



# Particulate emissions of a modern diesel passenger car under laboratory and real-world transient driving conditions<sup>☆</sup>

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## ABSTRACT

Exhaust emissions from diesel vehicles are significant sources of air pollution. In this study, particle number emissions and size distributions of a modern Euro 5b -compliant diesel passenger car exhaust were measured under the NEDC and US06 standard cycles as well as during different transient driving cycles. The measurements were conducted on a chassis dynamometer; in addition, the transient cycles were repeated on-road by a chase method. Since the diesel particulate filter (DPF) removed practically all particles from the engine exhaust, it was by-passed during most of the measurements in order to determine effects of lubricant on the engine-out exhaust aerosol. Driving conditions and lubricant properties strongly affected exhaust emissions, especially the number emissions and volatility properties of particles. During acceleration and steady speeds particle emissions consisted of non-volatile soot particles mainly larger than ~50 nm independently of the lubricant used. Instead, during engine motoring particle number size distribution was bimodal with the modes peaking at 10–20 nm and 100 nm. Thermal treatment indicated that the larger mode consisted of non-volatile particles, whereas the nanoparticles had a non-volatile core with volatile material condensed on the surfaces; approximately, 59–64% of the emitted nanoparticles evaporated. Since during engine braking the engine was not fueled, the origin of these particles is lubricant oil. The particle number emission factors over the different cycles varied from  $1.0 \times 10^{14}$  to  $1.3 \times 10^{15}$  #/km, and engine motoring related particle emissions contributed 12–65% of the total particle emissions. The results from the laboratory and on-road transient tests agreed well. According to authors' knowledge, high particle formation during engine braking under real-world driving conditions has not been reported from diesel passenger cars.

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## 1. Introduction

Traffic emissions are a major concern since the emission sources are in the proximity of population. Large European capitals such as London and Paris are struggling with air quality issues although much effort has been put into updating the vehicle fleets to comply with the latest emission standards. The adverse health effects of ambient particles or particulate matter (PM) are often associated with the concentrations of PM<sub>2.5</sub> (mass concentration of particles smaller than 2.5 μm) or PM<sub>10</sub> in the ambient air. In addition, black

carbon (Janssen et al., 2011) and nanoparticles (Oberdörster et al., 2005) have been proposed as proxies for health risks. The World Health Organization (WHO) has classified diesel exhaust to be carcinogenic to humans (IARC, 2012). Especially, metals, metal oxides and specific semi-volatile organic compounds are considered as the hazardous material in the exhaust related particles. Nevertheless, epidemiological short-term studies on ultrafine particles (UFP) and mortality is still limited due to the lack of continuous monitoring of UFP, while there is abundantly ambient PM data available (Lanzinger et al., 2016 and references therein).

Vehicles increase particle concentrations in urban air by the emissions of primary particles, fresh exhaust PM, and aged exhaust PM (Rönkkö and Timonen, 2019; Maricq, 2007). Primary emissions are formed in the engine during the combustion processes, and the particle number size distribution may consist of two modes; an

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accumulation mode in the size range of 30–100 nm and a core mode in sub-10 nm the size range (Rönkkö and Timonen, 2019). The accumulation mode particles are non-volatile soot agglomerates (Kittelson, 1998; Tobias et al., 2001). The core mode particles consist of amorphous carbonaceous compounds (Sgro et al., 2012; Seong et al., 2014) or metallic ash compounds from fuel and lubricant oil (Fushimi et al., 2011; Lähde et al., 2014). Furthermore, exhaust contains semi-volatile compounds that condense onto the surfaces of nonvolatile particles or nucleate forming new particles when the exhaust rapidly cools at the end of the tailpipe (Arnold et al., 2006, 2012; Rönkkö et al., 2013; Shi and Harrison, 1999; Tobias et al., 2001; Schneider et al., 2005; Khalek et al., 2003). The newly formed particles consist of volatile material such as water, sulfate and hydrocarbons. These particles are hereafter called fresh exhaust particles. Secondary organic aerosol (SOA) is formed in the atmosphere from gaseous precursors emitted by vehicles. Photochemistry converts these high vapor pressure organic compounds to low volatility compounds which can form SOA via gas-to-particle conversion (Kroll et al., 2012; Chirico et al., 2010).

Due to the hazardous health effects and climate-warming potential (e.g. Bond et al., 2013), the emissions of light-duty vehicles are regulated. As legislation has brought more stringent emission limits, measuring particulate mass emissions has come down to the limit of detection in the gravimetric PM weighting method. Positive loading caused by condensing hydrocarbons is a problem. The updates to regulatory emission limits have tackled this problem by introducing a particulate number (PN) limit. In Europe, the PN emission level of non-volatile particles for light-duty (LD) diesel vehicles were implemented in 2011 emission level (Euro 5b standards). In 2011, the USA implemented the EPA Tier 3 regulations. Additionally, starting from Euro 6 emission standards implemented in 2014, non-volatile exhaust PN has been regulated for gasoline direct injection (GDI) passenger cars and heavy-duty (HD) road diesel vehicles such as trucks and buses. This non-volatile particle regulation aims to non-volatile particles greater than 23 nm in size. The current regulatory method does not capture the non-volatile particles smaller than 23 nm, semi-volatile or secondary particles at all. Since September 2018, all new vehicles in the EU are certified according to the new Worldwide harmonized Light vehicles Test Cycle (WLTC) chassis dynamometer procedure, which has replaced the previous New European Driving Cycle (NEDC) test protocol (Dieselnet, 2020).

In order to meet the strict emission limits, modern diesel cars are equipped with diesel oxidation catalysts (DOCs) to reduce the emissions of hydrocarbons and carbon monoxide. They are also equipped with diesel particle filters (DPFs), especially due to PN regulation, to reduce nonvolatile soot particle emissions down to allowed levels. Moreover, in several vehicle models, selective catalytic reduction (SCR) systems are applied to reduce nitrogen oxides. The DOC having an open flow structure has not been reported to significantly affect the non-volatile particle size distributions (Davies et al., 2018).

Some recent studies have shown that vehicle exhaust can also contain particles in the nanoparticle size range under engine braking (motoring) conditions when the fuel injection is stopped preventing combustion inside the cylinder. So far, these particles have been observed in on-road studies of heavy-duty (HD) diesel vehicles (Rönkkö et al., 2014; Karjalainen et al., 2016), in on-road and laboratory studies of gasoline direct injection (GDI) passenger cars (Rönkkö et al., 2014; Karjalainen et al., 2014a; Pirjola et al., 2015), and in laboratory studies of a diesel passenger car without a DPF (Pirjola et al., 2019). Pirjola et al. (2015) found that a 10% decrease in the Zn content of engine oils was linked with a 5–13% decrease to the non-volatile particle number emissions while the effect of lubricant on semi-volatile particle fraction was even

higher, in the order of 20%. Particles during deceleration have not only been observed in the direct exhaust measurements, instead deceleration conditions have been observed to cause majority of ambient particles (sizes between 5 and 560 nm) around crossroads area (Goel and Kumar, 2015).

Engine motoring related particles may be a significant source of particle number emissions for diesel passenger cars without an after-treatment system (ATS). Additionally, the load of these particles may significantly reduce the lifetime of DPFs since ash components from the lubricant oil accumulate on the DPF (Dittler, 2012). As unburned components of the exhaust, they are not cleaned from the filter during regeneration, and thus lead to poisoning of the catalyst.

The objectives of this paper are (i) to characterize particle number emission from a modern diesel passenger car (Euro 5b standard) exhaust under the valid regulated test cycles as well as during various transient and steady driving conditions, (ii) to study the role of lubricating oil in particle formation. Thus, the engine-out particulate emissions were measured using three different manufacturer-approved lubricating oils. Emissions were measured for all lubricant oils when the DPF was by-passed. In order to evaluate the performance of the ATS, for one lubricant oil, the measurements were also carried out when the engine was equipped with the DPF. Objective (iii) is to compare real driving emissions with chassis dynamometer emissions. Therefore, all transient test cycles were conducted under laboratory conditions with instruments connected to partial flow sampling system, and under on-road conditions with a chase vehicle. Objective (iv) is to put a special emphasis of the analysis on the events where particulate emissions occurred even though the diesel engine was not simultaneously fueled, and on how these emissions were affected by the choice of lubricant oil. According to our knowledge, nanoparticle formation during engine motoring under real-driving conditions on-road has not been reported for diesel passenger cars before.

## 2. Experimental methods

### 2.1. Test vehicle, fuel and lubricants

The test vehicle was a Euro 5b emission level compliant TDI passenger car Audi A4 Sedan (displacement 2.0 dm<sup>3</sup>, model year 2012, odometer reading ~30,000 km). The car was front wheel driven and equipped with a manual gearbox. Engine was a modern common rail turbo diesel engine with maximum power output of 105 kW. The engine was equipped with an ATS including a diesel oxidation catalyst (DOC) and a diesel particulate filter (DPF) along with an exhaust gas recirculation system (EGR).

As mentioned in Introduction, the purpose of these measurements was to determine whether diesel passenger cars emit particles during engine motoring and furthermore, if lubricants influence particle formation and emissions. DPFs have been observed to act very efficiently in removing particles from the engine exhaust. There were no means to control the timing and extent of regeneration events, therefore, the exhaust sample was taken upstream of the DPF in order to measure the particulate emissions originating from the engine. This was realized by removing the DPF and replacing it by a ball valve and muffler that caused the same backpressure for the engine as with the original ATS.

The engine was run on low sulfur (<10 ppm) commercial EN590 specified diesel fuel. Three commercial SAE 5W-30 lubricants from well-known brands were used. All of them were fully synthetic high-performance motor oils designed for extended oil drain intervals. The largest differences between the lubricants were in the fractions of the evaporation losses of the lubricants (Noack wt-%) and in the additive contents, particularly, in calcium, zinc and

chlorine concentrations (Table S1).

## 2.2. Laboratory experiments

### 2.2.1. Driving modes

The measurements were conducted over the NEDC (duration 1180 s, mileage 10.9 km) and the US06 (596 s, 12.8 km) cycles. The NEDC comprises an urban driving cycle (UDC) with a total duration of 780 s, followed by a 400-s-long extra-urban driving cycle (EUDC). The UDC is driven at low engine load causing low exhaust gas temperature, and the average speed is only 18.4 km/h, whereas the EUDC part simulates highway driving with an average speed of 62.6 km/h and reaching 120 km/h at maximum. The US06 cycle during the first 500 s describes aggressive, high speed and high acceleration driving behavior up to 129 km/h, and during the last 100 s rapid speed fluctuations up to 48 km/h and finally to 80 km/h. For each lubricant oil these tests were performed three times. Engine parameters, such as wheel speed, rpm, engine load and intake air mass flow (MAF) were recorded with an on-board diagnostics (OBD) logger having 1 Hz time resolution.

Three steady state driving at 5 kW wheel power with gear 5, at 10 kW and at 20 kW wheel powers with gear 6, were measured during ~7 min for all and non-volatile particles. These tests were repeated with each lubricating oil. Furthermore, transient driving cycles shown in Table S2 were performed. The cycles began with a steady speed indicated by the first value (20 or 30 km/h), then accelerated to the second value (50, 70, or 90 km/h), followed by steady driving after which the speed was decelerated to the starting value by engine braking. The cycles took 3–5.5 min, and they were repeated 3 times. These cycles were the same as used in a similar study on the emissions of a GDI passenger car (Karjalainen et al., 2014a; Pirjola et al., 2015). Although these cycles do not necessarily represent typical real driving conditions, they demonstrate particle formation during actual and extreme situations of diesel engine motoring.

### 2.2.2. Exhaust sampling and instrumentation

The exhaust sample was taken using partial flow dilution system (Ntziachristos et al., 2004). A combination of a porous tube -type diluter (PTD) with a dilution ratio (DR) of about 12, an ageing chamber with a residence time of 2.7 s, and an ejector-type diluter (Dekati Ltd.) with a DR of ~8 were used for sampling and diluting the exhaust (Fig. 1). The primary dilution ratio was set with dilution air controlled by a mass flow controller and further heated to 30 °C. This type of sampling system was used in order to mimic the effects of atmospheric dilution that occurs at the exit of tailpipe and on roadside in real life (Rönkkö et al., 2006; Keskinen and Rönkkö, 2010). Pressurized dilution air was filtered of particles and gaseous contaminants. The dilution ratios were calculated based on the measurements of the CO<sub>2</sub> concentrations in the tailpipe and the diluted exhaust. The CO<sub>2</sub>-concentrations were measured by a SIDOR (Sick Maihak) non-dispersive infrared CO<sub>2</sub>-analyzer.

An engine exhaust particle sizer (EEPS 3090, TSI Inc.) (Johnson et al., 2004) was used to measure the particle size distribution in the size range of 5.6–560 nm (mobility diameter) and an electrical low-pressure impactor (ELPI, Dekati Oy) (Keskinen et al., 1992) in the size range of 7 nm–10 μm (aerodynamic diameter). The ELPI was equipped with a filter stage (Marjamäki et al., 2002) and an additional impactor stage designed for nanoparticles (Yli-Ojanperä et al., 2010). An Ultrafine Condensation Particle Counter 3776 (UCPC, TSI Inc.) was used for the number concentration measurement of particles larger than 2.5 nm and a particle size magnifier (PSM A10 and a CPC A20) (Airmodus Oy) (Vanhanen et al., 2011) for particles larger than 1.3 nm. An aerosol diluter designed to minimize nanoparticle losses was used to provide additional dilution

with a DR of 43 for the CPCs. All the instruments operated with 1 s time resolution.

The volatile fraction of the exhaust particles was determined by alternating the measurement position between upstream and downstream of the thermodenuder (TD) (Rönkkö et al., 2011) in consecutive repetitions of the same driving cycle. The temperature of the sample was raised to 265 °C within the TD and evaporated components were removed by activated charcoal. As mentioned above, these non-volatile particles are called primary particles whereas the particles monitored without the TD are called fresh particles.

## 2.3. On-road experiments

The chase studies were conducted on the same diesel passenger car in a remote environment on an approximately 2 km strip of a straight road as in Pirjola et al. (2015) and Karjalainen et al. (2014a). The location was chosen to minimize the amount of interfering traffic and local sources of air pollution. A mobile emission laboratory, the “Sniffer” (Pirjola et al., 2004; Rönkkö et al., 2006; Pirjola et al., 2015), was used to chase the tested passenger car at a distance of ~10 m. The sample was taken from the front bumper of the Sniffer at a height of 0.7 m and transported in a sample line inside the car to the aerosol measurement devices. Two UCPCs and an EEPS were used to measure particle number concentrations and size distributions in real time. The UCPCs were organized so that simultaneous measurements both upstream and downstream of the thermodenuder were possible. The EEPS measured always upstream of the TD. Each of the transient cycles (Table S2) were repeated at least 15 times for each lubricant.

## 2.4. Data processing

All aerosol instrument data were corrected for the total DR of the sampling system, and for the particle losses within the PSM (Järvinen et al., 2019) and the thermodenuder (Heikkilä et al., 2009). The maximum penetration factor (minimum loss correction) based on Heikkilä et al. (2009) was used for the thermodenuder loss correction for the UCPC data. All data from the different instruments were synchronized to the speed profiles obtained from the vehicle's OBD data. Intake mass airflow was used to calculate the instantaneous emissions from the vehicle.

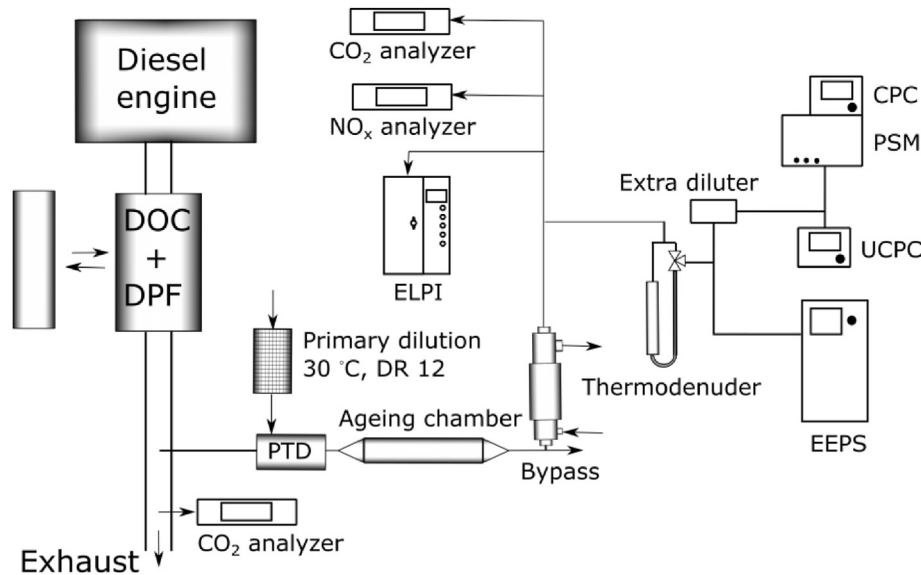
Emissions factors *EF* in #/km for particle number were estimated by the following equation

$$EF = \frac{\sum_{i=1}^n N_{raw,i} MAF_i}{\rho_{exh} \sum_{i=1}^n v} A \quad (1)$$

where *n* is the number of seconds of the cycle, *MAF<sub>i</sub>* is the momentary mass flow of the exhaust, *ρ<sub>exh</sub>* is the exhaust density (assumed to be air density), *v* is the vehicle speed, and *A* is a conversion factor for the units. *N<sub>raw,i</sub>* is the raw particle number concentration, and it is calculated by Eq. (2)

$$N_{raw} = \frac{\Delta N}{\Delta CO_2} \Delta CO_{2,raw} = \frac{(N - N_{bg})}{(CO_2 - CO_{2,bg})} (CO_2^{raw} - CO_{2,bg}^{raw}) \quad (2)$$

where *N* and *CO<sub>2</sub>* are the measured (diluted) particle number and carbon dioxide concentrations, and the subscript *bg* refers to the background concentrations.



**Fig. 1.** Measurement setup and instrumentation in the laboratory tests. Thermodenuder could be by-passed with a three-way valve. Dilution ratios were calculated as a ratio of the raw and diluted CO<sub>2</sub> concentrations. OEM DOC + DPF system was replaced with a valve and muffler during most of the experiments.

### 3. Results

#### 3.1. Particle number emissions during the NEDC and US06 cycles

Fig. 2 presents the time-series of the vehicle speed and number emissions of primary and fresh particles (>2.5 nm) during the NEDC and US06 cycles for different lubricants used. With all oils, driving conditions strongly affected exhaust emissions. The peak emissions occurred at vehicle accelerations and decelerations during engine braking. In the acceleration parts of the NEDC, the primary and fresh exhaust particle emissions were at a rather similar level, indicating that these particles were non-volatile in nature and already existed inside the tailpipe. However, under the engine braking conditions, especially when the car was braked from 120 km/h to 0 km/h, the primary particle emissions were clearly lower compared to the fresh particle emissions. Consequently, the majority of these particles were volatile. Overall, the highest emissions occurred with Oil 1; the emission factors (EF<sub>fresh</sub>) over the whole cycle were  $1.30 \times 10^{14}$ ,  $1.15 \times 10^{14}$  and  $1.18 \times 10^{14}$  #/km for oils 1, 2 and 3, respectively (Table S3), and the average fraction of primary particles of these were 76%, 89% and 90% for Oil 1, Oil 2 and Oil 3, respectively. As a curiosity, with Oil 1 automatic regeneration started before the EUDC during one cycle, and a huge number of particles was released as shown in Fig. S1. Consequently, the EF<sub>fresh</sub> was higher,  $1.58 \times 10^{14}$  #/km.

The emissions of nanocluster aerosol (NCA) with particle diameters in the range of 1.3–2.5 nm were measured during all cycles. As seen from Fig. 1, the highest NCA emissions occurred at the accelerations and decelerations, as for the fresh exhaust particles. The lubricant oils affected NCA emissions. During the NEDC, the largest EF<sub>NCA</sub> =  $3.23 \times 10^{13}$  #/km was estimated for Oil 1 covering 20% of the fresh exhaust EF<sub>PSM</sub>. The EF<sub>NCA</sub> was  $1.12 \times 10^{13}$  #/km for Oil 2 and  $4.5 \times 10^{12}$  #/km for Oil 3 being about 10% and 4% of the corresponding EF<sub>PSM</sub>.

Due to more aggressive driving conditions and higher speeds, the peak emissions during the US06 were much higher than during the NEDC (Fig. 1). The highest momentary emissions occurred during the fast accelerations with Oil 1 whereas the highest emissions during the engine braking were observed with Oil 2. Over the NEDC, the highest particle emissions occurred with Oil 1,

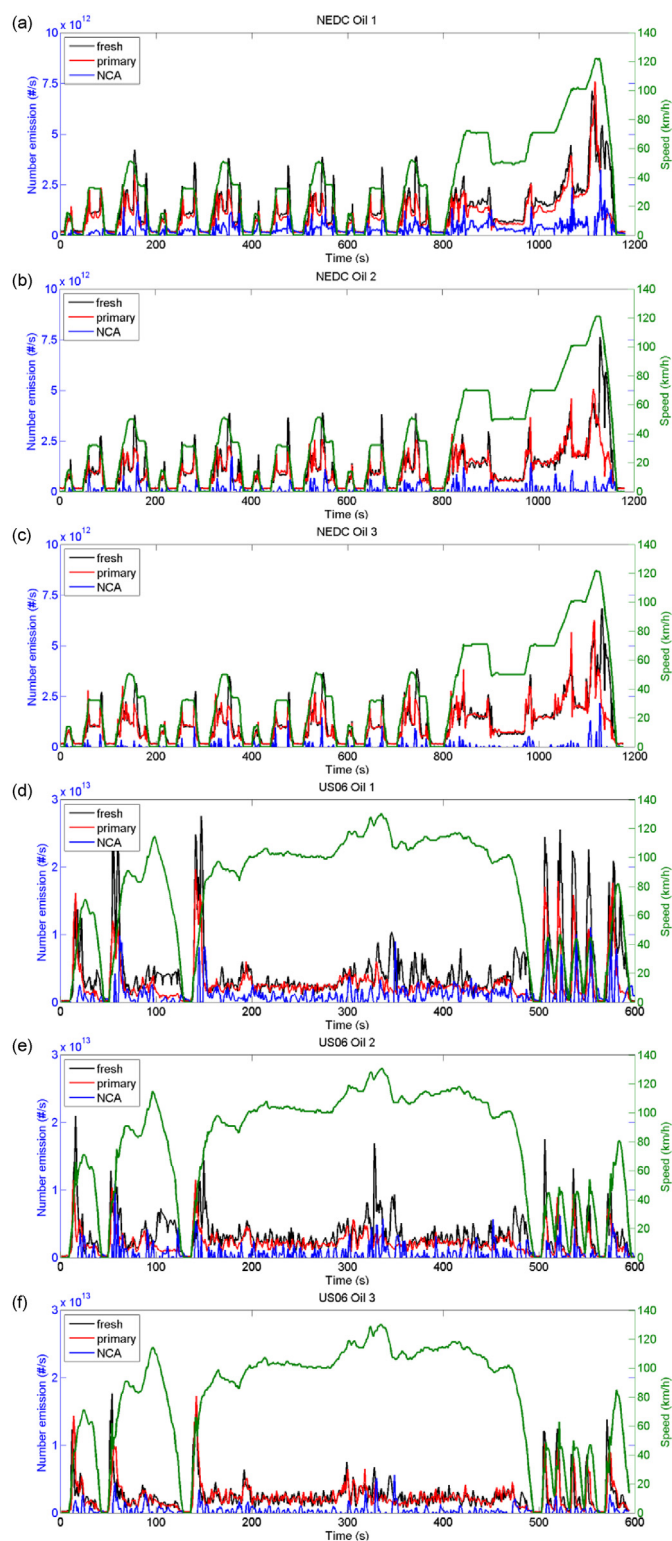
$2.04 \times 10^{14}$  #/km compared to  $1.48 \times 10^{14}$  #/km with Oil 2 and  $1.08 \times 10^{14}$  #/km with Oil 3 (Table S3). The average ratio of the primary and fresh particles were 63%, 66% and 96% for Oils 1, 2 and 3, respectively, indicating that with Oil 3 the particles were less volatile. The average NCA emission factors followed the order of the fresh exhaust emission magnitude (Table S3), and covered 22%, 20% and 17% of the fresh exhaust particle emissions factors (EF<sub>PSM</sub>) for Oils 1–3, respectively.

#### 3.2. Particle number size distributions during the NEDC, US06 and steady speed cycles

As an example, the time evolution of particle number size distributions measured by the EEPS during the NEDC and US06 with Oil 1 is shown in Fig. 3a–d, and with the other oils in Figs. S2–S3. During accelerations and high-speed driving in the EUDC of the NEDC, the particle size distribution was typically dominated by the soot mode particles, the mode peaking at ~60 nm (Fig. 3a). Instead, during deceleration, the size distribution was shifted to smaller particle sizes and the role of sub-20 nm particles became much more significant. Plausibly the origin of these particles was from the lubricant oil (Rönkkö et al., 2014; Karjalainen et al., 2014a).

During the US06 cycle, a stronger soot mode peaking at 70 nm was observed and furthermore, larger particles >200 nm appeared in aggressive accelerations during the first 150 and last 100 s (Fig. 3b,f). The smaller particle modes peaking at ~10 nm and 20 nm can clearly be distinguished over the both cycles (Fig. 3e and f). A major part of these particles was volatile and evaporated during the TD treatment so that the sizes of the primary particles were close to 10 nm. Some differences in the average size distributions of the emitted particles (#/km) between the oils can be discovered (Fig. 3e and f). The soot mode particle emissions were highest with Oil 1 over the both cycles, whereas the emissions of the particles smaller than 30 nm were highest with Oil 2.

The steady state tests (Table S2) indicate that the lubricants used have minimal effect on particle number emissions when the wheel power was low, 5 kW and 10 kW. Instead, at 20 kW wheel power a large mode peaking at ~80 nm was emitted with Oil 3 (Fig. S4). These particles were larger than the ones with Oils 1 and 2, and ~40% of these particles in number were volatile. Altogether, the



**Fig. 2.** Time series of particle number emissions (#/s) (measured by the UCPC) over the NEDC (a–c) and over the US06 (d–f) for the three lubricants. Vehicle speed is shown by green color. Note different y-scales for the NEDC and US06. Also shown are the NCA emissions measured by the PSM.

emissions of the soot particles decreased in increasing wheel power.

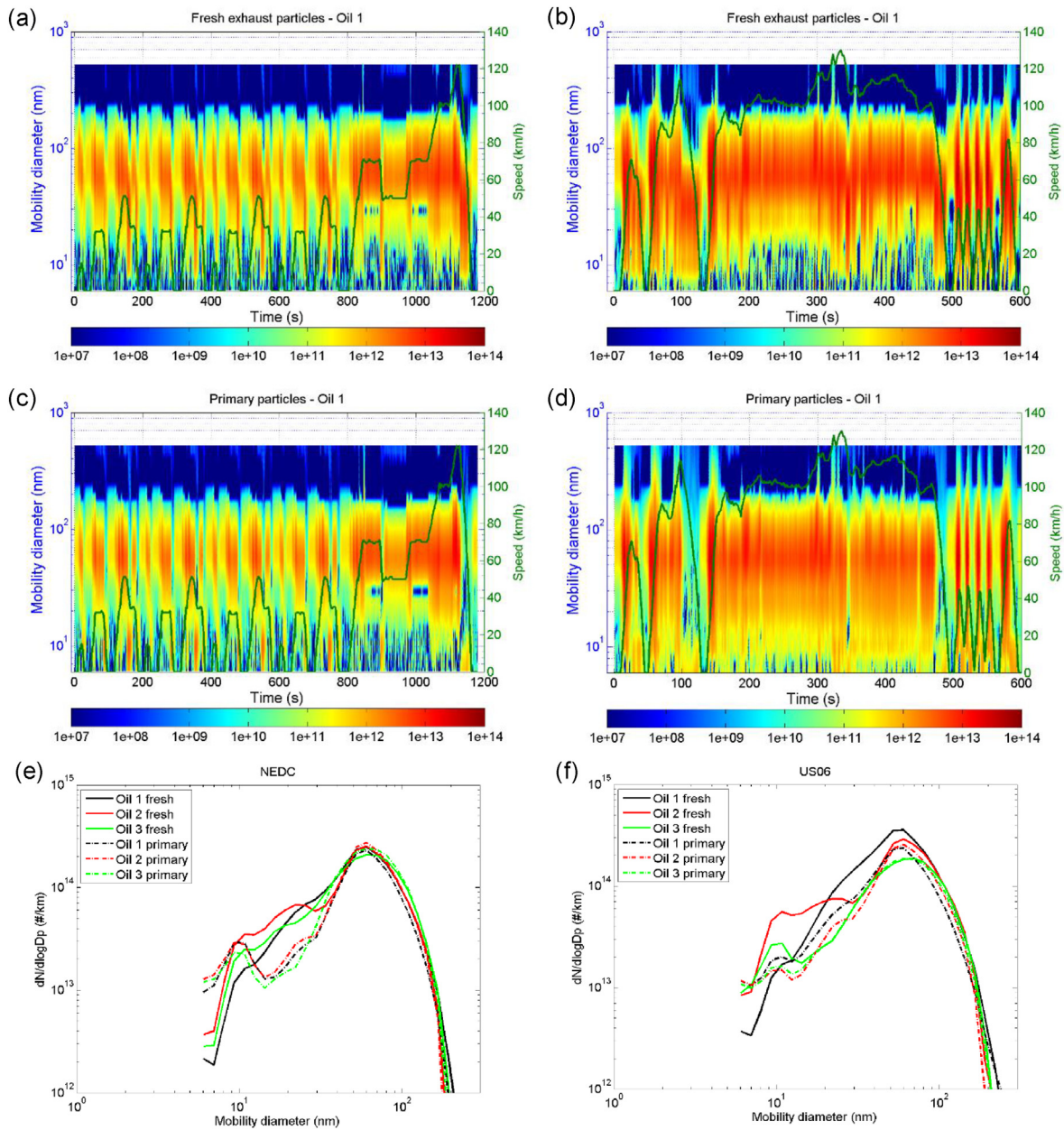
### 3.3. Particle number emissions during the transient cycles

Detailed investigations on the particle formation during different situations of diesel engine motoring were performed in the laboratory as well as on the road. The time evolution of the number size distributions measured by the EEPs of fresh exhaust particles emitted during the three transient cycles 30-70-30 with gear 3 (Table S2) are displayed in Fig. 4 for Oil 3 and in Fig. S5 for Oils 1 and 2. These measurements were performed on the chassis dynamometer with the ATS bypassed. As a general observation, the repeatability of the driving cycles was excellent. During accelerations, a wide soot mode peaking at  $\sim 60$  nm dominated both the fresh and primary particle number size distributions, and these emissions were similar. Instead, during deceleration by engine braking when the engine load was 0% (black curve in Fig. 4a and b), the size distribution totally changed; the wide soot mode vanished and a bi-modal size distribution was formed. The smaller mode was located at 10–20 nm and the larger mode, with much lower emissions, at around 100 nm (Fig. 4f). The larger mode was not practically affected by the thermodenuder while the smaller mode shifted towards the smaller particle sizes, and simultaneously, the number emissions dramatically decreased (Fig. 4d and e). The behavior of the emissions were similar with the other oils (Figs. S5 and S6).

The emission factors between the oils did not deviate much but the highest emissions over the transient cycle were observed with Oil 2. The fresh exhaust particle emission factors were  $1.43 \times 10^{14}$ ,  $1.56 \times 10^{14}$  and  $1.41 \times 10^{14}$  #/km for Oils 1–3, respectively, and the primary ones  $1.18 \times 10^{14}$ ,  $1.22 \times 10^{14}$  and  $1.12 \times 10^{14}$  #/km, respectively (Table S3) based on the UCPC data. No notable differences across engine lubricants was seen in the particle emissions during accelerations and steady speed parts of the cycle (Fig. S7). However, some differences were seen in the average size distributions of the brake particle emissions (Fig. 5e). While the larger mode emissions were highest with Oil 3, the smaller mode emissions were highest with Oil 2, leading to the highest brake particle emissions factor of  $1.82 \times 10^{14}$  #/km with Oil 2 over the braking distance and the lowest of  $1.34 \times 10^{14}$  #/km with Oil 1 (Table S3). With all the oils the majority of the smaller mode particulate material was volatile and consequently, the fraction of the primary brake particle number emissions of fresh brake particle emissions were 38%, 41% and 36% for Oils 1, 2, 3, respectively, whereas during the whole 30-70-30 cycle, the fraction of non-volatile particle number emissions of all particles were 83%, 78%, and 80%, respectively. Zn content was highest for Oil 2, 25% higher than for Oil 3, and sulfur content 7% higher than for Oils 1 and 3 (Table S1). These additives might have increased the particle emissions (Pirjola et al., 2015).

The parallel measurements with the PSM and UCPC for all oils and the estimated NCA emissions (particle diameter 1.3–2.5 nm) are presented in Fig. S8. During the acceleration and steady driving, the NCA formation is low while during the engine braking the NCA emissions cover 10% for Oil 1, 17% for Oil 2 and 20% for Oil 3, of all engine braking particle emissions.

In addition, the transient cycle was carried out when the vehicle was equipped with the DPF. During this experiment, the particle number emissions were very low (Fig. 5c and d), however, weak variability, similar as without the DPF, can be seen. Based on the UCPC data, with the DPF the EF was  $2.3 \times 10^9$  #/km, only 0.0016% of the case without the DPF. Thus, the filtration efficiency of the DPF was excellent (99.998%) during this cycle, and it was fairly independent of the altering driving conditions.



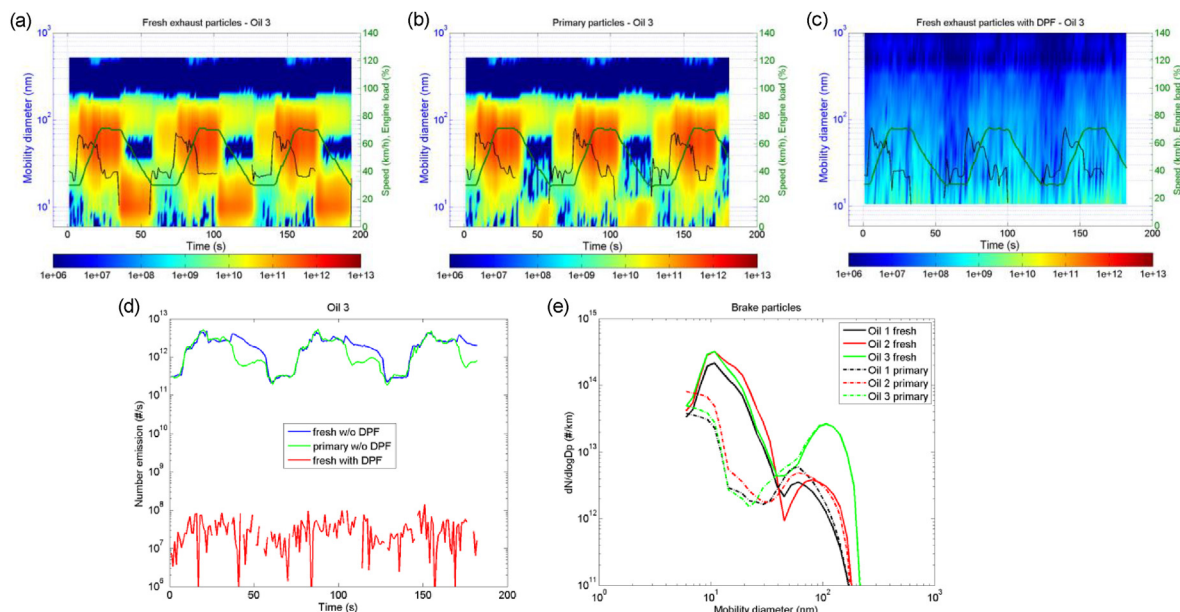
**Fig. 3.** Time series of the size distribution (measured by the EEPS) of the emitted fresh exhaust particles (a–b) and emitted primary particles (c–d) (#/km) during the NEDC (left panels) and during the US06 (right panels) when Oil 1 was used. Shown is also the vehicle speed. The average number size distributions of the fresh and primary particle emissions over the NEDC (e) and over the US06 (f) are presented for all oils.

Fig. 5 shows the emissions of particle number measured by the UCPC and the number size distributions measured by the EEPS with Oil 3 during one repetition of the 30-70-30/3 —cycle on the road. The experiment was conducted without the DPF in the test vehicle. The average dilution ratio of the exhaust was estimated as a ratio of the average  $\Delta\text{CO}_2$  concentration measured in the laboratory over the same cycle and the average atmospherically diluted  $\Delta\text{CO}_2$  concentration measured on road. In this case DR was 636. During the engine braking (after 30 s from the beginning) the smaller particle mode can be observed as in the laboratory tests, however, the larger particle mode seen in the laboratory measurements can hardly be detected on the road. Due to the much higher atmospheric dilution ratio their concentration is small and close to the detection limit of the EEPS.

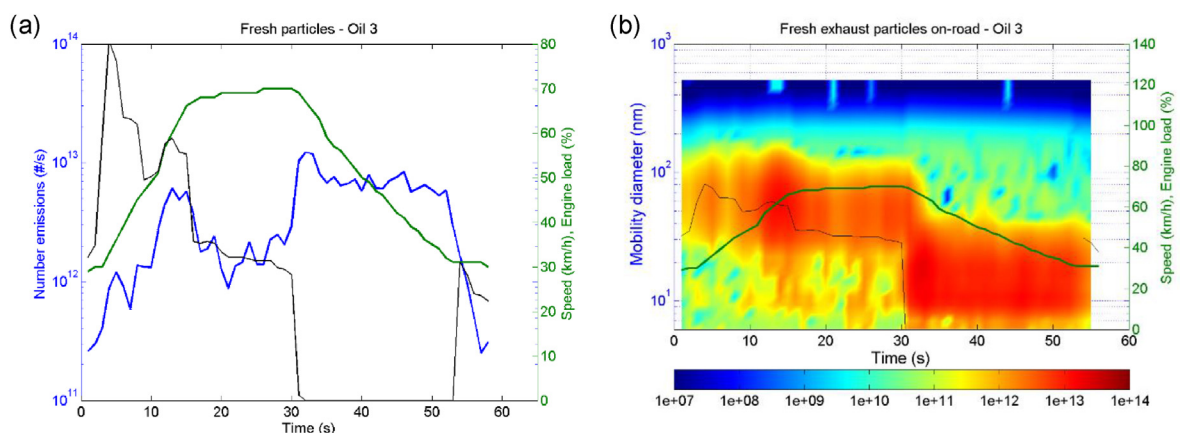
### 3.4. Emissions factors

Emission factors of all particles and engine brake particles for the three lubricant oils during the different cycles (Table S1) are shown in Fig. 6. The EFs were highly dependent on the test cycle. Over cycles 20-50-2/1, 30-70-30/2 and 30-90-30/2, the EFs were clearly higher compared to the NEDC and the US06 but it should be noted that the tests were driven by using a small gear. Altogether, the engine-out particle number emission factors varied from  $1.0 \times 10^{14}$  to  $1.3 \times 10^{15}$  #/km.

The emission factors calculated for different oils within a specific cycle did not vary much; however, the EFs with Oil 3 were the highest in the road environments compared to Oil 1 and Oil 2 whereas under the laboratory conditions they were most frequently the lowest. In practice, the fraction of brake particle



**Fig. 4.** Time series of the number size distribution of the emitted particles ( $\#/s$ ) during the 30-70-30/3 cycle for Oil 3; all particle emissions (a) and non-volatile particle emissions (b) without the ATS were measured by the EEPs, but all particle emissions with the DPF (c) were measured by the ELPI. Shown is also the vehicle speed by green color and engine load by black color. (d) Time series of the particle number emissions ( $\#/s$ ) measured by the UCPC. (e) Average size distributions of the emissions ( $\#/km$ ) during engine braking for the different engine oils. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)



**Fig. 5.** Time evolutions of the emission of particle number (a) and the size distribution ( $\#/s$ ) (b) measured from the vehicle plume during one 30-70-30 cycle on the road. Shown is also vehicle speed (green color) and engine load (black color). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

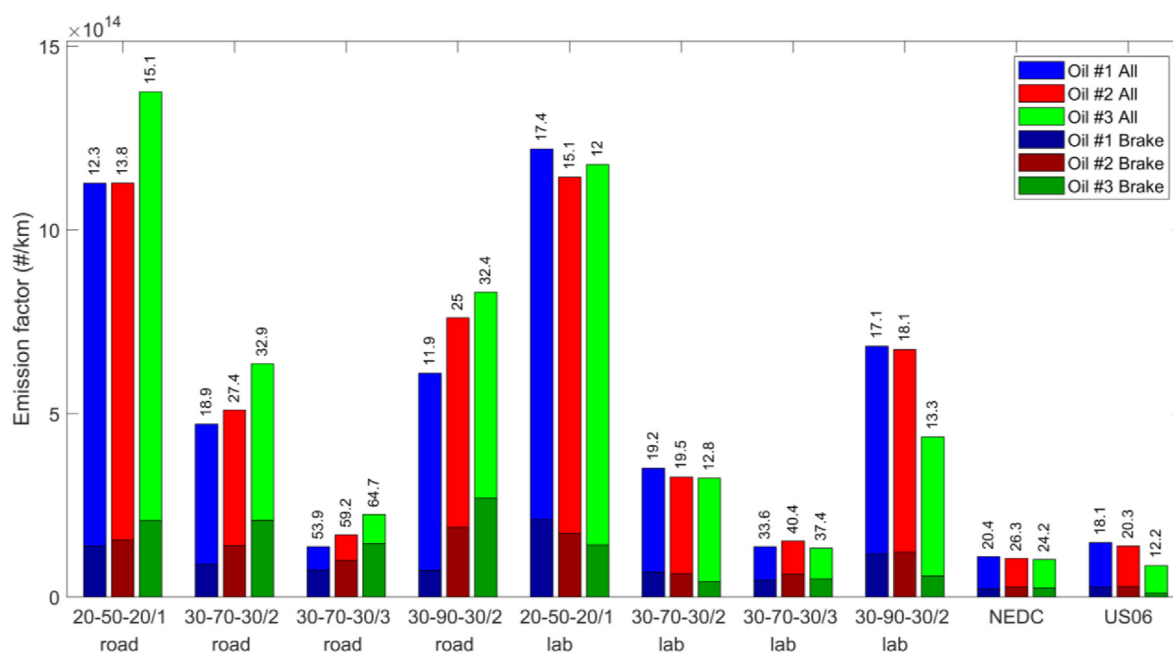
emissions of the fresh particle emissions followed the same mutual order as the fresh particle emissions.

**4. Discussion**

The sampling system used in the laboratory experiments of this study has been designed to mimic the particle processes in atmospheric exhaust dilution but it should be kept in mind that ambient temperature, humidity, residence time and turbulence of the exhaust plume can vary and affect the exhaust’s nanoparticle formation and growth (Keskinen and Rönkkö, 2010). In this study, the results corrected for the dilution ratio from the laboratory and from the on-road tests agree very well in terms of particle number concentration (Fig. 6). This may be caused mostly by the dominating role of non-volatile primary particles; both the laboratory experiments and on-road experiments showed that the engine out

exhaust, i.e., when the DPF was removed from the tailpipe system, contained soot mode particles. In addition, it seems that nanoparticle emissions during engine braking are not sensitive on measurement method, at least in terms of particle number emissions. However, the study underlines the importance to measure the diesel exhaust particle number concentrations over a large particle size range so that also the smallest particles are included in the particle number emission factors.

This study shows that, in addition to the variation in particle number emissions, transient driving conditions affect the size distribution of particles emitted by diesel cars. This is important to note because the potential effect of particles depends on particle size. We observed that while the acceleration conditions are clearly linked with soot particles (mean size > 50 nm) in engine out exhaust, significantly smaller particles (in sizes < 20 nm) are emitted under deceleration conditions conducted by engine



**Fig. 6.** Number emission factors (estimated by the UCPC) of fresh exhaust particles and engine brake particles for the different oils and cycles with no after-treatment system. The number on top of each column indicates the percentage of engine brake particle emission factor (calculated over the whole cycle distance) of the total particle emission factor.

braking. This was very clearly seen in our self-made transient cycle but also under acceleration and deceleration conditions during the official laboratory test cycles NEDC and US06. The nanoparticles emitted during deceleration were affected by the thermodenuder treatment, indicating that semi-volatile compounds participate in their formation and growth. However, it is possible that these particles contained non-volatile cores because a fraction of these particles did not evaporate in the thermodenuder treatment and, from the other hand, the results gathered from laboratory experiments are in line with the results from on-road experiments. Besides, the nucleation of new particles in the cooling dilution process of the exhaust typically requires high exhaust temperature, high oxidation capability of the exhaust after-treatment system, high fuel sulfur content and, based on some previous experiments (e.g. Karjalainen et al., 2014b), exhaust aftertreatment components that can store and release the nucleating compounds efficiently. In this study, the situations that led to the nanoparticle emission events (decelerations) do not fulfill these requirements.

Besides driving conditions, the effects of lubricant oil on particle emissions of the diesel passenger car were investigated. We did not see significant effects on soot mode particles and the effects on nanoparticles during deceleration were moderate, although the particle formation during deceleration can be considered as a lubricant oil driven process. The effect of lubricant oil was seen most clearly in the volatility properties of the particles; the fraction of non-volatile particles was 90–96% with Oil 3, 66–89% with Oil 2, and 63–76% with Oil 1 (Table S3).

Most importantly, this study demonstrates clearly how efficient the DPF is in reducing the real-world particle emissions of diesel passenger car. This effect is seen both in soot particles from fuel combustion (especially during accelerations) and in nanoparticles most likely originated from lubricant oil (emissions seen in this study especially during decelerations). Thus, this study encourages the implementation of efficient DPFs in new geographical and application areas of diesel-powered engines. Together with low sulfur fuels that decrease the nucleation mode particle emissions, DPFs may significantly decrease the particulate air pollutants and

thus benefit both human health and climate.

Significant nanocluster aerosol (NCA) concentrations have been observed in urban areas from road traffic (Rönkkö et al., 2017), in natural gas engine exhaust under high temperature conditions (Alanen et al., 2015), and in exhaust of city busses in real traffic (Järvinen et al., 2019). In the atmosphere, these NCA particles may act as nanosized condensation nuclei for the condensation of atmospheric low-volatile organic compounds, and thus influence human exposure in urban areas. This study indicates that also the non-filtered diesel passenger car exhaust may contain these particles, thus contributing to the atmospheric loading of smallest particles in urban areas.

## 5. Conclusions

In this study, the fresh and primary exhaust emissions from a Euro 5b diesel vehicle were measured under the NEDC, US06 and various transient cycles with three commercial engine lubricant oils on the chassis dynamometer. Furthermore, the real-world emissions during the transient cycles were measured by chasing the same vehicle. Driving conditions and lubricant oil properties strongly affected exhaust particles' number emissions, size distributions and volatility properties. During accelerations and steady speeds, the observed soot mode was non-volatile. During decelerations by engine braking high particle number emissions were observed although the engine was not fueled. The size distribution of the particle emissions was bimodal. The particles in the larger particle mode were non-volatile, whereas the nanoparticles had non-volatile cores with volatile material condensed on them. The origin of these particles was from a lubricant oil driven process and their volatility properties also depended on the used lubricant oil. The results obtained in the laboratory and on-road nicely agreed. Overall, the emission factors varied from  $1.0 \times 10^{14}$  to  $1.3 \times 10^{15}$  #/km. Engine brake emissions contributed 12–65% of total emission, and for the first time this phenomenon was observed for a diesel passenger car. However, with a DPF practically all particles were filtered. This study indicates that in regulated markets where



DPFs are mandated, particles formed during deceleration or otherwise under engine motoring conditions is not a problem. However, the role of these particles in urban air quality can be significant in the countries where the renewal of vehicle fleet is slow (more than a decade) or in several developing countries where DPF enforcing regulations do not exist.

### Declaration of competing interest

The authors declare that they have no competing interests.

### CRediT authorship contribution statement

**Hugo Wihersaari:** Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Visualization. **Liisa Pirjola:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Data curation, Writing - original draft, Writing - review & editing, Visualization, Supervision, Project administration, Funding acquisition. **Panu Karjalainen:** Conceptualization, Methodology, Investigation, Resources, Writing - original draft, Writing - review & editing, Visualization, Supervision, Project administration. **Erkka Saukko:** Methodology, Investigation. **Heino Kuuluvainen:** Methodology, Investigation. **Kari Kulmala:** Resources, Funding acquisition. **Jorma Keskinen:** Conceptualization, Resources, Supervision, Project administration, Funding acquisition. **Topi Rönkkö:** Conceptualization, Resources, Writing - original draft, Supervision, Project administration, Funding acquisition.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2020.114948>.

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