Reply to comments from Referee #1

We would like to thank the referee for the comprehensive and thoughtful review, and helpful comments which are addressed individually in the response below. The reviewer's comments are included in italics with the responses in blue.

The study by Lauster et al. describes a new method to quantify the NOx emissions from a motorway using two MAX-DOAS in parallel. This method is new and complementary to the existing ones, the analysis appears valid, and the presentation of the results in the paper is in general clear, although there is room for improvement in this respect. The experiment also addresses a hot topic regarding air quality. This work fits well in the scope of AMT. Therefore this work should be published, once the authors have taken into account the following remarks.

Major points:

One limitation of this study is its small database. To my understanding, there was only one day of measurements (10 May 2019). This date appears in the main text only in section 3 (I know it is in the caption of Fig. 1). It is fine to demonstrate a new technique with a small database, but this should be clear in the text. That means adding the date of experiment to the sentences of the abstract and conclusion which gives the factor 11+/-7. In case the authors performed more of such measurements but could only use those of May 10 for some reasons, it would be interesting to (briefly) explain what the problems were.

We have added the date to the respective sentences in abstract and conclusion. Also, we added (l. 39 in the revised manuscript):
"The presented results are based on one day of measurements (10 May 2019) for proof of concepts. Further measurements could then be used to analyse, e.g., different driving conditions in more detail."
Indeed, we only have one measurement day with this setup (including weather station and using this viewing geometry). The primary aim of our study is to present a proof of concept of the measurement method. Further studies could then include different measurement conditions (e.g. weekdays vs. weekend, different seasons) as well as additional measurement sites to investigate different driving conditions (e.g. speed limits, slope of the motorway). Such an extensive study, however, is beyond the scope of this manuscript.

It's confusing that the legends indicate 'west side', 'east side' in Fig. 2 and Fig. A1, since they show measurements when both instruments were on the west side to record reference measurements. I suggest to label the instruments e.g. A and B across the text and figures instead (keeping the west side, east side where it makes sense).

We see the point and adapted the legends and the text accordingly.

l. 125 and below: Can the authors explain why they use the non filtered SCDtraffic estimate in the main text, if they have filtered the clouds in A2? Does the statement that the 'clouds have only a small impact' refer to the 16% of A2? If so, this is more important than the standard error of the mean (5%) and thus not 'a small impact'.

In the main text, we refer to the unfiltered case as no clear relation between the cloudiness and the NO_2 signal is seen. However, we agree to the referee that a more accurate error estimation should include the deviation of 16%. We therefore added this deviation as an additional error to the traffic induced NO_2 SCD and the following processing steps. Also, we dropped the sentence that "clouds have a small impact" (l.132) to avoid further confusion. In the end, the additional error has no significant effect on the outcome of the emission estimation.

l.177: 'Our simulations with CAABA confirm...' -> The O3 concentration is indeed an important parameter is in the NO2/NO evaluation, one can imagine that the atmospheric mixing is as well. Could the authors add a figure with these simulations, e.g in the appendix? If the NO2/NO ratio is stable in the O3 conditions on 10 May 2019 in Mainz, it is interesting to know in which O3 conditions this ratio is not stable.

We thank the referee for this important comment. Indeed we did not yet consider the titration of ozone close to the source, where NO concentrations are very high and ozone becomes depleted. This will stop further conversion of NO to NO_2. However, turbulent mixing with ambient air increases with distance from the source. Thereby, ozone in the emitted air parcel is replenished and the oxidation of NO continues.
In the revised version of the manuscript, we apply a Gaussian dispersion model using Pasquill stability classes (Pandis and Seinfeld, 2006) based on the atmospheric stability on the measurement day. With this dispersion model we estimate the extent of the emission plume and derive the NO_2 mixing ratio from our measurements. While turbulence induced by the local topography and obstacles like trees is neglected, it helps to estimate the evolution of NO_2 mixing ratio between emission source and measurement location. From the comparison of the dispersion model and the observations, we conclude that the ozone-limited chemical regime only prevails very close to the emission source.
In order to consider this in our emission estimate calculation, we subdivide the transport of the air parcel in two sections: 1) Close to the emission source we assume that only negligible amounts of NO are converted into NO_2 and no further conversion takes place as ozone is depleted. 2) Turbulent mixing with ambient air refills the ozone reservoir and NO to NO_2 conversion can be described by the CAABA model simulations. For simplicity, we chose the distance at which the initial NO_2 mixing ratio of CAABA model simulations is reached as the transition between both sections.
As the new approach shortens the time for NO to NO_2 conversion, it is found that the NO_2/NO_x ratio is smaller than assumed in the previous approach without considering ozone limitations. Since both approaches yield the same results within the error estimation, a modification of the given NO_2/NO_x ratio was not deemed necessary.
The revised approach is described in the text (l. 195 and Appendix C1 in the revised manuscript) including a figure to the simulations.

l. 188 The authors could refer to previous experiments which indicate that it is unlikely that the NO2/NO equilibrium would be reached so close to a source, e.g. the airborne measurements of NOx fluxes from power plants (see for instance the Phd of A. Meier, Uni. Bremen), or similar studies.

We have added (in l.214 of the revised manuscript):

"However, it is rather unlikely that the equilibrium state is reached so close to the emission source (as also found for airborne measurements of emission fluxes from power plants; Meier, 2018)."
Similar to other studies, the NO/NO_2 emission rate only stabilises at a distance of 3-5 km from the source. Therefore, we do not expect to measure the equilibrium state already at a distance of a few hundred metres. But from our simulations we can conclude that a large part of the emitted NO was already converted to NO_2.

In appendix A1, the statement 'As for cloud free condition a constant CI is expected' is misleading since the CI varies, even without any clouds, with the sun position (see e.g. Gielen, 2014). In practice, this statement is only valid because of the short considered time period, please rephrase.

Agreed and changed to "An almost constant CI is expected for cloud free conditions in this time period."

The last sentence of the appendix 'a constant wind is advantageous for the measurements' is an important take-home message and should be explicit in Sect 3.2 and in the conclusions.

Agreed and added in l.173 and l.293 of the revised manuscript.

I have several smaller suggestions to improve the presentation, see below.

Minor points:

l.8: 'independent' -> independently ?

Done.

l.13: 'A large fraction of the global emissions' -> can the authors be quantitative on this fraction? 'Therefore'-> does not seem an appropriate adverb here since it's not linked by cause to the previous sentence, what about 'Moreover'?

According to the 5th assessment report of the IPCC (2013), the anthropogenic emissions of NO_x account for approximately three-quarters of the global NO_x emissions. The phrase reads now "About three-quarters of the global emissions of NO_x originate from anthropogenic sources (IPCC, 2013)." In the next sentence, "Therefore" was changed to "Moreover".

l.30: 'need to convert NO into NO2 as they directly measure the exhaust plume'-> can the author briefly explain why? (the emissions are mainly NO?). It makes sense to detail also since the reference is in German.

The paragraph was revised, focusing more on the general approaches used in other studies. Thereby, this sentence dropped out. The study by Pöhler and Engel (2019) internally measures NO_2, but as they directly measure the exhaust plume mainly NO (which is produced in the combustion process) is present in the sample. Therefore, the sampled NO is converted into NO_2 before the measurement.

l.55: 'the differential SCD yields the integrated tropo concentration of a specific trace gas'. This seems too short to be accurate. Please specify that the integration is along the photon path and that this quantity is relative (differential) to the column in the reference spectrum.

Agreed and rephrased more detailed. It reads now:
"Then, the differential SCD yields the integrated tropospheric concentration of a specific trace gas along the photon path (for an altitude range from the surface up to about 2 to 3 km; Frieß et al., 2019, and references therein), i.e. the column density relative to the reference spectrum."

l.68: perpendicular -> almost perpendicular?

Done.

l. 71: 'Possible source of NOx' although that may seem obvious to the authors, I suggest to add that 'since no fire was detected in the area' for other readers

We have added "since no other sources (e.g. fires) were detected in the area".

Spectral analysis: presenting the DOAS fit parameters (window, cross-sections, polynomial orders...) in a dedicated table would be more readable and synthetic.

Done.

l. 98: 'As can be seen in the grey area' -> actually not much can be seen in the grey area due to the y-axis scale of the lower subplot. I suggest to redo this figure 2, with the second subplot zoomed in the time period of the grey area so that we really see that the delta is about 4e14. This would also make the subplots less redundant.

Thanks for this suggestion. We changed the plot accordingly.

In the text, the authors should also explain what this delta is in practice (interpolated? one channel assumed constant?) since the measurements do not appear synchronized in time.

It is correct that the measurements are not ideally synchronised in time. Therefore, to obtain the difference between the two instruments, the time series of one instrument was interpolated onto the time axis of the other. A corresponding sentence was added to the manuscript (l. 106 in the revised manuscript).

l. 104 'spectra are being integrated' -> '... averaged' ?

We have changed this to „accumulated".

l. 285 'as shown in fig.2'-> 'fig. A2'?

We have changed "90° measurements as shown in Fig. 2" to "90° measurements (compare to Fig. 2)" as we refer to the same 90° measurements for which the NO_2 results are depicted in Fig. 2.

l. 292 'in Fig. A3 where the dashed line indicates ... threshold' -> Fig A2 ?

Thanks for pointing this out. We mixed up the sentences. It is now corrected to the following:
"The reference was inferred by fitting a 2nd order polynomial to the data and is depicted as dashed line. The filtered time series are displayed in Fig. A3."

l. 112 It's expected that the error trends follows the RMS, as it is expected that the RMS decreases with increasing integration times. Please add a few words on the physical explanation (shot noise ...)

We have changed the following sentence
"Although the average RMS decreases for longer integration times, the NO_2 retrieval yields the same result regardless of the integration time."
to
"For the short integration times of our measurements, the spectral residual of the fit is dominated by photon shot noise. This is also clearly demonstrated by the observed dependence of the RMS (and the fit error) on integration time. The RMS decreases for longer integration times as the ratio of the photon shot noise to the measured signal increases. In contrast to the fit error decreasing with integration time, the NO_2 retrieval yields the same average NO_2 DSCDs for different integration times."

l. 115-116 'Consequently ... to resolve specific traffic event' -> Please break this sentence in two for the sake of readability

Done.

l. 160 For the sake of readability, I suggest to be more explicit with the geometric approximation of the AMF at 20° i.e. to write 2.92.

Agreed and added to the text.

Figure 4 is important and should be improved. The y axis of panels A and B should be zoomed to better see the variations and mean values. Panels E and F are redundant, the authors could only show one of them (leading to larger remaining subplots and a clearer figure).

Thanks for this suggestion. We adapted the plot and text accordingly.

l. 199 'These emission standards' -> 'the emission standards of trucks' (would be clearer for the reader)

Done.