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Dear editor,

Concerning our study on NO_x lifetimes and emissions estimated by satellite observations (acp-2015-633), we have now submitted revisions of the manuscript and supplement.

We have considered all the comments and suggestions from the reviewers carefully and modified the manuscript accordingly. The changes mainly affect the discussion and treatment of uncertainties (section 2.3), in particular related to the involved ECMWF wind fields. In addition, the respective section in the supplement (sect. 3) was largely extended, including the discussion of possible effects of systematic spatio-temporal patterns (e.g., diurnal cycles of NO_x lifetimes) or the assumptions of a constant NO/NO_2 ratio and the treatment of the chemical decay of NO_2 as a simple first-order loss.

We have also made substantial changes to the supplement, however, In addition to the extended discussion of uncertainties (section 3), new sections about the potential impact of interfering sources (section 4) and possible application of the method for SO_2 (section 5) have been added.

While the basic method and thus the resulting lifetime and emission estimates have not been changed, the estimated uncertainties have been slightly modified by
a) adjusting the estimated uncertainty due to wind fields from 20% to 30%, and
b) using the standard mean error (instead of the standard deviation) for the estimated uncertainty of lifetimes from different wind directions.
Due to the latter choice, the final uncertainties slightly decrease.

Looking forward to hearing from you.
Sincerely yours,

Qiang Zhang

Anonymous Referee #1

This manuscript, titled "NO_x 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^{-1} from north, but the model wind is 5 m s^{-1} 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 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. SO_2 has a much longer lifetime, does it have any different spatial pattern? Sources of 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 NO_x lifetimes, we used the tropospheric H_2CO columns as provided by BIRA (De Smedt et al., 2015) from OMI observations. We averaged the H_2CO columns for the ozone season during 2005−2013, and explore their relationship with NO_x lifetime. We observed systematic spatial patterns for the H_2CO columns, e.g., the concentration of H_2CO is higher in the eastern US than the western US, which is similar to the spatial distribution of NO_x lifetime. However, the overall correlation between H_2CO TVCDs and 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 NO_x lifetime. We have discussed this in Sect. 3.1 of the revised manuscript.
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 SO_2 emissions (e.g., Fioletov et al. (2011)) and even to obtain estimates of 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 SO_2 directly, higher uncertainties have to be expected due to the longer lifetime of 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_2

We have presented a method for the estimation of NO_x lifetimes and emissions from space for strong sources on top of a generally polluted background.
Satellite observations of SO_2 have been used before for top-down estimates of emissions (e.g., Fioletov et al., 2011) and even to obtain estimates of SO_2 lifetimes under special circumstances. Beirle et al. (2014) analyzed downwind plume evolution of SO_2 from the Kilauea volcano on Hawaii and estimated the respective SO_2 lifetime and emissions by a method similar to that proposed in Beirle et al. (2011) for NO_2. 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_2 is much more complex than for NO_2: The NO_2 observations are sorted according to the wind direction at the time of the measurement, while the “history” (i.e. the potential impact of NO_x emissions from the previous day, transported under possibly different wind conditions) is not considered. While this is appropriate for NO_2 due to the lifetime of a few hours, this is fundamentally different for SO_2 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_2 emissions from an individual source would be more difficult.
Thus, it might be worth testing a similar method for SO_2, 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_2.”

7. The authors suggest that any uncertainty in the NO_2: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_3 is very low (e.g., as low as 20 ppb. Houston,Gulf air), O_3 production in the plume up to 100 ppb. will have a five fold effect on the NO_2:NO ratio downwind (1:1 vs 5:1 -> a 60% increase in NO_2:NOx), an apparent increase in NO_2 where the true NO_x lifetime should decrease (more NO_2 available to react with OH as well as more RO_2 and OH from O_3 photolysis). A second effect is related to mixing and the NO:NO_2 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_2 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[NO2]/k[NO][O3]). 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_2/NO_x ratio could influence the NO_x lifetime, in particular if the difference in O_3 concentrations between upwind and downwind plumes is significant.
We have discussed this in Sect.3 of the supplement, as follows:
"However, the NO/NO_2 ratio of course might differ locally, in particular when the difference in O_3 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_3. 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_x 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_x 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_2 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_2 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_x 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_2 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_x emissions is far less (20.7 and 24.9 Tg-NO2 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_2 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_2 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_2 product, also NASA provides an NO_2 "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_2 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_2 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_2 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^2 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_2:NO_x - 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_x 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_x 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_x 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_2 TVCDs over the US observed in this study is comparable with other studies. We observed a decline in NO_2 TVCDs from the period of 2005–2008 to the period of 2009–2013 with an average total reduction of 14 ± 9% for investigated US cities. Russell et al. (2012) reported consistent decreases of NO_2 TVCDs in cities across the US, with an average total reduction of 32 ± 7 % during 2005–2011. The two decrease rates are comparable.

19. 192 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_2:NO_x 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_2 to NO_x 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_x/NO_2 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 ± 0.08 for polluted (NO_x>1×10^{15} molec cm^{-2}) regions in China and the US for the 1st of July 2005, and similar values for all days of the ozone season (on average 1.32 ± 0.06).
While the NO/NO_2 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_x 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 r2 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^2 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^2. 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_2 measurements have been applied to quantify NO_x 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_2 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 ENOx"
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 NO2 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_2 from space: a case study for Kīlauea volcano, Hawai'i, Atmos. Chem. Phys., 14, 8309–8322, 10.5194/acp-14-8309-2014, 2014.

Boersma, K. F., Eskes, H. J., Veefkind, J. P., Brinksma, E. J., van der A, R. J., Sneep, M., van den Oord, G. H. J., Levelt, P. F., Stammes, P., Gleason, J. F., and Bucsela, E. J.: Near-real time retrieval of tropospheric NO_2 from OMI, Atmos. Chem. Phys., 7, 2103–2118, 10.5194/acp-7-2103-2007, 2007.

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO_2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, doi:10.5194/amt-4-1905-2011, 2011.

Bucsela, E. J., Krotkov, N. A., Celarier, E. A., Lamsal, L. N., Swartz, W. H., Bhartia, P. K., Boersma, K. F., Veefkind, J. P., Gleason, J. F., and Pickering, K. E.: A new stratospheric and tropospheric NO_2 retrieval algorithm for nadir-viewing satellite instruments: applications to OMI, Atmos. Meas. Tech., 6, 2607–2626, doi:10.5194/amt-6-2607-2013, 2013.

de Foy, B., Wilkins, J. L., Lu, Z., Streets, D. G., and Duncan, B. N.: Model evaluation of methods for estimating surface emissions and chemical lifetimes from satellite data, Atmos. Environ., 98, 66–77, 2014.

de Foy, B., Lu, Z., Streets, D. G., Lamsal, L. N., and Duncan, B. N.: Estimates of power plant NO_x emissions and lifetimes from OMI NO_2 satellite retrievals, Atmos. Environ., 116, 1–11, 2015.

De Smedt, I., Stavrakou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., Theys, N., Lerot, C., Gielen, C., Vigouroux, C., Hermans, C., Fayt, C., Veefkind, P., Müller, J. F., and Van Roozendael, M.: Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations, Atmos. Chem. Phys., 15, 12519–12545, doi: 10.5194/acp-15-12519-2015, 2015.

Dirksen, R. J., Boersma, K. F., Eskes, H. J., Ionov, D. V., Bucsela, E. J., Levelt, P. F., and Kelder, H. M.: Evaluation of stratospheric NO_2 retrieved from the Ozone Monitoring Instrument: Intercomparison, diurnal cycle, and trending, Journal of Geophysical Research: Atmospheres, 116, D08305, doi: 10.1029/2010JD014943, 2011.

Fioletov, V. E., McLinden, C. A., Krotkov, N., Moran, M. D., and Yang, K.: Estimation of SO_2 emissions using OMI retrievals, Geophys. Res. Lett., 38, L21811, doi: 10.1029/2011gl049402, 2011.

Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K. D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry Integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy, version 2.51), Geosci. Model Dev. Discuss., 8, 8635–8750, doi: 10.5194/gmdd-8-8635-2015, 2015.

Lamsal, L. N., Martin, R. V., van Donkelaar, A., Celarier, E. A., Bucsela, E. J., Boersma, K. F., Dirksen, R., Luo, C., and Wang, Y.: Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, Journal of Geophysical Research: Atmospheres, 115, D05302, doi: 10.1029/2009JD013351, 2010.

Lin, J. T., McElroy, M. B., and Boersma, K. F.: Constraint of anthropogenic NO_x emissions in China from different sectors: a new methodology using multiple satellite retrievals, Atmos. Chem. Phys., 10, 63-78, doi: 10.5194/acp-10-63-2010, 2010.

Marchenko, S., Krotkov, N. A., Lamsal, L. N., Celarier, E. A., Swartz, W. H., and Bucsela, E. J.: Revising the slant column density retrieval of nitrogen dioxide observed by the Ozone Monitoring Instrument, Journal of Geophysical Research: Atmospheres, 120, 5670–5692, doi:10.1002/2014jd022913, 2015.

Platt, U., and Stutz, J.: Differential absorption spectroscopy, 135–174, Berlin and Heidelberg, Germany, Springer, 2008.

Russell, A. R., Perring, A. E., Valin, L. C., Bucsela, E. J., Browne, E. C., Wooldridge, P. J., and Cohen, R. C.: A high spatial resolution retrieval of NO_2 column densities from OMI: method and evaluation, Atmos. Chem. Phys., 11, 8543–8554, doi: 10.5194/acp-11-8543-2011, 2011.

Anonymous Referee #2

General comments

The proposed method is an extension of a previous method developed by the same author's for estimating NO_x emission and lifetime from satellite-based observations. It is a very elegant approach, as not dependent on modeling assumptions. In this manuscript the method is extended to sources located in polluted background, while it was presented originally only for megacities with relatively low background pollution. Uncertainties on emission estimates are still very large and this study contributes n reducing these uncertainties. The paper is well written and the methodology appropriate. I recommend publication on ACP after addressing the following specific and technical comments.

Response: We thank Referee #2 for the encouraging comments. We addressed the comments carefully as below.

Specific comments

1) P24182 L9 You could maybe mention the nominal spatial resolution at nadir here.

Response: Thanks. We have mentioned it in the revised manuscript.

2) P24 L13-14 I think the reference to other works could be improved.

You might want to cite a similar methodology for fitting described by: Fioletov, V. E., C. A. McLinden, N. Krotkov, M. D. Moran, and K. Yang (2011), Estimation of SO_2 emissions using OMI retrievals, Geophys. Res. Lett., 38, L21811, doi:10.1029/2011GL049402.

Or more recently in: Fioletov, V. E., C. A. McLinden, N. Krotkov, and C. Li (2015), Lifetimes and emissions of SO_2 from point sources estimated from OMI. Geophys. Res. Lett., 42, 1969–1976. doi: 10.1002/2015GL063148."

You could also discuss more in the introduction for example the results (including e.g. the advantages and disadvantages) of the methodologies presented by Valin et al. (2013), Lu et al. (2015) and de Foy et al. (2015). At the moment these papers are just mentioned. What were their main features and results?

Response: We have clarified the main features and results of the above references in the revised manuscript, as follows:

"In a recent study, Beirle et al. (2011) averaged OMI NO_2 measurements separately for different wind directions, thereby constructing clear downwind plumes which allow a simultaneous fit of the effective NO_x lifetimes and emissions, without the need of a chemical model. Valin et al. (2013) adopted this approach, but rotated satellite NO_2 observations according to wind directions such that all the NO_2 columns are aligned in one direction (from upwind to downwind). The rotation procedure accumulated a statistically significant data set to examine the dependence of NO_x lifetime on the wind speed. Following studies e.g. de Foy et al. (2015) and Lu et al. (2015) adopted this plume rotation technique and quantified NO_x emissions from isolated power plants and cities over the US respectively, which showed that the method can gave reliable estimates over multi-annual averages and even provide

estimates of emission trends with reasonable accuracy. 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. Alternative approaches based on model functions with multiple dimensions, e.g. a two dimensional Gaussian functions (Fioletov et al., 2011) and a three dimensional function (Fioletov et al., 2015), were also proposed to estimate lifetimes and emissions. “

Could you also comment on the applicability of your methodology for SO_2 polluted sources too somewhere in the manuscript?

Response: We thank the reviewer for this request, which is of course an obvious question. 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 SO_2 emissions (e.g., Fioletov et al. (2011)) and even to obtain estimates of 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 SO_2 directly, higher uncertainties have to be expected due to the longer lifetime of 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_2

We have presented a method for the estimation of NO_x lifetimes and emissions from space for strong sources on top of a generally polluted background.

Satellite observations of SO_2 have been used before for top-down estimates of emissions (e.g., Fioletov et al., 2011) and even to obtain estimates of SO_2 lifetimes under special circumstances. Beirle et al. (2014) analyzed downwind plume evolution of SO_2 from the Kilauea volcano on Hawaii and estimated the respective SO_2 lifetime and emissions by a method similar to that proposed in Beirle et al. (2011) for NO_2. 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_2 is much more complex than for NO_2: The NO_2 observations are sorted according to the wind direction at the time of the measurement, while the “history” (i.e. the potential impact of NO_x emissions from the previous day, transported under possibly different wind conditions) is not considered. While this is appropriate for NO_2 due to the lifetime of a few hours, this is fundamentally different for SO_2 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_2 emissions from an individual source would be more difficult.

Thus, it might be worth testing a similar method for SO_2, 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_2. ”

3) P24186 L7-8 You mention here that 8 wind sectors are used for lifetime estimation but later in section 2.2.3a only 4 sectors are considered for Eq. 5 when emissions are estimated. Could you comment on that?
Response: We have clarified this in the sect. 2.2.3 of the revised manuscript, as follows:
“Note that the projections of line densities under calm wind conditions for opposite wind direction sectors, e.g., north and south, are just mirrored. Thus, we combined the projections for opposite wind direction sectors.”

4) P24190 L15 Because only clear sky pixels are considered you might want to comment also on the eventual bias on emission and lifetime due to for example to specific wind patterns and accelerated photochemistry under clear sky conditions.
Response: We agree that the selection of cloud-free OMI NO_2 TVCDs used for fitting lifetimes and emissions does not represent the average level for all days, due to the accelerated photochemistry and different meteorological conditions (e.g. boundary layer height, atmospheric transport) under clear sky conditions. But still the emission estimates are appropriate, as both the NO_x lifetime and total mass derived from the NO_2 TVCDs are derived consistently, both of which reflect the values under clear sky conditions. Thus, this effect is of minor importance for this study and is not expected to bias the estimates of NO_x emissions. We have included this aspect in Sect. 3 of the revised supplement.

5) P24191 L25 and Fig. S3 I think that this kind of methods would be useful to estimate changes in emissions over time. Would it be feasible to estimate the emissions for these two different periods (2005-2008 and 2009-2013) in order to quantify the emission reduction expected in US east-coast? If so, could you provide the results?
Response: We have reprocessed the data for the US according to the reviewer’s comment. Unfortunately, the fit procedure of emissions only works for a very limited number of sources for the period of 2009–2013, due to the lack of observations as a consequence of the row anomaly after 2008. However, the capability of estimating emissions for shorter time periods will be enhanced with future satellite instrument like TROPOMI (Veefkind et al., 2012) featuring higher spatial resolution, and in particular by upcoming geostationary satellite instruments, as stated in the conclusions.

6) P24200 L16-18 Could you comment more on how the methodology is applicable elsewhere, e.g. in Europe? I suppose there the emission source patterns might be even on smaller scale. In the original paper (Beirle et al. 2011) only 2 European cities, Madrid and Moscow, were included, and Helsinki (plus Saint Petersburg and Stockholm) in a following paper by Ialongo et al. (2014) so I suppose Europe would

be one of the main areas to assess the applicability of this new method. Could you comment on that?

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.

More in general, could the method be applied to sources smaller than 1x10ˆ15 molec/cm^2 if the fit results are good? How small the source could be? Is there a minimum ratio between the source and the background, which is critical for the fitting performances? And how close the sources can be to each other to successfully perform the fit? Could you comment on these issues?

Response: In general, we would agree that the method would work for smaller sources as well, if the statistics are sufficient (see e.g. Beirle et al. (2004)). But the uncertainty of the lifetime and emissions fit is much higher for smaller sources.

Thus, we dismissed the very small sources by applying a threshold of 1×10^{15} molec/cm^2 in order to assure the reliability of the fitted results, and avoid systematic biases due to potential spatially varying artefacts in spectral analysis or the calculation of AMFs.

As for the distance between sources, we performed a sensitivity analysis, which is included as a new section (Sect. 4 of the supplement), as follows:

"As for the distance between sources, we find that it is not critical for the fit of lifetime, as the actual distribution of sources is appropriately accounted for by *C(x)*. But for the fit of the total mass, a decision of the extent of the source under investigation has to be made. Here, we define the extent of the city to be $\pm$20 km and integrate the calm VCDs in across-wind direction over this interval. Thus, any interference within 20 km will automatically be assigned to the source of interest.

We performed a sensitivity analysis to investigate the effect of the distance between sources on the estimate of emissions. We simulated the line densities of a single source with emissions of 500 molec-NO_2/s and with an additional source with emissions of 10%, 25% and 100% of the source of interest at 0–50 km distance, respectively, assuming an a-priori lifetime of 3 hours with a spatial smoothing following a Gaussian function with a standard deviation of 10 km. We then performed a non-linear least-squares fit of the modified Gaussian function *g(x)* (Eq.(5)) to the synthetic line densities, as illustrated in Fig. S7.

Generally, the fit cannot distinguish the source and the interference within 20 km, which tries to "explain" the interference by a larger emission. In the examples shown in Fig.S7, a 10%–100% of interference results in emission estimates which generally include the interfering source. From a distance of 30 km on, the performance of the fit gets more and more unstable, due to the interference. For distances of 40 km (and larger), the fit works properly again with a bias of less than 5% for most cases, and correctly separates the source of interest from the interfering source.

However, if the interference is comparably large as the source (500 molec-NO_2), uncertainties are large. Thus, we conclude that our method generally is applicable for regional dominant sources within about a radius of 100 km. Interfering sources within 20 km cannot be separated, but will be included in the emission estimate. From 40 km on, interfering sources will not be included. ”

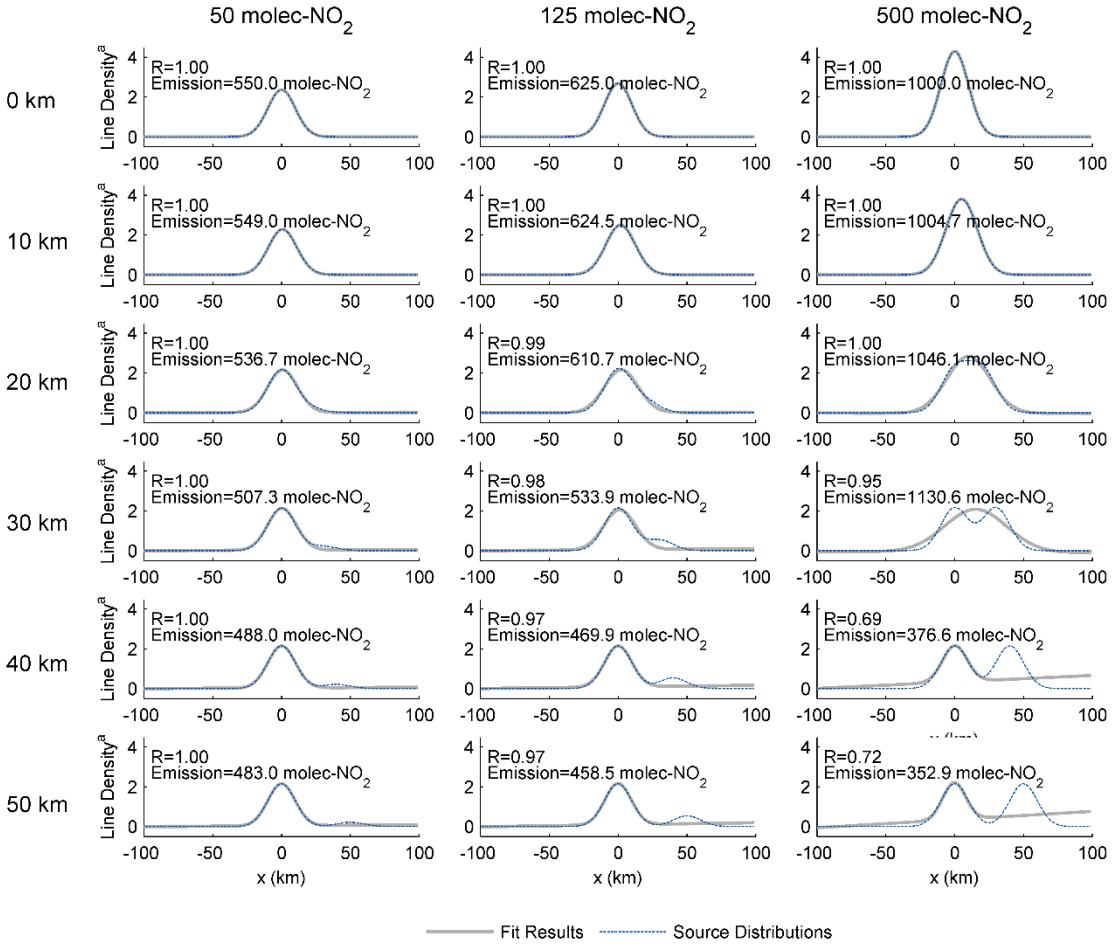

Figure S7. Sensitivity of the fitted emission to the distance between sources. Blue dot: synthetic line densities of a single source with emissions of 500 molec-NO_2/s under calm wind condition and with an additional source with emissions of 50, 125 and 500 molec-NO_2/s (from left to right) at 0–50 km (from top to bottom). Grey: emission fit based on *g(x)* (Eq. 5). The number indicates Emission resulting from the least-squares fit with 95% CI.

[a]Line Density: NO_2 line density (10^{23} molec/cm)

Technical corrections

P25197 L14 explaintion -> explanation

Response: Done.

Fig. S4 The yellow color chosen for spring and autumn are very similar, especially in a very busy figure like this is. Maybe you could replace the autumn yellow with something closer to lime or green? Or any other color you can distinguish a bit better?

Response: Thanks. We have revised the figure accordingly.

References section Several references (for example Butler et al., Gu et al., Levelt et al., Martin et al., Richter et al.) have the title not starting with a capital letter: you might want to check through. I think they should go with capital letter.
Response: Thanks. We have checked through and revised the references accordingly.

Reference

Beirle, S., Platt, U., von Glasow, R., Wenig, M., and Wagner, T.: Estimate of nitrogen oxide emissions from shipping by satellite remote sensing, Geophys. Res. Lett., 31, L18102, doi: 10.1029/2004GL020312, 2004.

Veefkind, J. P., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., Claas, J., Eskes, H. J., de Haan, J. F., Kleipool, Q., van Weele, M., Hasekamp, O., Hoogeveen, R., Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and Levelt, P. F.: TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications, Remote Sens. Environ., 120, 70–83, 2012.

Anonymous Referee #3

The manuscript introduces a creative way of quantifying the NO_x emissions from the satellite NO_2 retrievals for both power plant and urban sources located in the polluted background. It is well written and includes the detailed discussion on uncertainties in the developed method. I recommend publication of this manuscript after revisions based on the comments below. Since the manuscript can mislead the readers and future studies, careful revisions and another review of the revised manuscript may be necessary.

Response: We thank Referee #3 for the comments. We addressed the comments carefully as below.

The strength of this paper is the new method applicable to the sources in the polluted background. However, due to uncertainties in the estimated emissions from this method, the assessments of the bottom-up emission inventories with respect to the emissions in this study should be documented more carefully. For an example, the statement in the abstract, "Global inventory significantly underestimated NOx emissions in Chinese cities, most likely due to uncertainties associated with downscaling approaches" assumes that the emissions in this study are accurate. The emissions in this study from power plants are compared with the ones from CPED or eGRID, which is used as a strong support for excellent performances of the method. Looking at Figure 7, the agreement between the emissions in this study and the bottom-up inventories is not satisfactory, especially for the US, and numbers of power plants used are limited. Improved methodologies to derive the bottom-up emission inventory, MEIC are highlighted. But it does not guarantee accurate resulting emissions.

Response: We recognize the general concern raised by the reviewer and fully agree that the emissions derived in this study, as well as those provided by bottom-up inventories are subject to uncertainties. In this study, we try to quantify the uncertainties of our method as best as possible, and we have extended the uncertainty discussion in the revised paper accordingly.

Bottom-up emission inventories, developed by different researchers, often differ significantly from each other, due to the application of various assumptions and extrapolations associated with the limited knowledge of activity data and emission factors. The method developed in this study provides a top-down estimate which can be used for an independent evaluation of bottom-up inventories.

Concerning the comparison of our emission estimates with eGRID for power plants in the US, we consider the agreement to be not perfect, of course, but rather good (within 50% for all power plants, which is well within the estimated uncertainties). A larger number of included power plants would of course be desirable, but we carefully defined automated selection criteria where our method yields robust emission estimates.

We agree that the accuracy of urban emissions in MEIC is probably not as good as that of emissions from power plants. However, MEIC included multiple in-house

high-resolution databases, which is expected to improve the accuracy of emission estimates. In addition, the accuracy of MEIC has been validated by extant researches, e.g., Ding et al. (2015). We thus consider it as state-of-the-art bottom-up emission inventory, and well suited for a comparison to our top-down estimates.

In addition, errors in the ECMWF wind speed were not discussed in the manuscript. Table S3 in the supplementary material shows overestimated wind speed in ECMWF, which could underestimate NO_x lifetime and increase the estimated emission rate.

Response: Thanks. We have emphasized the discussion on effect of uncertainties of wind speeds on fitted results 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^{-1} from north, but the model wind is 5 m s^{-1} 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 covers uncertainties of the sorting procedure caused by deviations of the wind direction, and allow 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). **"**

To evaluate the method thoroughly, extensive validations of the developed emission estimations (and bottom-up emission inventories) utilizing independent data set and/or regional chemical transport models will be required.

Response: We consider our manuscript as proposal of a new method for top-down emission estimates of NO_x in polluted background, which was not possible with previous methods. We carefully discussed and quantified the uncertainties of our method, and extended the revised manuscript in this respect. Of course, further evaluation of the performance of our method with independent data sets and regional CTMs would be desirable, but is beyond of this conceptual study. We feel that using CTMs is a good plus, but not necessary for this work. In fact, in many previous published studies which used Gaussian fitting models to derive emissions, CTMs are not involved (e.g., Beirle et al., 2011; Fioletov et al., 2011; Lu et al., 2015).

We would like to point out, however, that our method provides an independent emissions quantification approach for the comparison to, and validation of, bottom-up inventories without involvement of CTMs.

Regarding the method developed in this study, the background level of NO_2 ($\varepsilon_i+\beta_i x$) can have information on the emissions from the source of interest since the lifetime of NO_2 is much shorter than relatively passive scalars such as CO and CH_4.

Response: Our method aims for emission estimates of local sources in generally polluted regions. Thus, we cannot estimate the emissions directly from the absolute measured tropospheric column, but have to account for the "background". In a first approach, we have just fitted Gaussian functions plus a constant offset to *C(x)*, which, however, often is not sufficiently reflecting the observed spatial patterns for calm winds. We thus added one further parameter, i.e. a spatially variable (linear) background, as the simplest possible expansion of the model function, which improved the performance of the fit significantly in many cases.

The reason for the need of a spatial variation of the background is related to the spatial distribution of sources, which is often not symmetric, and a possible gradient in the upper tropospheric NO_2.

In addition, the chemical lifetime defined in this study is an e-folding time. Whether the lifetime can be directly used for derivation of emission rate without application of an empirical coefficient or a weighting factor is a question.

Response: In this study, we assumed that the removal of NO_2 can be simply described by a first order loss, and thus the chemical decay of NO_2 follows an exponential decay function *e(x)* (Eq. 2) with an e-folding distance x_0, which yields an overall, effective lifetime τ. From the good lifetime fit performance, we see no indications that this assumption is insufficient.

In Beirle et al. (2014), it was investigated how far the estimated lifetime by this approach might be biased in case of temporal fluctuations of both emissions and instantaneous lifetimes. The impact of such fluctuations was found to be rather small. In the revised paper, we briefly discussed this effect in section 2.2.2.

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 SO2 from space: a case study for Kīlauea volcano, Hawai'i, Atmos. Chem. Phys., 14, 8309–8322, doi:10.5194/acp-14-8309-2014, 2014.

Ding, J., van der A, R. J., Mijling, B., Levelt, P. F., and Hao, N.: NO_x emission estimates during the 2014 Youth Olympic Games in Nanjing, Atmos. Chem. Phys., 15, 9399–9412, 10.5194/acp-15-9399-2015, 2015.

Fioletov, V. E., McLinden, C. A., Krotkov, N., Moran, M. D., and Yang, K.: Estimation of SO2 emissions using OMI retrievals, Geophys. Res. Lett., 38, L21811, doi:10.1029/2011gl049402, 2011.

Lu, Z., Streets, D. G., de Foy, B., Lamsal, L. N., Duncan, B. N., and Xing, J.: Emissions of nitrogen oxides from US urban areas: estimation from Ozone Monitoring Instrument retrievals for 2005–2014, Atmos. Chem. Phys., 15, 10367-10383, doi:10.5194/acp-15-10367-2015, 2015.

Anonymous Referee #4

Overall an interesting and relevant paper. The data are well presented, the measuring and analysis methods seems to me sound although the many fitting, scaling and filtering functions used under different situations with different areal extend makes me confused from time to time.

Response: We thank Referee #4 for the comments. In order to avoid confusion, we tabulated the fit intervals for lifetime and emission fits in a new Table 1 of the revised manuscript.

The authors state that the mean lifetime is derived from the change of the observed NO_2 patterns under windy vs. calm conditions. But if I understand the text well enough, N is derived from C and C is the line density under calm wind only as states into the text (near Eq 4). So this would be the blue lines in Figure 2 since these are the line densities for calm winds? In the figure caption on the contrary, N is fitted to the windy conditions for the different wind sectors (grey line on red crosses). Please clarify, since I am confused.

Response: The basic idea of the method is that patterns of line densities under windy conditions result from the transport, chemical decay and spatial smoothing of emission patterns. We used the line density under calm conditions, *C(x)*, as the proxy of emission patterns and performed a non-linear least-squares fit of *N(x)* (Eq. 4) to the observed NO_2 patterns under windy conditions, with the observed *C(x)* as fixed input and x_0, a and b as fit parameters. Thus, we state that results are derived from the change of the observed NO_2 patterns under windy versus calm conditions. We have clarified this in Sect.2.2.2 of the revised manuscript, as follows:

"The patterns of line densities under windy conditions result from the transport, chemical decay and spatial smoothing of emission patterns. The basic idea is to use the NO_2 patterns observed under calm conditions, *C(x)*, as proxy of emission patterns instead of assuming a single point source as in previous studies. Lifetime information is then gained based on the observed change of the NO_2 patterns under windy versus calm conditions."

It is also not clear to me why you subtract wind speeds between windy and calm conditions for use in deriving the life time. If it is not of a big effect as stated in the footnote 1 why bother?

Response: As the mean wind speed for the selection of days classified as calm is low, but not zero, line density under calm wind conditions *C(x)* is already shifted with respect to the emission pattern. In our study, the correction of this effect (i.e. taking the wind speed offset to calm conditions) is only marginal (so we put it in a footnote). However, we still would like to discuss this systematic effect in the manuscript as a general characteristic of our method; for different conditions (e.g. if a higher threshold for calm is chosen and wind directions are persistent), this effect might actually become significant.

The NO_2 amount A on top of the background is determined by fitting the functions $g_i(x)$ simultaneously for all available wind directions. What do the authors mean with" simultaneously"? Do they mean that they fit it for the 8 different wind sectors at the same time and still only retrieve one A? Please rephrase and clarify.

Response: We have rephrased this in Sect.2.2.3 of the revised manuscript, as follows:
"While the e-folding distance is fitted for each wind direction separately (and mean lifetimes might actually be different for each wind direction), the emissions are not expected to depend on wind direction. We thus use all available wind directions to perform one fit of all functions $g_i(x)$ simultaneously with wind sector dependent backgrounds, but one overall parameter *A*."

The possible linear gradient in the back ground of Equation 5: how can this be explained? Is it also possible that it results from interannual trends in the emissions over the area for the NO_2 period under investigation?

Response: Our method aims for emission estimates of local sources in generally polluted regions. Thus, we cannot estimate the emissions directly from the absolute measured tropospheric column, but have to account for the "background". In a first approach, we have just fitted Gaussian functions plus a constant offset to $C(x)$, which, however, often is not sufficiently reflecting the observed spatial patterns for calm winds. We thus added one further parameter, i.e. a spatially variable (linear) background, as the simplest possible expansion of the model function, which improved the performance of the fit significantly in many cases.
The reason for the need of a spatial variation of the background is related to the spatial distribution of sources, which is often not symmetric, and a possible gradient in the upper tropospheric NO_2.

The fit interval h is not well introduced in the main text. Suddenly it pops up. Please clarify.

Response: We have clarified this in Sect.2.2.3 of the revised manuscript, as follows:
"The fit of total NO_2 mass is performed over the interval *h* in wind direction (see Fig. S2)."

L26, P24189: replace "division" by "dividing". L9, P24192: should be "visually inspection".

Response: Thanks. We have revised the manuscript accordingly.

Figure 5: Why not using the same color bar range for both panels to stress the difference in total NO_2 columns between China and US?

Response: Thanks. We have chosen the same color bar for NO_2 TVCDs for both China and the US in the revised manuscript.