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

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