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Particle emissions of Euro VI, EEV and retrofitted EEV city buses in real traffic☆

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ABSTRACT

Exhaust emissions from traffic significantly affect urban air quality. In this study, in-traffic emissions of diesel-fueled city buses meeting enhanced environmentally friendly vehicle (EEV) and Euro VI emission limits and the effects of retrofitting of EEV buses were studied on-road by chasing the buses with a mobile laboratory in the Helsinki region, Finland. The average emission factors of particle number (PN), particle mass (PM_1) and black carbon mass (BC) were $0.86 \cdot 10^{15}$ 1/kg_{fuel}, 0.20 g/kg_{fuel} and 0.10 g/kg_{fuel}, respectively, for EEV buses. For Euro VI buses, the emissions were below $0.5 \cdot 10^{15}$ 1/kg_{fuel} (PN), 0.07 g/kg_{fuel} (PM_1) and 0.02 g/kg_{fuel} (BC), and the exhaust plume concentrations of these pollutants were close to the background concentrations. The emission factors of PM_1 and BC of retrofitted EEV buses were at the level of Euro VI buses, but their particle number emissions varied significantly. On average, the EEV buses were observed to emit the largest amounts of nanocluster aerosol (NCA) (i.e., the particles with size between 1.3 and 3 nm). High NCA emissions were linked with high PN emissions. In general, results demonstrate that advanced exhaust aftertreatment systems reduce emissions of larger soot particles but not small nucleation mode particles in all cases.

1. Introduction

Outdoor air pollution, especially particulate matter smaller than 2.5 μm ($PM_{2.5}$), has been estimated to cause 3.3 million premature deaths per year worldwide (Lelieveld et al., 2015). Emissions from combustion sources, including residential energy use, traffic and power generation, are frequently the most important air pollution-related contributors to premature deaths in many countries, such as China, the USA and Japan (Lelieveld et al., 2015). Exhaust emissions from diesel vehicles are specifically harmful to human health and have been classified as carcinogenic by the World Health Organization (IARC 2012). In general, the increasing knowledge of the negative effects of air pollutants has led to active and continuous regulatory actions aiming to limit the pollutant emissions and to technological development in emission reduction techniques, especially in road traffic.

The bus exhaust emissions under real operation depend on several factors, such as the vehicle's technical condition, type of engine, exhaust aftertreatment (EAT) system, fuel and lubricating oil, as well as the driving and environmental conditions. The effects are seen, for instance, in the number of exhaust particles emitted, the particle size distributions, gas-phase emissions and the chemical composition of exhaust particles and potential contribution of emitted compounds to secondary particulate-matter formation (Kado et al., 2005; Jayaratne et al., 2008; Hallquist et al., 2013; Pirjola et al., 2016; Saarikoski et al., 2017; Watne et al., 2018). Buses, like other motor vehicles, have typically larger emissions during acceleration, e.g., from bus stops (Pirjola et al., 2016). Localized emissions may generate air pollution hot spots in certain parts of the bus route, which may contribute to exposure of the people associated with these hot spots.

Diesel bus emissions have diminished due to the implementation of novel engine and EAT technologies. Diesel particulate filters

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(DPFs) are typically ceramic wall-flow filters used in Euro VI diesel buses to remove soot particles. In the DPF, the exhaust gas is forced through a porous filter medium, resulting in very high filtration efficiencies for soot particles, even over 99.9%, also depending on the filter loading (Vaaraslahti et al., 2004; Bergmann et al., 2009; Herner et al., 2011). The trapped particulate material increases the pressure drop over the filter, necessitating active or passive filter regenerations. Oxidative EAT systems, such as diesel oxidation catalysts (DOCs), have been applied for some time in buses to reduce the organic fraction in exhaust particles and gaseous emissions, such as hydrocarbons and carbon monoxide (CO). Exhaust gas recirculation (EGR) or selective catalytic reduction (SCR) systems have been used to reduce nitrogen oxide (NO_x) emissions from diesel exhaust (Koebel et al., 2000) (e.g., in Euro VI buses). NO_x is reduced in an SCR using ammonia as the reductant. Ammonia is typically generated by hydrolysis of a urea solution injected into the hot exhaust gas upstream of the SCR. The SCR efficiency depends on various parameters, such as placement and operational parameters of urea injection, loading of catalysts and exhaust gas temperatures (Johnson, 2009).

As described above, current emission control strategies largely rely on exhaust aftertreatment. This strategy offers the possibility to reduce the emissions by retrofitting existing buses. Buses have a relatively long service life (12 years on average in Finland; see Autoalan tiedotuskeskus, 2019), which may make the retrofitting of existing buses a beneficial alternative to replacement with new ones (Li et al., 2015).

Some studies have been conducted on the effects of retrofitting on buses' emissions of gaseous and particulate pollutants. For instance, in Zhang and Zhu (2011) study, 11 buses were retrofitted with DOCs and crankcase filtration systems. In their study, the retrofitting resulted in an approximately 30% reduction of particle number emissions in particle sizes larger than 5 nm when the emissions were measured during idling conditions. Tartakovsky et al. (2015) retrofitted three buses of different emission classes (Euro II, Euro III, Euro IV) with DPFs. For the studied buses, they conducted 2000 h field durability tests followed by particle filtration efficiency tests. They reported filtration efficiencies of more than 90% for particle number emissions during the conditions tested. DPF's effectiveness in particle emission reduction has also been observed in recent studies by Fleischman et al. (2018) and Zhang et al. (2018); the average particle filtration efficiency reported by Fleischman et al. (2018) was 96% (a 12-month study of 18 buses retrofitted with DPFs) and 90% in the study conducted by Zhang et al. (2018) (one bus retrofitted with a DPF). Regarding the need to reduce NO_x emissions, Carslaw et al. (2015) retrofitted 96 DPF-equipped buses with DPF-SCR devices. For these vehicles, they performed real-world NO_x emission measurements on a street in one location with a slight (+1.9°) uphill gradient. Compared to similar DPF-equipped buses operating under the similar driving conditions, NO_2 and NO_x emissions of DPF-SCR equipped buses were 61% and 45% lower, respectively. Along with emission reduction, it should be noted that the retrofitting of buses with advanced EAT systems can have additional effects; for example, Fleischman et al. (2018) found that the retrofitting increased buses' fuel consumption by 0.6–1.8%.

In this study, exhaust particle emissions were measured from a group of city buses representing various emission levels. The goal was to evaluate the particle emission differences between the enhanced environmentally friendly vehicle (EEV), Euro VI and EEV buses retrofitted with new EAT systems. Measurements were conducted with a mobile laboratory by chasing the buses on-road during their normal operation. In addition to particulate mass, which is traditionally connected to the negative effects on human health and the environment, we estimated the particle emissions by measuring particle number concentration, recently found small sub-3 nm emission particles (called nanocluster aerosols, NCA, see Rönkkö et al., 2017), lung-deposited surface area (LDSA) concentration of particles and black carbon (BC). BC emissions have been connected to particles' effects on health and climate (e.g. Highwood and Kinnersley, 2006). This study was made to compare vehicle technologies' effects on these important quantities and to understand how these quantities are interlinked with each other.

2. Experimental

2.1. Routes and measurement routines

Measurements were conducted by chasing the buses during their normal scheduled operation with a mobile laboratory and measuring the exhaust plume with the instruments installed in the mobile laboratory (e.g. Rönkkö et al., 2017). The measurements took place over an approximately 2.9 km-long stage (~10 min of driving) of Helsinki region bus line 550 (Fig. 1). The location of the measurement route in the middle of the bus line ensured that the bus engines were warm during measurements. The measurement route included seven bus stops (named A-G, Fig. 1) where buses stopped regularly and a 1.7 km-long part, where only bus traffic was allowed (approximately between stops C and D). Stop F was in different locations for westbound (FW) and eastbound (FE) traffic. One measurement consisted of chasing a bus under normal operating conditions in one direction. The distance between the bus and the mobile laboratory was approximately 10 m during normal driving and reduced to approximately 2 m at bus stops. The measurements were conducted between morning and afternoon rush hours (between 10 a.m. and 4 p.m.) on four working days (11th, 12th, 15th and 16th February 2016). Measurement times were selected to reduce other vehicles' effects on the chase measurement procedure and pollutant concentrations. During the measurements, the outdoor temperature varied from −4.5 to +2.5 °C and relative humidity from 52.4 to 86.6%.

2.2. Buses

The study consisted of 32 individual measurements for the exhaust emitted by multiple types of diesel-fueled buses (Table 1). The most common bus type during measurements was the EEV-level bus, with exhaust aftertreatment consisting solely of an EGR (EEV corresponds to Euro V emission levels but with lower limits for hydrocarbons and smoke number). Three of the EEV buses had been retrofitted with a DPF and an SCR, manufactured by two manufacturers. Here, we call those buses Retrofit 1 and Retrofit 2. Retrofit 2 maintained the original EGR, which had been removed in Retrofit 1. Five Euro VI buses from two manufacturers were also included in the study. The exhaust aftertreatment systems of Scania's Euro VI buses consisted of EGR, DPF and SCR, and the exhaust aftertreatment systems used in Volvo's Euro VI buses consisted of DPF and SCR.

2.3. Mobile laboratory and instrumentation

The detailed description of the mobile laboratory and sampling lines is given in the Supplementary material. A weather station (200WX, Airmar Technology Corporation) on the mobile laboratory's roof was used to log outdoor temperature, pressure and humidity, and an integrated GPS receiver logged the mobile laboratory's location and speed. Carbon dioxide (CO_2) concentrations were measured with an infrared absorption-based instrument (Sidor, SICK AG) and NO_x concentrations with a chemiluminescence based analyzer (T201, Teledyne API). A more detailed description is

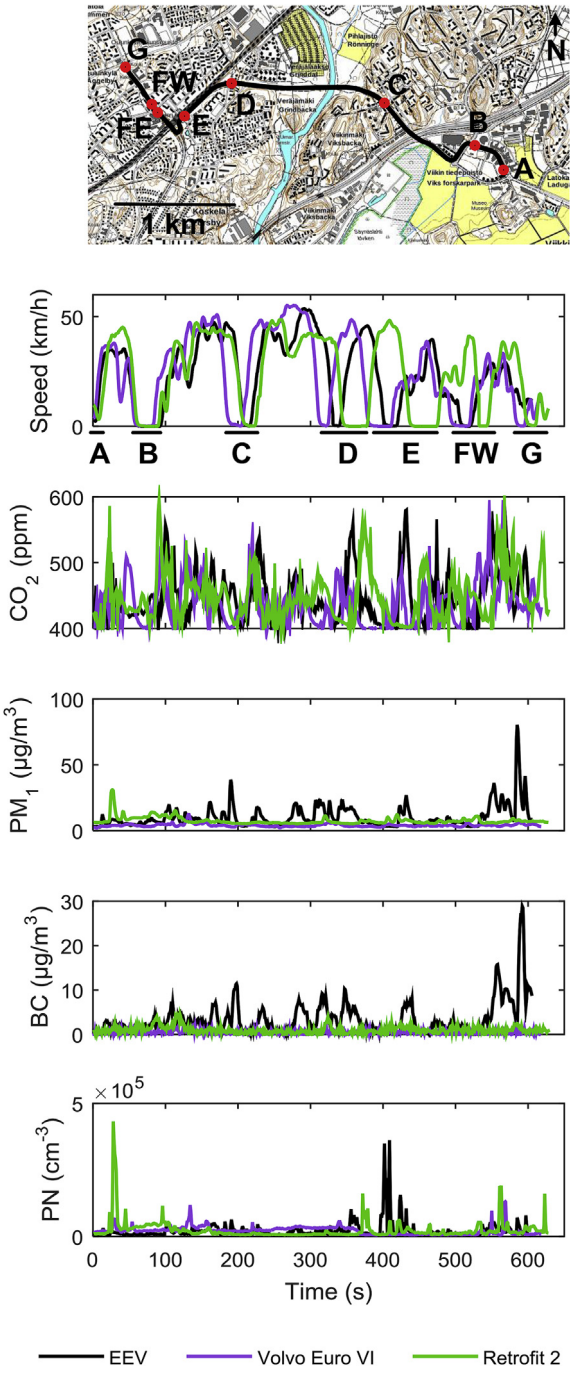

Fig. 1. Measurement route and time series of speed, CO_2 and pollutant concentrations for three selected individual chase measurements. The bus stops (A–G) are marked on the map and in the panel showing the time series of speed. The time series representing individual measurements are shown for three types of buses, all driving westward. Compared to average emissions of all studied EEV buses, the EEV bus selected for the figure had slightly higher PM and BC emissions and slightly lower PN emissions. The emissions of selected Euro VI and Retrofit 2 buses presented average emissions of all DPF-equipped buses. The approximate WGS 84 coordinates of the measurement route are: N:60.231, E:24.990. (The map is courtesy of the National Land Survey of Finland; a larger map is available in the Supplementary material (Figure S1)).

provided in the Supplementary material. Particle emissions were measured without any conditioning to resemble the real concentrations and characteristics of fresh exhaust emissions. Number concentrations of particles larger than 3 nm were measured with a condensation particle counter (CPC, 3776, TSI Inc.) and particles larger than 1.3 nm with a combination of a particle size magnifier (PSM, A10, Airmodus Oy) (Vanhanen et al., 2011) and a CPC (A20, Airmodus Oy), which is further denoted as the PSM. This parallel setup, which Rönkkö et al. (2017) also used, allowed for the measurement of the NCA concentrations in particle sizes between 1.3 and 3 nm. A dilution bridge with a dilution ratio of 40 was used in front of the CPC and the PSM. Particle size distributions from 6 nm to 10 µm were measured with an electrical low-pressure impactor (ELPI, Dekati Ltd.) (Keskinen et al., 1992; Marjamäki et al., 2000) equipped with a filter stage (Marjamäki et al., 2002) and an additional impactor stage with a cut diameter of approximately 17 nm (Yli-Ojanperä et al., 2010). A dual-spot aethalometer AE33 (Magee Scientific) (Drinovec et al., 2015) equipped with a PM_1 cyclone measured the black carbon (BC) mass concentration. The BC mass concentration was calculated using an 880 nm wavelength and a mass-absorption cross section of 7.77 m^2/g (Drinovec et al., 2015). All instruments sampled at a rate of 1 Hz or faster, except the NO_x analyzer, which sampled at a rate of 0.125 Hz. The real time responses are somewhat slower than the sampling rates (see the Supplementary material).

2.4. Data processing

The measurement data were first divided into individual time series according to the logged starting and ending times of each one-way chase measurement, including correction of the instrument time axes.

The emission factor calculation requires background concentrations, which were acquired every day by driving the route and measuring concentrations without chasing a bus. This calculation takes emissions from other vehicles into account as an average but not individual high-emitting vehicles. Average gas and particle concentrations were calculated over individual chasing events, and day-specific background concentrations were subtracted from these values. The emission factors were calculated from these background-level corrected values using the CO_2-based method Pirjola et al. (2016) described, using a CO_2 emission factor 3160 g/kg_{fuel} (Huss et al., 2013). Regarding the emission factors, see also a more detailed description included in the Supplementary material.

The ELPI data was used to calculate the PM_1 values and particle size distributions (see Supplementary material). Particles' LDSA (Oberdöster, 2000; Brown et al., 2001) was also calculated from the ELPI current data by using a calculation factor of 33 $\mu m^2/(cm^3 \cdot pA)$, defined for alveolar deposition in a road environment (Kuuluvainen et al., 2016). The ELPI was used in the measurements on February 15th and 16th. Particle losses in the PSM were approximately 20% over the entire size range, and they were corrected in the data analysis. More information on the PSM data analysis and particle losses in the sampling lines is given in the Supplementary material.

3. Results

3.1. Exhaust plume concentrations along the bus route

An example of time series representing three bus types (EEV, Euro VI and Retrofitted EEV (Retrofit 2)) are shown in Fig. 1. The figure shows the measurement route, speed, exhaust plume concentrations of CO_2, PM_1, black carbon (BC) and particle number (PN) for individual chase measurements. These values and other plume concentration values in Chapter 3.1 are given as measured (i.e., without background correction). In Fig. 1, the bus stops are marked as A-G. Between the stops, the buses reached speeds of approximately 50 km/h during the first half of the route and approximately 30 km/h during the latter half.

For all three buses, the exhaust plume's CO_2 concentrations

Table 1
Detailed description of the studied bus types.

Model	Manufacturing/retrofitting year	Emission level	Exhaust aftertreatment system	Number of buses studied
Scania K280UB LahtiScala 6 × 2	2013	EEV	EGR	12
Scania K280UB OmniExpress 320LE 6 × 2	2015	Euro VI	EGR-DPF-SCR	2
Volvo B8RLE 8900LE 6 × 2	2015	Euro VI	DPF + SCR	3
Scania K280UB LahtiScala 6 × 2 + Retrofit 1	2013/2015	EEV + Retrofit 1	DPF + SCR	1
Scania K280UB LahtiScala 6 × 2 + Retrofit 2	2013/2016	EEV + Retrofit 2	EGR + DPF + SCR	2

varied between 400 ppm and 600 ppm. The highest CO_2 concentrations were attributed to accelerations from the bus stops, conducted under high engine-load conditions. At more stable speeds, CO_2 concentrations typically remained below 500 ppm, except at the end of the route with a moderate uphill (average gradient 3.7%), which likely increased the engine load and CO_2 emissions. The chasing distance, wind and the location of the tailpipe may slightly affect the measured pollutant concentrations.

Pollutant concentrations in exhaust plumes significantly depended on the bus type. The highest exhaust plume PM_1 and BC concentrations were measured for EEV-level buses, for which the typical exhaust plume concentrations were below 30 $\mu g/m^3$ (PM_1) and 10 $\mu g/m^3$ (BC), and maximum concentrations, when examined with a 1 s time resolution, reached values up to 85 $\mu g/m^3$ (PM_1) and 30 $\mu g/m^3$ (BC). The appearance of high PM_1 and BC concentrations coincided with high CO_2 concentrations and high engine-load conditions. The highest PM_1 and BC plume concentrations occurred during accelerations and uphill driving. In contrast, the PM_1 and BC concentrations in the exhaust plumes of the Euro VI bus and the retrofitted bus were very low, close to background concentrations, and they did not systematically coincide with the plume's CO_2, vehicle speed or engine-load conditions. Therefore, the advantage of the Euro VI or retrofitting of EEV buses with DPF is clearly observed in the decrease of PM_1 and BC concentrations. Interestingly, the general level of exhaust plume PN concentrations was relatively similar among all bus types, and it did not coincide clearly with PM_1 or BC. This finding is explained by atmospheric dilution, sampling without heat treatment and small cut size of the CPC, which all bias the PN measurement toward small nucleation mode particles. These small particles produce only minimal signals in mass based metrics such as PM_1. At 400 s, a very high particle number concentration peak was measured from an EEV bus's exhaust plume. During this situation, the bus was approaching a bus stop, and the other emissions (CO_2, PM_1 and BC) were rather low. Therefore, the high particle number concentration was possibly caused by engine motoring (Rönkkö et al., 2014). Particles during the engine motoring have been associated with lubricant oil, and the increase in concentrations is linked to the reduced cylinder pressure of non-fired operation (Karjalainen et al., 2016).

The PM_1 concentrations of exhaust plumes are shown in Figure S3 as a function of location (same measurements as in Fig. 1). It is apparent that for the EEV bus, the PM_1 concentration and likely the human exposure to particles are typically relatively high near the bus stops, indicating that bus traffic can cause localized increases in pollutant concentrations. Figure S3 also indicates that with more advanced emission control technologies, those localized high concentrations can be significantly reduced because in the case of the Euro VI or Retrofit 2, no significant PM_1 concentration peaks occurred except the peak detected in the eastern part of the route for Retrofit 2. This peak may also originate from other traffic, which is present in this part of the route. In addition, it should be noted that the tailpipe outlet's location was not uniform between the bus types, and during the stops, the dilution might differ from normal driving.

3.2. Particle size distributions and nanocluster aerosol (NCA) emissions

In addition to PM_1 concentrations, the ELPI data was used to calculate the average exhaust plume particle and LDSA size distributions separately for each bus type. These measurements and the average background distributions are presented in Fig. 2. The average particle size distribution from an EEV bus exhaust plume, consisting of a clear soot mode (mean particle size approximately 40–60 nm in number size distribution and 70–110 nm in LDSA size distribution), differs significantly from other distributions. In general, retrofitting the EEV buses or the Euro VI emission level reduces the soot mode (20–200 nm in particle diameter) concentrations to the background level. The particle concentrations in the 10 nm size range vary significantly and do not systematically depend on the EAT system. The lowest values for these small particles are observed for Euro VI buses and the highest for the retrofitted buses, regardless of DPFs retrofitted to these buses. In the case of the retrofits, especially Retrofit 1, the high emissions of small particles may be related to the EAT system, and it is likely that these particles are formed after the DPF, most likely in cooling dilution of exhaust. Herner et al. (2011) and Thiruvengadam et al. (2012) measured small particles (diameter < 20 nm) after a DPF and SCR from a heavy-duty diesel engine. The formation is explained by the oxidative potential of the aftertreatment system, which increases oxidation of SO_2 to SO_3, promoting the formation of sulfur-driven nucleation. In addition, the excess ammonia from the SCR may increase the formation of nanoparticles. Lemmetty et al. (2007) calculated increased nucleation mode particle concentrations when ammonia was present in the exhaust, and Amanatidis et al. (2014) observed this increase experimentally. Therefore, in principle, differences in the concentrations of the smallest exhaust particles, for instance between Euro VI buses and the Retrofit 1 bus, may be caused by different catalysts used in DPFs, which can produce different sulfuric acid concentrations in the exhaust. Additionally, exhaust temperature and engine load may affect the formation of sulfuric acid. These effects can be combined with the effects of other exhaust compounds (such as ammonia and hydrocarbons) on exhaust nanoparticle formation (Arnold et al., 2012).

The parallel measurements with the PSM and the CPC were used to estimate the NCA (particle diameter D_p 1.3–3 nm) number concentrations. For example, a time series from EEV bus number 1310 measured on day 12 is shown in Fig. 3. The PSM measured almost equal concentrations with the CPC during certain times, indicating low NCA emissions relative to the total number of particles. However, most of time, the PSM measured higher concentrations than the CPC, indicating that the exhaust plume contained NCA particles. These NCA emissions are easily seen, for instance, during the high concentration peak at 215 s (see Fig. 3).

The averaged ratios of the NCA concentrations to the total particle number concentrations for each chase measurement are also shown in Fig. 3. These values include the compensation of the PSM's internal particle losses and losses in the sampling tubes

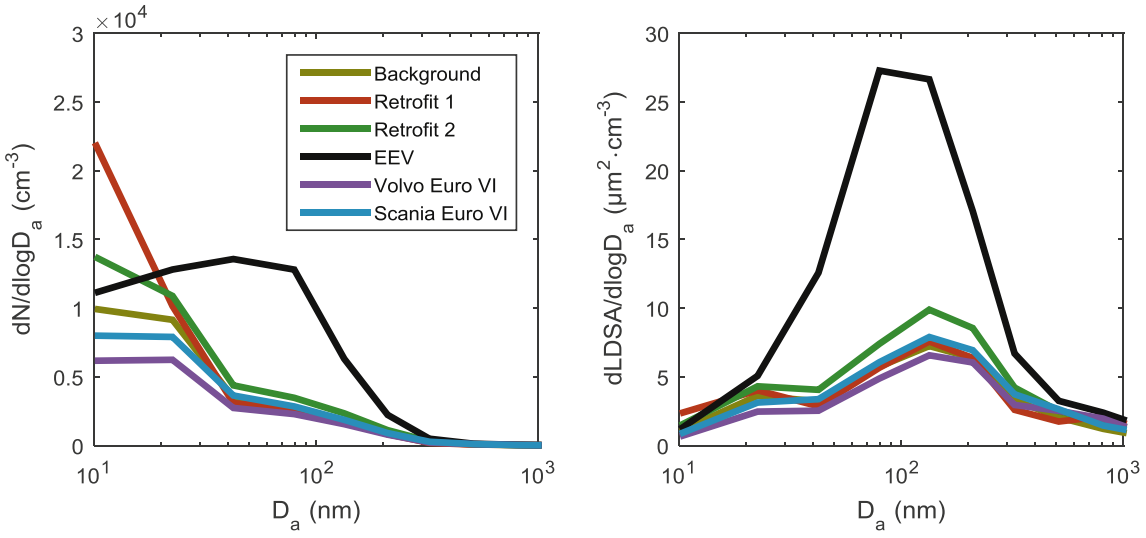

Fig. 2. Average aerodynamic particle number-size distributions and alveolar LDSA distributions in the exhaust plumes of various city buses. The average background from the separate measurements is presented in both figures (see also Figure S4) but it is partly covered by distributions from Euro VI and retrofit measurements.

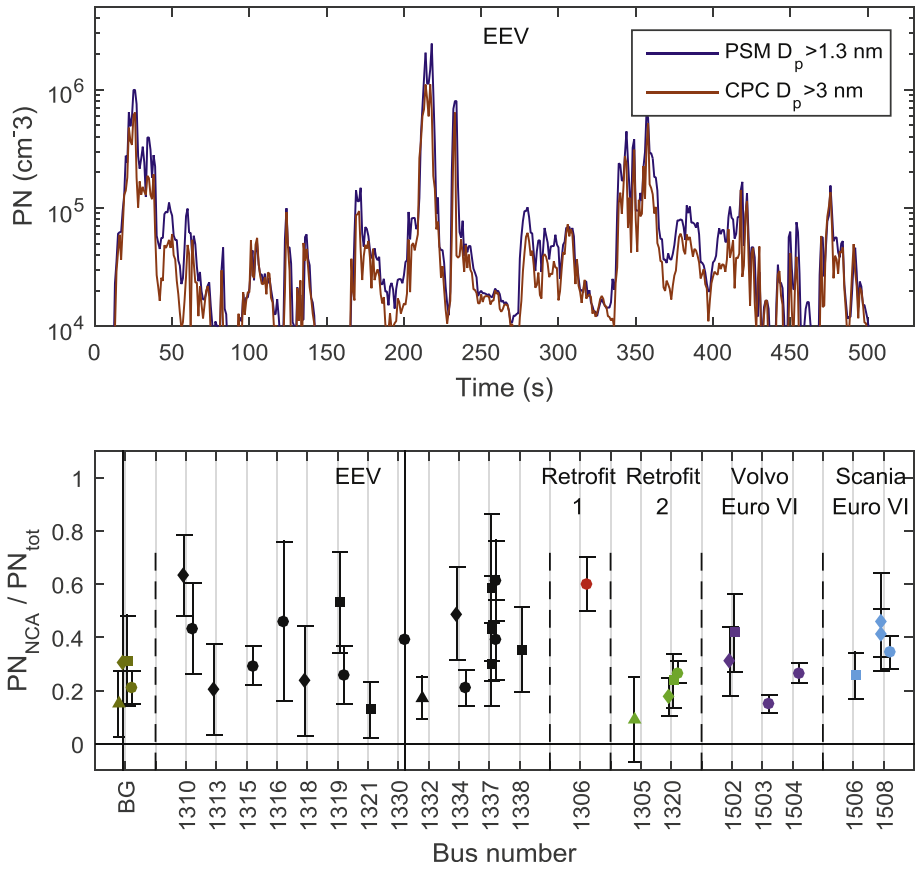

Fig. 3. Nanocluster aerosol (NCA) concentrations in city buses' exhaust plumes. In the upper part, an example of the particle number concentration time series measured using the PSM (including detection efficiency correction) and CPC are shown. Note the logarithmic y-axis. In the bottom part, the number concentration of the NCA in proportion to the total particle number concentration is shown for individual buses as an average over the individual chase measurement. The bars indicate standard deviations, which were calculated by dividing one measurement into 10 approximately 60 s periods. The BG refers to the urban background measurements. Different symbols represent different measurement days in February 2016: ▲ 11th, ◆ 12th, ■ 15th, and ● 16th.

calculated for particles with diameter of 2 nm. The relative NCA emissions varied between the buses and bus types. The mean relative NCA background concentration was 0.24, and the mean values measured from the exhaust plumes were 0.38 (EEV), 0.60 (Retrofit 1), 0.37 (Scania Euro VI), 0.29 (Volvo Euro VI) and 0.19 (Retrofit 2). Note that the buses producing high NCA emissions during one measurement can produce low NCA emissions during another measurement, such as bus number 1337 in this study. This

discrepancy can result from differences in driving style or e.g. the storage and release of sulfuric compounds in exhaust system, e.g. Rönkkö et al. (2013) and Karjalainen et al. (2014) reported relatively high variation in exhaust sulfuric acid concentrations during steady-state driving conditions. The exhaust's sulfuric acid has been linked to the formation of exhaust nanoparticles.

3.3. Emission factors

Emission factors for PN, NCA, BC and LDSA are presented in Fig. 4 as correlation plots. The background concentrations were subtracted from the reported values. First, the effects of diesel particle filters on soot mode particles are easily seen in Fig. 4 in the alveolar LDSA, PM_1 and BC metrics. The newest Euro VI and the retrofitted buses produced significantly lower emissions than the EEV buses, which were not equipped with DPFs. As a result, the DPFs in diesel buses, installed by the vehicle manufacturer or as a retrofit, can be seen as an effective particle-emission reduction method improving the urban air quality. They simultaneously decrease the emitted aerosol's light absorbing properties, thereby reducing the emitted aerosol's climatic effects.

A nearly linear correlation was observed between the emission factors of BC and LDSA. The result indicates very strongly that the soot mode particles consisting mostly of light-absorbing BC are also responsible for the LDSA emissions. A strong correlation was also observed between BC and PM_1 emissions. The buses equipped with DPFs have somewhat higher PM_1 to BC ratios. This difference may result from the DPF's capability to remove soot (BC) efficiently but not necessarily semivolatile compounds that condense on existing particles increasing the PM_1 value.

The BC emission factors were not correlated with PN emissions factors. In the particle number emission factors PN EF ($D_p > 3$ nm), no obvious difference emerged between the emission reduction technologies. Although the DPF removes the soot particles, nucleation can occur when the hot exhaust is mixed with the ambient air. This process typically produces high concentrations of particles smaller than 20 nm, which are responsible for most of the measured PN emission factor. The standard PN emission measurement with heated aerosol conditioning does not measure these nucleation mode particles although they can exist in fresh exhaust.

The NCA emission factors varied significantly, from very low levels to higher than $8 \cdot 10^{15}$ #/kg_{fuel}. Therefore, the city buses may significantly contribute to the mean NCA emission of traffic; e.g. Hietikko et al. (2018) reported an NCA emission factor of $9.36 \cdot 10^{14}$ #/kg_{fuel} for the traffic in street canyon in Rönkkö et al. (2017) reported slightly higher emissions factors. In this study, the NCA emissions were linked with the PN emissions of particles larger than 3 nm. This finding may suggest that the NCA mode is formed during the same processes with the larger nanoparticles but not in the process where larger soot particles are formed.

The average emission factors for each bus type are presented in Table 2, along with the NO_x emission factors and emission factors determined previously for EEV-level buses, adapted from Pirjola et al. (2016). It can be seen that the shift from EEV to Euro VI technology decreased the emissions in all studied emission categories. Soot emission seen in multiple metrics (PM_1, LDSA, BC) as well as NO_x emissions from Euro VI buses were extremely low, often below the detection limit of the measurement procedure. As a

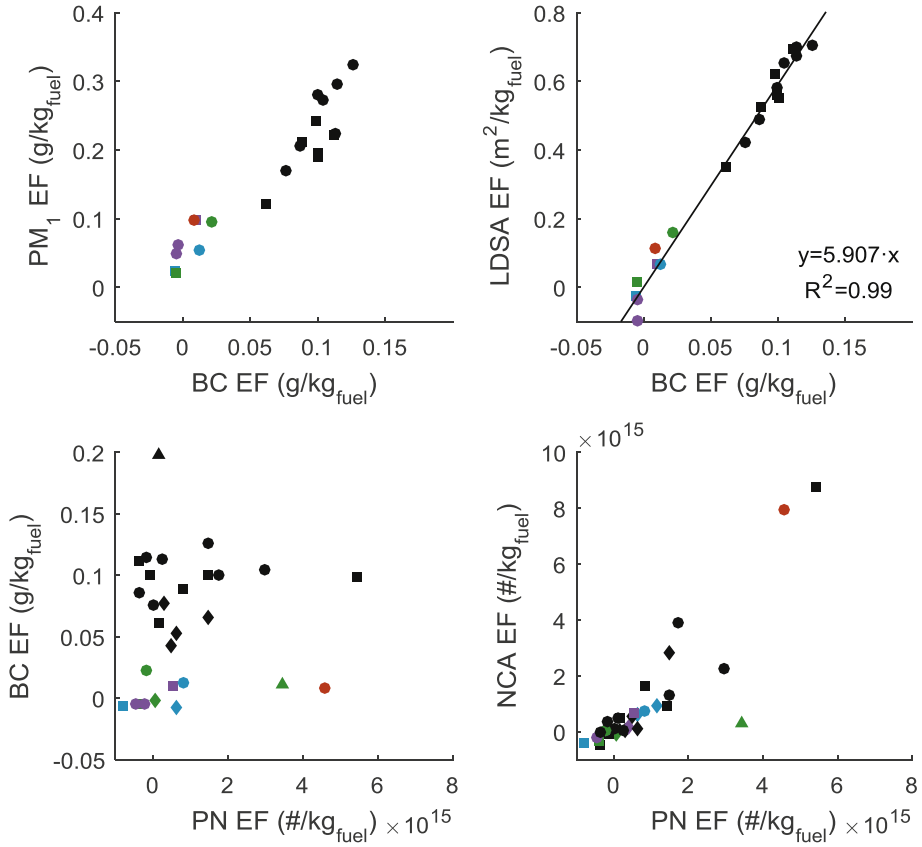

Fig. 4. Correlation of various emission factors. Color coding: EEV, black; Volvo Euro VI, purple; Scania Euro VI, blue; Retrofit 1, red; Retrofit 2, green. Different symbols represent different measurement days in February 2016: ▲ 11, ◆12, ■15 and ● 16. Note that PN EF is defined for particles larger than 3 nm and NCA EF for particles between 1.3 and 3 nm. The background variation produces emission factor uncertainties for individual chase measurements as follows: PM_1 ±0.14 g/kg_{fuel}, BC ±0.027 g/kg_{fuel}, LDSA ±0.20 m^2/kg_{fuel}, PN ±$1.4 \cdot 10^{15}$ #/kg_{fuel}, NCA ±$0.57 \cdot 10^{15}$ #/kg_{fuel}. Calculations are shown in the Supplementary material.

Table 2
Average emission factors ± standard deviations for various pollutants. The number (PN) emission factor was defined for particles $D_p > 3$ nm and the NCA emission factor for particles 1.3 nm < D_p < 3 nm. The NCA emission factors include diffusion correction with an assumption of an NCA diameter of 2 nm (see the Supplementary material). The LDSA emission factor was defined for alveolar-deposited particle surface area. Negative values indicate that the measured pollutant concentrations were lower than in the background measurements due to the measurement method being at its limit in these particular cases.

Model	PN ($\cdot 10^{15}$/kg$_{fuel}$)	NCA ($\cdot 10^{15}$/kg$_{fuel}$)	LDSA (m^2/kg$_{fuel}$)	PM_1 (g/kg$_{fuel}$)	BC (g/kg$_{fuel}$)	NO_x (g/kg$_{fuel}$)
EEV	0.86 ± 1.40	1.23 ± 2.14	0.52 ± 0.26	0.20 ± 0.09	0.10 ± 0.03	22.2 ± 7.1
Scania Euro VI	0.45 ± 0.85	0.47 ± 0.61	0.03 ± 0.09	0.05 ± 0.07	0.01 ± 0.03	−0.5 ± 1.3
Volvo Euro VI	0.01 ± 0.47	0.19 ± 0.37	−0.02 ± 0.07	0.06 ± 0.03	0.00 ± 0.01	0.1 ± 1.7
Retrofit 1[a]	4.56	7.97	0.11	0.10	0.01	31.6
Retrofit 2	0.72 ± 1.83	−0.01 ± 0.27	0.07 ± 0.12	0.03 ± 0.05	0.01 ± 0.01	7.3 ± 7.2
EEV EGR-DPF[b]	0.21 ± 0.01	–	–	0.40 ± 0.28	0.28 ± 0.17	32.9 ± 7.6
EEV SCR[b]	0.70 ± 0.38	–	–	0.28 ± 0.17	0.22 ± 0.01	39.8 ± 4.2

[a] Only one chase measurement, standard deviation cannot be calculated.
[b] Pirjola et al. (2016),chase measurement with mobile laboratory.

result, differences between the Euro VI busses from different manufacturers cannot be estimated. Retrofitting decreased the soot emissions to the same level as that of Euro VI buses but not the particle number. Particle number emissions are most likely caused by nucleation in the cooling and diluting exhaust. Retrofit 1 emitted NO_x at comparable levels with the original EEV buses. Unfortunately, this bus was measured only once, and it is not possible to make certain conclusions from this specific bus's emissions. As a comparison, Pirjola et al. (2016) made measurements in a more controlled environment, in a bus depot. They measured somewhat lower particle number emissions but higher NO_x and BC emissions. In addition, Pirjola et al. (2016) and Jerksjö and Hallqvist (2016) observed relatively high NO_x concentrations for buses equipped with SCRs. It should be noted that driving cycles are probably different, and during the study conducted by Pirjola et al. (2016), the ambient temperature was higher, possibly resulting in lower emissions of nucleation-mode particles (Kittelson et al., 2000; Rönkkö et al., 2006). Jerksjö and Hallqvist (2016) reported mean particle mass emission factor values from 0.05 to 0.3 g/kg$_{fuel}$ for diesel-fueled Euro V buses, and NO_x emission factors ranged from 13 to 27 g/kg$_{fuel}$. These values are close to what we obtained for EEV buses. They have also measured a low mean NO_x emission factor of 4 g/kg$_{fuel}$ and an extremely low particle mass emission factor of 0.002 g/kg$_{fuel}$ for Euro VI buses using rapeseed methyl ester fuel, which are in line with our observations of Euro VI diesel bus emissions. Note that the European particle number emission limits cover only solid particles larger than 23 nm (European Comission Regulation 582/2011), but in this study, the particle number emissions cover all particles larger than 3 nm. Therefore, the particle number emissions determined in this study are not comparable with regulatory measurements.

4. Discussion

In recent studies (Zhang et al., 2014; Yang et al., 2015; Ntziachristos et al., 2016), diesel vehicles' NO_x emissions have been measured to be significantly higher in the real world than in laboratory measurements, mostly due to the differences in engine loading patterns but also due to the disabling of emission reduction systems. The evaluation of vehicle particle emissions is even more complicated: real particle emissions can differ from emissions measured under laboratory conditions for the particle number, mass, size distribution, physical properties and chemical composition. These challenges can be tackled by studying the emissions under real-world conditions (i.e., in real driving and environmental conditions) which is possible with chase measurements (this study, see also Rönkkö et al., 2006; Pirjola et al., 2016; Saarikoski et al., 2017), roadside measurements, traffic tunnel measurements or on-board measurements (Franco et al., 2013). However, the on-board measurements frequently utilize tailpipe sampling, possibly leading to results not reproducing real exhaust plumes' characteristics in terms of semivolatile particulate matter. In chase measurements, these non-idealities are avoided. On the other hand, background aerosol can affect the measurements, which can be compensated for by using long measurement times and a high number of repetitions. In this study, measurement time was relatively long compared to remote sensing measurements (Franco et al., 2013). Fewer retrofitted EEV and Euro VI buses were measured than EEV buses, but in PM_1, BC and LDSA metrics, emissions from these DPF-equipped buses were low, which indicates that the ambient aerosol was not significantly affecting these results. In addition, results indicate that chase measurements are capable of distinguishing technological differences between the buses.

This study showed that with technology changes in vehicles, the particle and NO_x emissions can be diminished, and that it is beneficial to verify the effects of investments under real operating conditions. The advantage of the Euro VI or retrofit over the EEV-level bus was clearly observed in the decrease in PM_1, LDSA and BC emissions. In the case of Euro VI and retrofitted buses, plume PM_1, LDSA and BC concentrations were practically at the background level, indicating very low emissions. The NO_x emissions of one retrofitted bus were significantly lower than the NO_x emissions of EEV buses. One retrofitted bus produced NO_x emissions comparable to EEV buses and one retrofitted bus emitted NO_x even more than EEV buses on average. This finding may be related to different EGR strategies used in retrofitting, but a larger measurement set with retrofitted buses would clearly improve understanding of NO_x emission reduction in larger vehicle fleet.

The NCA emissions took place simultaneously with the number emissions of larger nanoparticles. Importantly, the NCA emissions were observed to differ between the buses, and they depended on driving conditions. The fraction of NCA emissions among total particle number was the lowest, 0–50%, for the newest technologies (i.e., for Euro VI buses and EEV buses retrofitted with the SCR and DPF) except Retrofit 1, which was a high-NCA emitter. For EEV buses, the fraction of the NCA emissions among the total particle number emissions was on average 0–65%, which was higher than for Euro VI or Retrofit 2. On average, the highest NCA emissions were measured for the EEV buses, which also had the greatest BC emissions. Therefore, our study indicates that the condensation sink and coagulation sink, formed in the exhaust by the soot-mode particles, do not prevent the emissions of NCAs. Our hypothesis is that without the DPF, the NCA is already formed in the combustion process or just after it, which results in nonvolatile nano-sized (1–3 nm) particles. This phenomenon has been observed previously e.g. by Alanen et al. (2015), who reported that natural gas engine exhaust was dominated by NCA already formed under high-

temperature conditions (i.e., in the cylinder or exhaust manifold). Furthermore, Alanen et al. (2015) suggested that the size of these particles increased as the exhaust cooled during dilution. When vehicles are equipped with DPFs (Euro VI buses and retrofitted EEV buses in this study), NCA formation can take place during the dilution process, most likely via the exhaust's sulfuric-acid-driven nucleation processes. However, technologies such as fuel, lubricant oil, engine design and EAT devices affect the semivolatile compounds that, on the other hand, have a significant effect on the final particle size of emitted particles. It is known that ambient temperature and humidity can affect nanoparticle formation (e.g., Rönkkö et al., 2006). Therefore, this study's results should be interpreted considering the measurement conditions (winter).

Regarding the NCA emissions, results indicate that emissions of the smallest particles can be reduced by advanced EAT systems. However, it should be noted that the smallest particles of vehicle exhaust can have several formation mechanisms, meaning that a change in vehicle technologies can also change the formation mechanism of NCAs, and, hence, their composition and physical characteristics.

5. Conclusions

Exhaust particle emissions from EEV, Euro VI and retrofitted EEV buses were studied during normal operation. Chase measurements were made utilizing a mobile laboratory equipped with on-line instrumentation for aerosols. Buses equipped with DPFs (i.e., Euro VI and Retrofitted EEV buses) produced minimal BC, PM_1 and LDSA emissions, indicating that the DPF removes soot particles from exhaust with high efficiency in real operation. In addition, shifting in bus fleet from EEV to Euro VI was found to effectively reduce NO_x emissions. Particle number emissions ($D_p > 3$ nm) measured without any temperature conditioning for the sampled aerosol varied significantly between the bus types studied. The largest average particle number emission factors from individual chase measurements were obtained for EEV-rated buses and retrofitted buses. Buses' NCA ($D_p < 3$ nm) emissions varied between the bus types, individual buses and driving conditions. Buses producing low NCA emissions during one measurement can produce high NCA emissions during another measurement. Hence, NCA emissions should be studied in more detail to find the most relevant factors affecting NCA formation.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2019.04.033.

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