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. ”

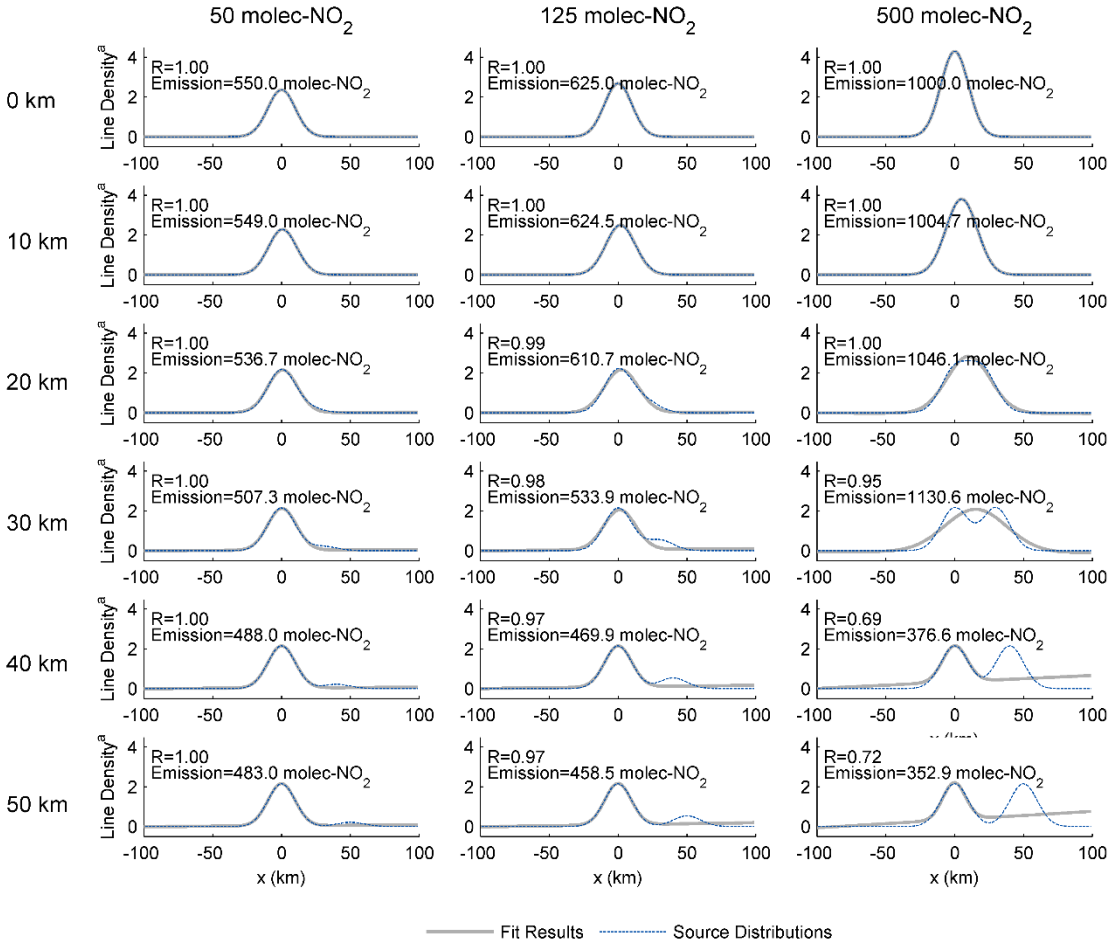

Figure S7. Sensitivity of the fitted emission to the distance between sources. Blue dot: synthetic line densities of a single source with emissions of 500 molec-NO_2/s under calm wind condition and with an additional source with emissions of 50, 125 and 500 molec-NO_2/s (from left to right) at 0–50 km (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.