

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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub>.

*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<sub>2</sub> 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<sub>2</sub> columns between China and US?*

**Response:** Thanks. We have chosen the same color bar for NO<sub>2</sub> TVCDs for both China and the US in the revised manuscript.