

Interactive comment on “Particle and gaseous emissions from individual diesel and CNG buses” by Å. M. Hallquist et al.

Anonymous Referee #1

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Johnsson et al. measured particle and NOX emission factors (EF) under conditions of ambient dilution from 35 individual busses taken from the actual bus fleet. Three types of engines were studied, including diesel EURO III-IV busses with and without diesel particle filter, and busses fuelled by compressed natural gas (CNG). Among the findings were the highest observed particle number EF from CNG busses, which however emitted the smallest particles mass compared to diesel busses. Accordingly, the particle mode was much smaller for the CNG busses relative to the diesel fuelled ones. The manuscript is scientifically sound, well written and the results are nicely transparent.

Specific comments

Page 2, line 24 (Page 27739, line 19). This could be further elaborated. Following dilution, semivolatile organic compounds (SVOC) partition from the condensed phase to the gas phase until equilibrium is achieved. Other secondary particles originating from vehicular emissions are formed during atmospheric oxidation of semivolatile and volatile organic compounds (Robinson et al 2007. Science 315, 1259). Contrary to condensates, these are formed in a time scale from hours to days, i.e. on a regional scale.

Authors' comments and action: This is true. We have now added the following (Page 2, line 26-30) “Additionally, traffic contributes to the formation of secondary organic aerosols (SOA); however, the magnitude of this contribution is very uncertain (Robinson et al., 2007). This is a chemically-induced particle formation (time scales of hours to days) which is very important on a regional and global scale” (Hallquist et al, 2009).

Page 10, line 13-16 (Page 27749, line 1-4): Please clarify this paragraph. For comparable particle mass, a shift from larger to smaller particles would increase the surface area. The smaller particles from CNG busses most probably reflects their chemical composition, e.g. diesel particles with no after-treatment have a soot core within a size range exceeding the 10-25 nm mode observed from the CNG busses. The small particles from CNG busses are expected to coagulate on a short timescale.

Authors' comments: We have now clarified this sentence

Authors' action: This sentence now reads: “The lack of larger particles in the emissions from CNG-fuelled buses decreases the available surface area and hence favouring nucleation over adsorption/condensation of supersaturated vapours. This enhanced nucleation is one reason for the larger average particle number emissions for the tested CNG buses (Kumar et al., 2010).”

Table 2, page 15 (Table 2, page 27760). Modelled EF for PN is lowest for CNG and highest for diesel busses, whereas the opposite is found in this study. Comment on this reverse relationship.

Authors' action: The reverse relationship is now commented on in the text (Page 27748; Page 10, line 3-5). The following has been added “A reason for this can be that the particle number emissions that the HBEFA model is based on often follow the PMP protocol, involving heating the particle sample to 300°C, and the CNG particles are suggested to be volatile (Jayaratne et al., 2012).”

Table 3, page 16 (Table 3, page 27761-27762). Comment on the high variation observed for apparently similar vehicles, e.g. busses EURO III(SCR, DPF) #1 and #2 EFs differ by a factor 10 for PN, acc and PN, const, and even more for PM,acc. The same PN EF for EURO III Busses # 3-8 differ by a factor of 400 and 35, respectively. Use specific examples.

Authors' comments: The data presented is a reflection of the true variation in an in-use regional bus fleet, where the variations found between similar buses can be due to engine specifics, maintenance or malfunction.

Authors' action: Have added the following into section 3.2 (page 27748; Page 10, line 28-31) "The data presented in this study (Table 3) is a reflection of the true variation in an in-use regional bus fleet, where the variation found between similar buses (e.g. regarding fuel type and after-treatment technology) within the same Euro class can be due to engine specifics, maintenance and malfunction."

A general discussion about variation in emissions factors would be appropriate, also including table 4. How many repetitions were the obtained EF based on, and could you provide standard deviations in the tables. Considering the large variation, please discuss the need for repetitions on different days as well.

Authors' comments and actions: The buses were tested at least three times but often more repetitions were performed. This information has now been added to Page 5, line 8-9. In the Tables the statistical 95% confidence intervals are included and we do not think that adding standard deviations will add more information. In Table 4, the tested vehicles have been divided into three groups (CNG, Diesel buses with and without DPF) so there is a large variation in engine specifics, Euro class and after treatment technology within each group that explain some of the variations. The variation in the ambient conditions has been reduced by using a thermodenuder as described in the paper. However, maintenance is also an important factor that may differ and it would be very interesting to follow how one bus behave e.g. throughout a year but that is beyond the scope of this paper. Instead, the major aim of this paper was to study, present and understand the range of EFs within an in-use fleet (35 different vehicles).

Technical corrections Page 12, line 1-3 (Page 27750, line 22-24). Rewrite sentence for clarity.

Authors' actions: Sentence has been changed to: "The average EF_{CO} for the diesel buses with DPF tested in this study were $9 \text{ g (kg fuel)}^{-1}$ (17 buses in total). Here values below one times the standard of the noise were assigned to $6 \text{ g (kg fuel)}^{-1}$."

Interactive comment on “Particle and gaseous emissions from individual diesel and CNG buses” by Å. M. Hallquist et al.

Anonymous Referee #2

22 January 2013

General comments:

The authors characterized the particle and gaseous emissions from 28 individual diesel fuelled and 7 compressed natural gas (CNG)-fuelled buses under real-world dilution when the buses were driving in an accelerating mode or in a constant speed mode. The buses used different after-treatment systems. The results showed that the particle number emission factors for CNG-fuelled buses were higher than for the diesel buses, just opposite to the mass emissions factors. Differences were also found in the particle number size distributions indicating that the emitted particles from the CNG buses were smaller. From the climatic and health point of view this issue is topical, interesting and important. The manuscript is mostly well-written; however, I raise some critical questions which should be addressed.

Specific comments:

1. Introduction: All nucleation mode particles do not need to be secondary particles. For example, Rönkkö et al (EST, 2007) found that a Euro IV heavy-duty diesel vehicle with EGR emits nucleation mode particles that have a nonvolatile core formed before the dilution process.

Authors’ comment: Our statement is not that all nucleation particles are secondary. What we say is “Particles measured in close vicinity of the emission source are primary, i.e. emitted as particles from the tailpipe, or secondary, i.e. formed during the expansion and cooling of the hot exhaust gases.”

2. Section 2 should be rewritten throughout. Experimental method is poorly described, and it is hard to understand how the experiments were performed; when and where (other traffic?), how many repetitions, and under which conditions. Did you measure background concentrations and are they subtracted.

Authors’ comments: Yes background concentrations were measured and $EF_{PN/PM}$ were derived by simultaneous measurements of CO_2 and particle number/mass concentrations from the same air volume in the bus plume and compared to the concentration before the passage. Also the gaseous concentrations were compared to background concentrations; here the increase in absorption due to a bus passage was measured. Line 1 section 2 now reads “In this study particle and gaseous emissions from individual vehicles were determined by measuring the concentration change in the diluted exhaust plume compared to the concentrations before the passage and relative to the corresponding change in CO_2 concentration”

Authors’ action: In section 2 we have now added information about number of repetitions for the tested buses (Page 5 line 8-9) “Each bus was tested at least three times, but often more repetitions were performed.” and conditions (Page 5, Line 4-5) “The measurements were performed at five different locations in connection to the bus depots with limited influence from other traffic.”

No information was given about the engine and driving parameters (bus type, engine type, engine speed, gear, torque etc.), please, add into Table 1. The fuel sulphur content of diesel fuel should also be mentioned.

Authors' comments and actions: The diesel fuel used as well as the sulphur content of the fuel has been added to Table 1. The measurements were conducted during acceleration from stand still and before changing gear, typically 2-4 s into the acceleration. Engine speed torque etc was not measured and therefore not included in Table 1.

Description of CO₂ measurements from Section 2.1. (Particle sampling) could be moved to Section 2.2. (Gas sampling); and description of emission factors for gases from Section 2.2. to Section 2.3.(Calculation of emission factors).

Authors' actions: We have moved the description of EF for gases to section 2.3. However, the description of the CO₂ measurements has been left unchanged as this is part of the particle emission measurements.

3. The gaseous NO, HC and CO were measured by a remote sensing device (AccuScan RSD 3000). How close to the particle measurements did the transmitter and the receiver locate. I am not familiar with the system; I expect the method is fine for rather low ambient concentrations. Therefore, I am wondering the high concentrations of NO, NO_x, CO and CO₂ in the calibration gas, the mixing ratios sound to be valid for raw exhaust measurements. Could you, please, explain more about this issue.

Authors' comments: It should be noted that CO₂ is measured both by the RSD instrument and in the air sampled for particle emission factor determination. This is clearly stated in the paper and shown in Figure 1. The inlet of the tubing used for the particle sampling (and to the LI-COR CO₂ instrument) was placed as close to the RSD beam as possible. However, an optimal placement of the tubing had to be determined for each bus type since the placement of the exhaust pipe can differ between different bus models. This means that the distance between the RSD beam and inlet of the tubing varies from approximately 0 meter to about 1 meter. Two calibration procedures are performed on RSD instruments. The first is performed by the manufacturer in the lab in order to establish the sensitivity of each detector/filter combination to the pollutant gas and to ensure linearity of the instrument. The second calibration is performed in the field by the operator. The gas mixture used for this calibration is meant to be representative for a high emitting vehicle.

Authors' actions: We prefer to not add information as we think the information provided is sufficient (see e.g. Figure 1 and the reference given for the RSD instrument).

4. Particle mass concentration and emission factors EF(PM) were calculated from the EEPS measurements by assuming spherical particles with unit density. No mass monitor was installed in the measurements setup. I am somewhat skeptical about these values because soot particles are agglomerates. Is it necessary to include EF(PM) in this paper? At least, uncertainty of the results should be estimated and discussed. Table 4 compares the EF(PM) with other studies. By taking into account that this study concern PM_{0.56} while the others mostly give PM₁₀ and PM_{2.5}, the paragraph (p.27748, lines 7-) needs more precise discussion.

Authors' comments and action: Recently we have conducted on-board measurements on a Euro V diesel bus using a soot-sensor (AVL 483 Micro Soot Sensor) measuring particle mass and an EEPS. The results from these measurements showed a good agreement between the two instruments when using the same assumptions (i.e. spherical particles and unit density) as in this study for determining PM from the EEPS measurements.

For combustion related traffic emissions most particles are below 560 nm as is shown in Fig. 4 hence PM_{0.56} and PM₁₀ and PM_{2.5} are comparable. However, road measurements of PM₁₀ can include re-suspension and hence these values can be higher. A section about this has been added to section 3.2 (Page 10, line 13-18).

5. The particle sample was led through a thermodenuder at 298 K. Rönkkö et al. (2007) measured 95% losses at 6 nm particles, 74% at 10 nm, and 28-40% at 30 nm for the temperature range 28-275 oC. Did you correct the losses in the thermodenuder?

Authors' comments and action: Yes corrections for particle losses in the thermodenuder have been performed. Have added the following sentence to section 2.1 (Page 5 Line: 22-23) "Size-dependent aerosol losses within the TD were accounted for".

6. The measured EFs (Table 3 and Fig.3) were compared to the modelled data (Table 2) from the HBEFA 3.1 model for a standard urban bus with a posted speed of 30 km/h and with stop and go traffic. The authors conclude that the modelled values are generally significantly lower than the measured values. What might be the reasons; these should be discussed. Comment also why the EF(PN) is smallest for a CNG bus, contrary to the results obtained in this work.

Authors' comments and action: Possible reasons for the lower modelled EFs compared to the measured ones as well as the reason for low modelled EF_{PN} for CNG buses are discussed. The following has been added (Page 9, Line 29-31 and Page 10, Line 1-5). "The modelled values are generally significantly lower than the measured values. A possible explanation for this can be different driving modes, acceleration versus route, including start and stops but also constant speed mode. As indicated by Table 3, EF_{PN/PM} was generally lower for constant speed mode compared to acceleration. Modelled EF_{PN} was the lowest for CNG buses and highest for diesel busses, whereas the opposite was found in this study. A reason for this can be that the particle number emissions that the HBEFA model is based on often follow the PMP protocol, involving heating the particle sample to 300°C, and the CNG particles are suggested to be volatile (Jayaratne et al., 2012)."

Technical comments:

7. The sentence "The shape of the CO₂ peak..." on p.27746 lines 14-17 should be clarified.

Authors' action: The sentence has been changed to: "The shape of the CO₂ peak is broader than the particle peak, which is due to the use of a small volume before the CO₂ analyser, extending the time available for the instrument to process the gas sample in order to prevent concentration peaks out of the instrument's measurement range."