

Interactive comment on “Estimating real driving emissions from MAX-DOAS measurements at the A60 motorway near Mainz, Germany” by Bianca Lauster et al.

Anonymous Referee #2

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The manuscript present a new approach to derive average vehicle NO_x emissions on a motorway using passive MAX-DOAS. This method is a further adaptation / modification of emission estimates like performed with car MAX-DOAS [e.g. Ibrahim et al. 2010] or stationary MAX-DOAS for volcanoes [Galle et al. 2010]. While the basic measurement principle is similar, the setup was here adapted to the task of vehicle emission measurements. The applied method are well described and clear. They have the potential to be established for general vehicle emission monitoring of whole fleets. The topic fits well in the scope of AMT. There are two major weak points. First, several parameters which influence the measurement are not well considered. They lead to a further increase of the derived emission factor error, which is already quite large with 4.3+/-

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2.5×10^{19} molec/(ms). Second, the calculation of the expected NO_x emissions are incorrect and underestimated, which make the manuscript and likely the measurement principle disputable. This does not mean that a significant higher emission is derived than expected, but a more realistic expected emission would strengthen the value of the manuscript. The manuscript should first be corrected before publication.

Mayor points.

Chapter 3.4: Expected traffic emissions The EURO emission standard is a limit which is based on a lab test cycle on a chassis dynamometer (NEFZ, and now WLTP) and only needs to be fulfilled on average over the whole test cycle. The emissions can be for some driving situations higher and for other lower. Especially on a motorway where the engine load is high, emissions are typically higher than the average especially for passenger cars (e.g. HBEFA data base, Lashkina and Lashkin 2016, Athanasios et al. 2019, for trucks e.g. TNO 2016). Second, It is expected and well known, that real driving emissions (RDE) will be higher as the driving and surrounding properties in the test cycle are not realistic (like also, mentioned in the manuscript l. 26.). For trucks this is also limited since EURO VI with a RDE factor of 1.5. This mean that EURO VI trucks are allowed to emit on average $1.5 \times 460 \text{mg/kWh} = 690 \text{mg/kWh}$ in RDE (not the applied 460mg/kWh). For passenger cars the RDE confirmation factor is 2.1 since EURO-6d-Temp. RDE are thus for these diesel cars $2.1 \times 80 \text{mg/km} = 168 \text{mg/km}$. For older vehicles RDE is not tested and thus an emission confirmation with a confirmation factor is not defined. In conclusion this mean that the emission standard (Table 1) are not the expected RDE even if the vehicles confirm to the emission limit. The error become obvious as the expected weighted emission limit of the vehicle fleet (l. 213) is with 116mg/km below the RDE limit newest EURO 6d-temp diesel passenger cars need to confirm (168mg/km). Third, there are engine situations where significant higher emissions are allowed like cold start. If directly comparing measured emissions with calculated emissions, it should be excluded that these driving situations could contribute. Else they need to be considered.

There are different ways to handle the comparison more correctly: a) The expected emissions are modeled using the vehicle fleet, number, driving property at the measurement site and emission RDE data from HBEFA data base. This can also be made if it is expected that all vehicles confirm to the legislation or if included known RDE emission values. The expected emissions will increase in comparison to the authors calculation. That does not mean that they are than in agreement with the measurement, but this would allow a comparison between expected and measured average emissions.

b) The derived total emission is compared to the average emission limit on the chassis dynamometer (like currently done in the manuscript), however than a direct relation of how much the derived emissions are higher needs to be avoided. It must be clearly stated that the calculated emissions do not represent the expected emissions on the motorway, which is higher even if the vehicles confirm to the legislation. The comparison just gives the reader a relation between the numbers. In general the manuscript would than focus more on the derived total emission and less on the comparison.

c) The calculated expected emissions are at least more realistic. That mean that emission factors for motorways need to be used. Additional the RDE conformity factor need to be applied, which however only exist for newer EURO 6 / VI. How to deal with older cars is thus difficult. Additionally some estimated emissions of the trains need to be considered. Even if they are not clearly seen in the DSCD's (like mentioned in I. 130), they are still included in these data.

l. 209: The number of total traveled distance of trucks may not represent the real truck composition on the motorway. Especially on the motorway typically more foreign trucks are present than on average on the road. A more realistic number can be found from the toll collect system (https://www.bag.bund.de/DE/Navigation/Verkehrsaufgaben/Statistik/Mautstatistik/mautstatistik_node.html)

Chapter 2.2: Deriving DSCD's A spectral fit is missing in the appendix.

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l. 110: The given NO₂ DSCD error of 0.006×10^{16} molec cm⁻² does not agree to the given RMS values. Please provide the correct NO₂ DSCD errors of the spectral analysis. The error for the average DSCD is not reducing with Gaussian error propagation. Also in l. 125 / 126 the given error seem to be calculated with gaussian error propagation of the mean which is not valid. Systematic measurement errors do not behave like a statistical standard error.

The typical averaging (2s) is very short with resulting noisy NO₂ DSCD's. The authors derive an average emission factor over a longer time period. It is explained that there is no significant difference between the different time resolutions (l. 113). It is not clear which difference the authors mean here. The one for the example in Fig. 3? Or the difference for the whole SCD_traffic? Even if the difference is small, I do not understand the argument in l. 116 "to resolve specific traffic events", as non of these events are analyzed in the manuscript. If analyzing averaged spectral data (16 s or longer) the section 2.4 and Fig. 3 can be shorten.

The influence of clouds is not clear through the manuscript. Clouds seem to have a large influence on the results. It is not clear if both MAX-DOAS point at 90° at the same location, why a difference in DSCD is observed? Both should see the same cloud and thus same variation of DSCD. From Fig. 2 it looks like a systematic offset for the "East side" instrument is observed. If such small variations already cause such large difference between the instrument (east side instrument measure higher DSCD), how can you exclude that this is not the case when the instrument measure at different locations at 20°? From Fig. 2 only the measurement situation without clouds are used for the reference. The argument in l. 132. that clouds have only a small influence is not clear as Fig. A3 show the influence also for the emission measurement. With the argument from Fig. 2 (both instrument at west side) also only data without clouds should be used for the emission analysis (like Fig. A3 instead data from Fig. 4).

Are the "West side" and "East side" instrument at the same height? If not, what would be the influence on the NO₂ DSCD if they are not at the same height? Could this cause

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some bias in the DSCD_traffic.

Chapter 2.2: Estimation of real driving emissions

I. 168: The vehicles emit also directly NO₂. The share is for diesel engines (the main NO_x emitters) rather high with 30%.

I. 170: Specify that “the share of NO₂ in total NO_x” need to be known “at the measurement location”.

I. 174: The conversion of 2/3 of NO to NO₂ is estimated to 4 minutes. This conversion needs O₃. As NO emissions are very high at the emissions source, O₃ is completely titrated, and thus NO can not further react to NO₂. Even if the background conc. (I. 180) is at 42-44ppb, it will be zero at the motorway (like typical on high traffic roads). The further reaction requires dilution with O₃ rich air. Is this considered in the CAABA model? How this would effect the result?

I. 178: The estimated ratio from CAABA is 0.7 +/-0.4. It is not clear which solar radiation data are used for this calculation. The same is the case for the steady state conversion factor in I. 188.

Minor points:

Abstract: The whole approach is based on the conversion of NO to NO₂, which depends on Ozone. It is important to mention this in the abstract.

I.7: “...concentration over the lowermost 2 to 3km.” But what is the most relevant height in such a study. The plume will not be uplifted to several 100m, but will be rather below 100m if you are so close to the highway. So this statement is confusing. What is the expected height of the plume at the “East side” location?

Fig. 1: Following on the plume hight → How the area of highest sensitivity is calculated marked in Fig. 1?

I. 24: Since EURO 6c, the WLTP test cycle is the new test standard. For EURO VI the

WHSC.

I. 30: What do you mean with “need to convert NO to NO₂” which sounds like a problem? The measurement systems observe NO_x, how the instruments measure internally NO_x is not relevant for the derived emission data.

I. 31 & I. 34: The statement “Furthermore, this approach is dependent on the exact position of the emission source and the inlet of the measuring instrument.” and “Both approaches are able to resolve the emission of individual vehicles but are depending on the wind field and the position of the exhaust pipe with respect to the measuring instrument.” is not correct. Remote sensing and plume chasing observe ratios of gases e.g. NO_x/CO₂ and derive from this the emission factor. The dilution between emission source and inlet is not relevant for the emission value. It may have an effect if a sufficient signal is captured at all, but not for the value itself. It needs to be correct that these systems directly observe RDE of individual vehicles and measurement position and wind field is not relevant.

I. 33: The statement “However, these require an estimate of the amount of primary NO₂ in the exhaust.” is only valid for older remote sensing systems, as newer directly measure also NO₂ and NO. Additionally the direct NO₂ emission is small in relation to NO, thus this error is not so large. An other reason is valid why remote sensing has large errors: The snap shot emission measurement at very specific driving conditions where these systems work are not representative for the average emissions of an individual vehicle and also not necessary on average over many measurements as many driving conditions are not covered (e.g. motorway). That would be the motivation to derive fleet average emission factors and compare it with expected emissions from models.

The advantage of the described method in this manuscript over remote sensing and plume chasing is that it derives the average emission directly, where the other techniques would require a large data set.

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I. 57: If the authors state “measure the NO₂ emissions of vehicles” this would mean the direct NO₂ emissions, not including NO afterwards converted to NO₂. But the manuscript focus on NO_x emissions, derived from NO₂ DSCD.

I. 58: The background NO₂ DSCD subtraction is one of the main new methods applied in the manuscript. It should thus also be described in more detail in the method description.

I. 79: The wind measurement was performed upwind, but the important wind speed of the plume is downwind. Can this be different due to shading of trees etc.? What would be the estimated error?

Chapter 2.1: Does not include the measurement date and how many measurements are performed. It looks like there was only few hours of measurements. What were the conditions during this day? Are they representative. From a statistical point of view this is a quite small data set.

I. 151: The wind speed is only measured at ground level. But the landscape includes trees and hills. The wind speed may not represent the true speed of the plume. Can a better wind speed be estimated from the time shift of “West side” to “East side” of the NO₂ data (expecting that these variations are also at plume height)? An additional error for the wind at plume height should be included. What would be the influence if the wind velocity on the motorway (between the trees) is lower? Is the average wind speed derived over all wind speed data points or only in these periods when you have valid DSCD_traffic? This is even more relevant when analyzing data from Fig. A3.

I. 285: Here it is stated that Fig. 2 show CI, but it show NO₂ DSCD.

I. 227: Include the applied molar mass (46,01 gÅmol⁻¹).

I. 244: The sentence “trucks only account for a small amount of the total traffic volume”, is confusing, as they cause a large portion of the total NO_x emissions.

Fig. 4: A description is missing why traffic number is only shown for few times. What is

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with the gaps?

Fig A1: Include an explanation why CI is different for both instrument even if they point both at 90° at the same location.

A3, Fig A4 and A5: The difference between the two plots is only the averaging of the data over 12min instead of 2s. The explanation why there is no correlation in A4 is hidden in I. 300. As wind speed measurements are not at the location of the plume, the correlation at the high time resolution seem to be prone for errors and confusing. I suggest directly to show only averaged data (A5), where a small time shift has only a minor effect.

References:

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