Atmospheric Measurement Techniques Discussions

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Comment on amt-2021-43

Anonymous Referee #3

Referee comment on "Evolution under dark conditions of particles from old and modern diesel vehicles in a new environmental chamber characterized with fresh exhaust emissions" by Boris Vansevenant et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-43-RC2, 2021

The authors investigate the emissions of 6 diesel passenger cars from Euro 3 to Euro 6 under 2 different driving cycles. They determine emission factors of particle number, particle mass, black carbon, NMHC and IVOC. They present a method to correct the evolution of particle mass concentration for dilution and wall loss of particles in the chamber. They claim that under dark conditions particle mass concentration (PM) increases over time as a function of initial particle number or particle surface concentration. Below a particle number concentration of (8-9)E4 cm-3 the particle number concentration (PN) increases, while above it decreases.

The experiments and the data analysis are well described. However, there is much speculation regarding the interpretation of the data and a lack of proof of their claims.

The authors interpret their observation of a sustained increase of particle mass and number concentrations by condensation and nucleation, respectively. However, to have continuous condensation or nucleation a constant production of condensable or nucleating vapors need to occur. Otherwise, vapors will rapidly condense on particles and on the wall and condensation or nucleation stops. In their small chamber the lifetime of such vapors will be less than ten minutes. Therefore, the claim that PM increases over many hours could be due to condensation of IVOCs is not plausible. The authors may also check if the saturation vapor pressure of IVOCs is low enough to partition to the particle phase at the particle mass concentrations of their experiments.

There are also some issues regarding the PN increase and the particle number concentration threshold. This observation depends on the available instrumentation. The smallest particles they can measure is 14 nm. They do not present the particle size distributions of their different experiments and therefore it is not clear in which cases a nucleation mode is formed. This would happen very fast during injection and there is not a steady nucleation going on as explained above. If an increase of PN occurs, this happens

because of coagulation of nucleation mode particles, which produces particles of larger size that become measurable in their SMPS. This is not an increase of PN but the fact, that they did not measure the particles below 14 nm. Thus, the threshold and the time of increase depend on this measurement parameter. The fact, that a growing nucleation mode influences the PN at larger sizes is not a new finding. One should always be aware of it when analyzing such data.

At high PN concentrations when "coagulation prevails" the Figures 11c,d,e show first the expected decrease of PN due to coagulation, which is however after 1-2-h followed by an increase of PN. Do the authors have an explanation for this observation? Are these wall loss corrected PN data? If yes, this would mean the newly developed algorithm could eventually not correctly compensate for the losses.

The authors need to provide more evidence why PM should grow over hours. Condensable vapors are lost to the walls and it is not plausible how an oversaturation is maintained over hours without production. Thus, the wall-loss correction method could induce such an artifact. Although Figure 5 shows a good correlation between the mean loss coefficients and k_{BC} it is also obvious, but not mentioned, that there is a large off-set. Why does this occur and how does this affect the PM correction? The authors need to show that the PM growth is a real phenomenon and not an artifact from their correction method. By calculating corrected PM do they use the average loss rate or the size dependent loss rates?

Line 27: "Condensation is 4 times faster when the available particle surface if multiplied by 3". How did the authors calculate this? Is this for a certain particle size?

Overall, I do not see much scientific progress in this paper.