

We gratefully thank all reviewers for the careful reading and valuable comments. Below we provide our point-by-point responses to the reviewers' comments. In the following context, raised comments/suggestions are marked in **black**, responses are presented in **red**, and changes to the manuscript/supplement information are indicated in **blue**.

Reply to Anonymous referee #1

The paper presented by Leinonen et al. summarizes different methods to determine dilution ratios and emission factors from vehicle chase studies. The study location was a well-suited test road for wintertime investigation. Mostly, no other vehicle intervened the setup. The study appears to be scientifically sound and adds valuable data to the literature of exhaust chasing. It should be accepted for publication after some revision.

We thank the referee for these positive comments towards our manuscript.

The study would have been much more useful and credible if the test vehicle would have been equipped with a PN-PEMS. This would have proven if the order of magnitude for PN of the Diesel vehicle (assumed with DPF) is correct. An average of 5×10^{12} #/km at -11C to -23C could have easily been confirmed, or disproven. The same question arises for the gasoline vehicle: 4×10^{11} – 1×10^{12} #/km (Skoda-24C to -26C). How does this compare to PN PEMS during the same drive?

The referee is correct that PEMS could have been used to compare the emission factors of vehicles. In earlier study, such as (Karjalainen et al., 2014) have shown that the emissions of particles measured from the chase measurements are in line with the dynamometer results. Based on those earlier results, we believe that the emission factors calculated in this study are in line with the results that would have been achieved with the PN-PEMS system. In the future, the comparison between PN-PEMS and chase measurement methods would be interesting to conduct.

The results for the Skoda-2 -24C/-26C selecting downhill driving are puzzling: Why is the EF not substantially lower (downhill Figure 7) if compared to the overall trip? Rather the opposite occurs comparing Skoda -24C Figure 5 (about 4×10^{11} #/km) with downhill Figure 7 (about 6×10^{11} #/km)?

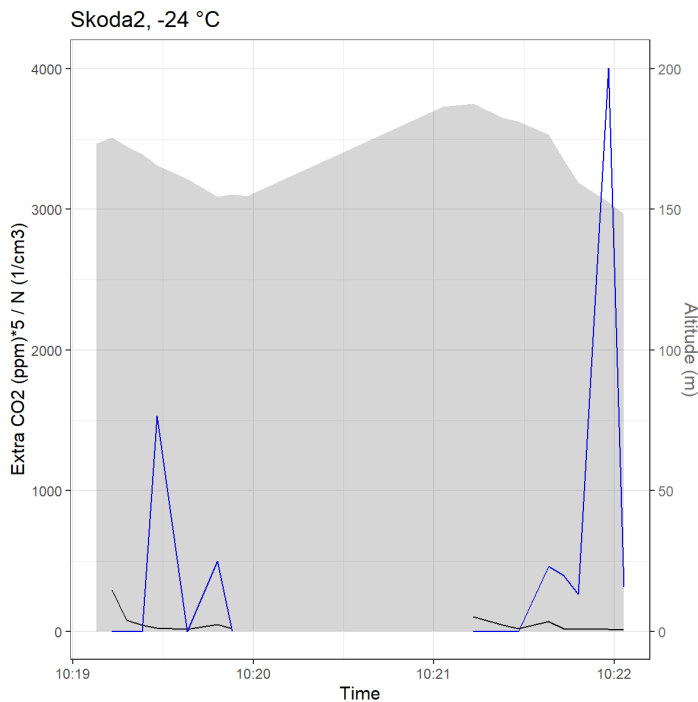


Figure 1. Time series of $CO2_{meas} - CO2_{bg}$ (black) and $N_{meas} - N_{bg}$ (blue) for the downhill section for Skoda2, -24 C round. Altitude profile is shown with gray background ribbon. Note that the data between 10:20:00 and 10:21:00 is not used in the calculation of downhill EF:s.

For the high EF of Skoda2 -24 C round, Fig. 1 shows the time series $CO2_{meas} - CO2_{bg}$ and $N_{meas} - N_{bg}$ for the downhill section. We also checked the video recording for the downhill sections. For the first downhill, between 10:19 and 10:20, there was no clear braking period. Hence the sources of particles measured from that time period were not clearly identified. For the second downhill, after 10:21, there were multiple brakings starting from 10:21:25 and 10:21:55, which is close to the increase of number concentrations observed. However, the chemical composition of those particles was not studied in detail for this study. Those concentrations shown in figure 1 explain the high EF (in #/km) shown for the downhill section in Figure 7.

What were the chasing distances and can could the NWD, or MARS be applied in open traffic, where this short distance would be potentially unsafe?

When the chased vehicle was moving, the chasing distance was between 5 and 10 meters. So short distances won't be safe in open traffic, especially under winter conditions, if the chased vehicle is driven by a random driver. Chasing methods, however, could be used for other (longer) distances as well, as shown e.g. in Olin et al. (2023).

We added the information about estimated distance between vehicles into section 2.3.

“Based on our estimation, the chasing distance was between 5 and 10 meters when the chased vehicle was moving.”

The authors are measuring total PN, and no size distributions. Therefore, it is left entirely open if nucleation particles would have occurred at the low temperatures. If nucleation would have occurred, it would question the method of NWD, particle formation could not have been completed in the exhaust plume.

Maybe, for a next study a setup w and w/o thermal treatment of the PN will be included?

It is difficult to estimate whether the nucleation and condensation processes are complete prior to the sampling. The plume residence time was at minimum about 0.5 s. The focus of the article is rather on different computational methods to define the dilution and emission factor, not to cover the fine details of aerosol particles and properties. More thorough aerosol characterization study for sure would require the use of thermal treatment and measurement of different particle sizes.

Other editorial:

Line 23: please clarify what is meant with “but the regulation even for new vehicles is still under development and the new regulations do not completely cover the existing fleet”? If a future regulation is meant, it is normal. It has to be developed and can only apply to vehicles coming into production at that date.

We agree with the reviewer that the meaning of the sentence was not clear. The sentence was modified to “Vehicle emissions are regulated in legislation but the regulation for new vehicles is under constant development (type approval, periodical technical inspection (PTI), and real driving emissions (RDE)). The new and upcoming regulations are effective only for the vehicles produced after the regulation have become effective.”

Line 28-30: “The limits for PN only consider nonvolatile particles, and the particle mass (PM) formed from the precursor gases via nucleation and condensation as the exhaust gas dilutes and cools upon exiting the tailpipe is mostly neglected. The amount of particle matter (both in terms of PN and PM) formed this way can be considerable.”

These sentences are misleading: With the elimination of fuel sulfur the occurrence of volatile PN formed upon cooling and nucleation has been decreased. The hydrocarbons potentially nucleating are measured and regulated via gaseous HC requirements. Also, potential health effects are more likely to be related to solid PN (i.e. more long-lived, and not dissolved and diluted in the alveoli).

Therefore, until data is available, or a strong reference is added, the sense of the statement “neglected” should be revised.

We clarified the message based on the comments from the reviewer. The updated version of the sentences is as follows:

“The regulation limits for PN mostly considers nonvolatile particles. The particle mass (PM) formed from the precursor gases via nucleation and condensation as the exhaust gas dilutes and cools upon exiting the tailpipe is not fully considered PN measurements, however the regulation for gaseous hydrocarbons limits the amount of precursor gases produced by the vehicle. The amount of

secondary particle matter (both in terms of PN and PM) formed from precursor gases can be considerable. However, the amount of secondary PM has decreased in 21st century as the fuel does not contain as much sulfur as before.”

Tab 1: Add if particle filter was installed. Euro-5 should have DPF, for gasoline vehicles not clear.

We added a column to Table 1 indicating filtering technologies in each vehicle. Particle filter was in every vehicle, except Ford Focus. An updated version of Table 1 is presented below.

Table 1: Information on the studied vehicles. DPF = diesel particle filter, GPF = gasoline particle filter, MHEV = mild hybrid electric vehicle, SCR = selective catalytic reduction

Car	Fuel	Filter	Registration year	Engine displacement (l)	Emission class	Odometer reading (km)	Number of drives
Audi A6	Diesel	DPF	2008	3.0	Euro 5	236,000	6
Seat Alhambra	Diesel	DPF + SCR	2012	2.0	Euro 5	169,000	6
VW Transporter	Diesel	DPF + SCR	2019	2.0	Euro 6	36,000	4
Ford Focus	Gasoline		2018	1.0	Euro 6	78,000	5
Skoda Octavia 1.0	Gasoline (MHEV)	GPF	2020	1.0	Euro 6	1,000	6
Skoda Octavia 2.0	Gasoline	GPF	2019	2.0	Euro 6	21,000	6

Line 377: “NWD and MARS Method would be extendable to non-exhaust emissions”

This statement should be further explained. How to differentiate from Exhaust PM? How to differentiate tire wear, and brake wear? Applicable to electric vehicles only?

We agree with the reviewer that the distinction of non-exhaust emissions from exhaust emissions might not be easy, and that it is probably easiest to study non exhaust emissions from electric vehicles. The methods presented did not differentiate any emissions from each other, as the methods focus on the dilution of emissions. The differentiation, if required, needs to be done using some other data/methodology. Additionally, the differences in dilution of emissions, i.e. exhaust vs. non-exhaust and also different sources of non-exhaust, needed to be considered.

We added the following text after the sentence:

“For NWD, the method is based on the estimated slope κ of the vehicle. For example, for tire emissions, if the emission from the tires is C_{raw} and mass exhaust flow rate of the emission is Q ,

then $EF = C_{raw} * Q$. On the other hand, it was assumed that $DR = \kappa * v/Q$. Then $C_{raw} = C_{meas} * DR = C_{meas} * \kappa * v/Q$. For EF, we get that $EF = C_{raw} * Q = C_{meas} * \kappa * v$. Hence, an explicit value of mass exhaust flow rate Q is not needed to calculate EF of non-exhaust emission. The κ value can be estimated from the other vehicle with similar estimated dilution of emissions, or in case of hybrid vehicle, the κ can be determined during the time when the combustion engine is running. For MARS the basic idea is that from the test dataset of measurements, the dilution ratio of emissions could be estimated in different driving situations. Then in the new dataset, the DR is estimated based on splines estimated from the test dataset.

In both methods, the emission factor of the non-exhaust emission can be determined during the times when the vehicle is running with electric engine only. For the non-exhaust emissions, some correcting coefficient for the dilution ratio might be needed.”

References

- Karjalainen, P., Pirjola, L., Heikkilä, J., Lähde, T., Tzamkiozis, T., Ntziachristos, L., Keskinen, J., and Rönkkö, T.: Exhaust particles of modern gasoline vehicles: A laboratory and an on-road study, *Atmos. Environ.*, 97, 262–270, <https://doi.org/10.1016/j.atmosenv.2014.08.025>, 2014.
- Olin, M., Oikarinen, H., Marjanen, P., Mikkonen, S., and Karjalainen, P.: High Particle Number Emissions Determined with Robust Regression Plume Analysis (RRPA) from Hundreds of Vehicle Chases, *Environ. Sci. Technol.*, <https://doi.org/10.1021/ACS.EST.2C08198>, 2023.