles). Overall, both products show a good quantitative agreement (see Fig. 9 in Bucsela et al., 2013). Note also that any additive offset between different products (as caused by different stratospheric corrections) would have no effects on our estimated emissions due to the fitted background in Eq.(5). We have clarified this in Sect.3 of the supplement, as follows:

“The retrievals of NO<sub>2</sub> TVCDs performed by KNMI (used in this study) and NASA (OMI “Standard Product”) are based on the same spectral analysis, but differ in the separation of stratospheric and tropospheric columns and AMF calculations (Bucsela et al., 2013; Boersma et al., 2011; Boersma et al., 2007; Dirksen et al., 2011), which resulted in some significant differences in their early released products (Lamsal et al., 2010; Platt and Stutz, 2008). With the development of NO<sub>2</sub> retrieval algorithms, however, the two products are increasingly converging (Bucsela et al., 2013; Boersma et al., 2011).”

*15. 186 L4: Please list instead of  $r^2$  the range of inferred lifetimes and other important parameters. The model may be over-determined .*

**Response:** Thanks. We listed both the range of inferred lifetimes and R<sup>2</sup> in the revised manuscript.

*16. 186 Footnote: Does this mean that calm winds are only 2-3% of faster winds?*

**Response:** Yes, the projected wind speed under calm wind and windy conditions is 0.1 and 17.4 km/h on average respectively for investigated sources (for calm, slightly positive and negative projected winds almost cancel out).

*17. 189 - see major comment on NO<sub>2</sub>:NO<sub>x</sub> - a few sentences or a paragraph here should be sufficient.*

**Response:** Please see the response for comment 7.

*18. 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?*

**Response:** In theory, any large changes in NO<sub>x</sub> emissions after 2008 would affect results when comparing top-down estimates with bottom-up ones for the years 2005–2008. However, the effect is of minor importance for China. The emission changes in China is not linear after the year 2008: NO<sub>x</sub> emissions rebounded after the economic crisis around 2008 and declined again around 2012 associated with emission control regulations. Based on MEIC inventory, the average NO<sub>x</sub> emission for investigated Chinese cities for the years 2005–2008 is only 3% less than that for the years 2005–2012. Thus, we only emphasized the effect for the US in the main text.

The decline in NO<sub>2</sub> TVCDs over the US observed in this study is comparable with other studies. We observed a decline in NO<sub>2</sub> TVCDs from the period of 2005–2008 to the period of 2009–2013 with an average total reduction of  $14 \pm 9\%$  for investigated US cities. Russell et al. (2012) reported consistent decreases of NO<sub>2</sub> TVCDs in cities across the US, with an average total reduction of  $32 \pm 7\%$  during 2005–2011. The two decrease rates are comparable.

*19. L192 I think that there should be some justification as to why European sources were not analyzed.*

**Response:** For this study, we choose large sources across China and the US as the pre-selected candidates, of which the good-quality and countrywide consistent bottom-up emission information, particularly for power plants, is available. Further investigation on sources located in other regions, in particular, Europe, will be performed in the near future, with collating the corresponding bottom-up emission inventories. We have clarified this in the Sect.4 of the revised manuscript.

*20. 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.*

**Response:** Please see the response for comment 3.

*21. 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.*

**Response:** The concrete number of 1.3 used for scaling up the NO<sub>2</sub> to NO<sub>x</sub> is based on the typical assumptions made in the section 6.5.1 of Seinfeld and Pandis (2006) for “typical urban conditions and noontime sun”. Note that conditions are quite consistent in this study due to the overpass time of OMI close to noon, the selection of cloud-free observations, the focus on the ozone season, and the focus on polluted regions with generally high tropospheric ozone.

In addition, we have checked the NO<sub>x</sub>/NO<sub>2</sub> ratio at OMI overpass time within the boundary layer (up to 2 km) with the CTM EMAC (Jöckel et al., 2015) and found



values of  $1.28 \pm 0.08$  for polluted ( $\text{NO}_x > 1 \times 10^{15}$  molec  $\text{cm}^{-2}$ ) regions in China and the US for the 1<sup>st</sup> of July 2005, and similar values for all days of the ozone season (on average  $1.32 \pm 0.06$ ).

While the NO/NO<sub>2</sub> ratio of course might differ locally (in particular close to strong sources), we still consider the applied correction (with an assumed uncertainty of 10%), to be adequately represented by the CTM, as it has to represent the mean conditions over spatial scales of ~100–200 km. We have clarified this in the supplementary information of the revised manuscript.

*22. 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.*

**Response:** The sonde data are indeed incorporated in ECMWE assimilation, but still they are not expected to be the same, as multiple input data are used, and model values are not simply overwritten, but only regulated.

Thus, the deviation between the resulting assimilated ECMWF wind fields and individual sonde measurements can still be significant, in particular in mountainous regions, like shown in Table S3.

*23. 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?*

**Response:** The underestimation of EDGAR inventory is less significant at regional scale than urban scale when comparing with top-down estimates using an averaged lifetime or MEIC inventory. As stated in the response for comment 12, the underestimation could not be attributed to the total budget, as the national total NO<sub>x</sub> emissions of different inventories are comparable, but is most likely due to different downscaling approaches.

*24. 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<sup>2</sup> is wind speed. Is there some way to indicate agreement of direction, or u and v components?*

**Response:** Thanks for the suggestions. We have revised the heading and the sign in the revised manuscript. r<sup>2</sup> does not refer only to the wind speed, it considers the wind direction. We firstly sorted ECMWF wind fields for the years 2005–2013 into 8 wind direction sectors and classified the simultaneous sonde data into the same wind direction sector, and then calculate the mean of the projected wind speeds from both datasets to calculate r<sup>2</sup>. We have added a note to clarify this in the table.

*Technical comments:*

*25. 181 L10: "Emissions . . ." "sentence should be re-phrased."*

**Response:** Thanks. We have re-phrased the sentence as follows:

“Emissions at city level are often downscaled from regional emission estimates, based on surrogates (e.g. population density and industrial productivity), which however often just roughly reflects the magnitude and spatial distribution of urban emissions.”

26. 181 L22: “. . .allow. . .” *consider re-wording.*

**Response:** Thanks. We have re-phrased the sentence as follows:

“The satellite NO<sub>2</sub> measurements have been applied to quantify NO<sub>x</sub> emissions.”

27. 182 L4 “hotspots” *re-word*

**Response:** We have done it (please see the response for comment 11).

28. 183 L10 “by” *different word choice*

**Response:** We have replaced “by” by “from”. We would welcome proposals for a better formulation from the reviewer (or the ACP language editor) if needed.

29. 184 L3: *More descriptive section heading “Outflow model”?*

**Response:** Thanks. We have revised the label as “NO<sub>2</sub> outflow models and lifetime/emission fits” in the text.

30. 184 L8 “recap” -> “summarize”

**Response:** Thanks. We have re-phrased the word in the revised manuscript.

31. 184 L16. *This source is actually reasonably isolated relative to the others. Please identify Harbin on Figure 5.*

**Response:** Thanks. We have identified Harbin on Fig.5 in the revised manuscript.

32. 184 L1 *New label? “Isolated point source outflow model: Lifetime and ENO<sub>x</sub>”*

**Response:** Thanks. We have revised the label as “Isolated point source outflow model: Lifetime and Emissions” in the text.

33. 185 L1 *New label? “Mixed source outflow model: Lifetime”*

**Response:** Thanks. We have revised the label in the text.

34. 186 L12 *New label? “Mixed source outflow model: Emissions”*

**Response:** Thanks. We have revised the label in the text.

35. 186 L9 *What is the typical number of fits that meet the criteria of the 8 possible?*

**Response:** The number of fits that meet the criteria is 4 on average. We have added this in Sect.2.5 of the revised manuscript.

36. 192 L9 - *It seems like a global database of urban areas or population density would be a better classification for future reference.*

**Response:** The relationship between urban emissions and socio-economic parameters

is complex. For instance, a city with low population density does not necessarily correspond to a small amount of emissions if it has strong industrial activity. However, we do not deny that a global database of urban areas or population density would help to identify the candidates. We would like to explore which indicator is better in a future study.

37. 211 - As mentioned elsewhere. Please label and emphasize Harbin. If possible label all locations.

**Response:** Thanks. We have labelled all locations on Fig.5 in the revised manuscript.

38. Supp 5 L5 direction -> direct

**Response:** Thanks. We have revised the sentence as follows:

“The accuracy of wind fields affects our analysis twofold, by sorting the NO<sub>2</sub> TVCDs according to wind directions as well as by transferring the fitted e-folding distance into a lifetime.”

## Reference

Beirle, S., Boersma, K. F., Platt, U., Lawrence, M. G., and Wagner, T.: Megacity emissions and lifetimes of nitrogen oxides probed from space, *Science*, 333, 1737–1739, 2011.

Beirle, S., Hörmann, C., Penning de Vries, M., Dörner, S., Kern, C., and Wagner, T.: Estimating the volcanic emission rate and atmospheric lifetime of SO<sub>2</sub> from space: a case study for Kīlauea volcano, Hawai‘i, *Atmos. Chem. Phys.*, 14, 8309–8322, 10.5194/acp-14-8309-2014, 2014.

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