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Reducing particle emissions of heavy-duty diesel vehicles in India: Combined effects of diesel, biodiesel and lubricating oil

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ABSTRACT

Diesel engines contribute significantly to deteriorating air quality. Tightening legislation has led to various technological advances, but developments differ between countries. In India, air quality has not improved and fine particle ($PM_{2.5}$) related premature deaths are predicted to increase. In this study, we characterized the particle emissions of an Indian-manufactured BS IV (Bharat Stage, comparable to Euro emission standards) heavy-duty diesel vehicle and studied the effects of different fuels, fuel blends and lubricating oils. The main aims of the study were to investigate the particle emission dependency on fuel types and fuel blends used in India and to produce useful data for further use (e.g. legislative parties and modeling): emission factors (PN, PM, BC, other chemical compounds), size distributions and volatility of particles. Additionally, the sensitivity of the emissions to the lubricating oil choice was studied. Two lubricating oils, two fossil fuels conforming to BS IV and BS VI emission standards and two biofuel – BS IV fossil fuel blends were tested, one containing Renewable Paraffinic Diesel (RPD) and the other renewable Fatty Acid Methyl Ester (r-FAME). The tests were conducted on a chassis dynamometer (Delhi Bus Driving Cycle, DBDC). Our results show that the emitted particles were in ultrafine particle size range, and both the soot mode particles and smaller nanoparticles were affected by fuels and lubricating oils. The transition from BS IV grade diesel to BSVI was shown to have potential in reducing particle emissions (PN and eBC) of heavy-duty diesel vehicles in India. Blending fossil fuel with biofuel strongly affected particle number emissions, chemical composition, and eBC emissions and the emissions were highly sensitive to biofuel type. Changing the lubricating oil had a comparable magnitude of effect as changing the fuel and the results indicate that in order to reduce particle emissions, a combination of fuel and lubricating oil should be chosen, instead of choosing them separately.

1. Introduction

Particle emissions from road traffic deteriorate the air quality of streets, roads and highways, enveloping urban areas as they disperse from their source (Pant and Harrison 2013; Kumar et al., 2014; Rönkkö and Timonen 2019). Globally, ambient $PM_{2.5}$ (particle mass consisting of sub-2.5 μm particles) is the leading cause of excess mortality (Lelieveld et al., 2020). Diesel engines have been recognized as major particle emitters, and their emissions have been regulated since the early 1990's with gradually stricter legislation. This has led to advancements in aftertreatment systems, fuels, and lubricating oils in order to reduce the emissions below legislated limits. In European countries and more recently in China, this development is one of the main reasons for improved air quality in urban areas (Cheng et al., 2019; Font and Fuller, 2016; Querol et al., 2014). In Europe, the adoption of a particle number emission limit in Euro V and Euro VI standards has practically forced the

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use of diesel particle filters (DPF) in diesel-powered on-road vehicles, which has reduced the solid particle emissions of diesel cars, buses and trucks to very low levels (Grigoratos et al., 2019). However, diesel vehicle emission regulation is not on the same level everywhere in the world and in many highly populated urban areas, air quality has not improved: for example, BS IV (Bharat Stage) emission standards were implemented nationwide in India in 2017 (Lathia and Dadhaniya, 2019), whereas in the European Union the corresponding emission standard EURO 4 had been implemented in 2005 (Querol et al., 2014). Many Indian cities struggle with air quality issues (e.g. Balakrishnan et al., 2019) but the implementation of BS VI in 2020 (corresponding to EURO 6) may improve the situation (Anenberg et al., 2019). Extensive air quality problems and the relatively rapid movement from BS IV emission standard directly to BS VI makes India one of the most interesting countries for investigating the effects of technology on road traffic emissions.

Pandey and Venkataraman (2014) estimated that the emissions of the entire transport sector of India in 2010 were 276 Gg/y of $PM_{2.5}$, 144 Gg/y of Black Carbon (BC), and 95 Gg/y of Organic Carbon (OC). Heavy-duty diesel vehicles were the largest source of $PM_{2.5}$ and BC; however, two-stroke vehicles and old vehicles (pre-2005) were the largest source of OC. In a later review of emission factors for 2013 by Prakash et al. (2020), the estimated contribution of $PM_{2.5}$ had increased slightly from 276 to 355 Gg/y, whereas BC and OC estimations remained almost the same (137 and 106 Gg/y, respectively). Traffic-related emissions are an exceptionally large problem in megacities like Delhi, where population density and traffic volumes are high. For instance, in 2010, outdoor $PM_{2.5}$ and ozone caused an estimated 19 700 premature deaths in Delhi, and an increase to over 50 000 is predicted by 2050 (Lelieveld et al., 2015). The transport sector contributes 16% of the total $PM_{2.5}$ emissions in Delhi and 20% of the emissions in the National Capital Region (NCR) (ARAI, 2021; Teri, 2018).

In addition to air quality, the Indian government has another motive for cutting diesel usage. In India, the road transport sector accounts for 6.7% of annual Gross Domestic Product (GDP) and 28% of annual energy demand. Diesel alone meets an estimated 72% of transportation fuel demand (about 92 billion liters during 2016–2017) followed by petrol (23%) and other fuels such as Compressed Natural Gas (CNG) and Liquefied Petroleum Gas (LPG) (MoSPI, 2018). It has been estimated that diesel demand in the transport sector of India will increase to 158 billion liters by 2026 (International Energy Agency, 2017). India is dependent on import to meet its crude oil demand; during 2017–2018 about 220 million tons were imported (MoSPI, 2019). The increase in demand and the large-scale dependency on imported crude oil are major challenges for energy sustainability.

To combat these challenges, India has modified its vehicle emission norms and fuel quality standards several times during last two decades. Initially, fuel standards aimed to decrease fuel lead content and later standards have added sulphur content limits. India moved to BS VI engine and fuel standards in April 2020, which limits the sulphur content in both gasoline and diesel to less than 10 ppm. The emission standard for heavy duty vehicles under BS VI limits CO, NO_x and PM to 4 g/kWh, 0.46 g/kWh and 0.01 g/kWh respectively, and particle number to 6×10^{11} #/kWh (ARAI, 2021). The government of India has also undertaken several initiatives to address the increasing demand for fossil fuels and the dependency on imported crude oil. Blending biofuel with gasoline and diesel is one such initiative, and has the potential benefit of emission reduction (Ribeiro et al., 2007). However, in 2016, the target for large-scale blending of biodiesel with conventional diesel was not very successful, but India was able to achieve its highest ever ethanol market with a blending rate of 3.3% across the country (Aradhey, 2017). The enforcement of BS VI norms is expected to bring down NOx emissions by 25% in gasoline engines and 68% in diesel engines, whereas PM emissions from diesel engines is expected to decrease by 80% (ICCT, 2016).

Diesel engines emit particles with various compositions, morphologies and sizes, depending on the engine- and vehicle technologies, fuel and lubricant qualities, as well as on driving conditions (Burtscher 2005; Maricq, 2007; Giakoumis et al., 2012; Heikkilä et al., 2009a, 2009b; Karjalainen et al., 2019; Kittelson et al., 2006; Pirjola et al., 2015). Typically, the emitted particle mass from diesel engines is dominated by soot particles composed of solid elemental carbon (EC), semi-volatile hydrocarbons, and sulfuric compounds. The soot particle size distribution of freshly emitted exhaust usually forms a lognormal mode (Harris and Maricq, 2001) with a mean diameter between 30 and 100 nm (Maricq, 2007). While the agglomerated, solid fraction of soot mode particles forms in a high temperature combustion process, the semi-volatile fraction of these particles is linked with the condensation and adsorption processes that take place when the exhaust cools down to ambient temperature. The use of a DPF efficiently reduces the emissions of solid exhaust particles (Lähde et al., 2009). Along with soot mode particles, diesel exhaust emissions can contain a high number of smaller nanoparticles, referred to as the nucleation mode, which can either be solid particles formed in high-temperature conditions (Rönkkö et al., 2007; De Filippo and Maricq 2008; Lähde et al., 2009) or particles condensed from vapors when the exhaust cools down to ambient temperature (Arnold et al., 2012), hence the term *nucleation mode*. In modern diesel engines the adoption of high-pressure fuel injection has decreased soot mode particle emissions but, on the other hand, increased the emissions of smaller exhaust particles (see e.g. Rönkkö et al., 2007). In recent studies, at least two high-temperature formation processes have been proposed for the solid portion of these nanoscale particles, one related to the lubricating oil and the other to the fuel (Kuuluvainen et al., 2020).

Lubricating oil is used to decrease friction between moving parts in an engine. Oil properties, such as chemical composition, can have a major impact on the particle emissions of a diesel engine. For instance, in a study by Sakurai et al. (2003) at least 95% of the volatile component of both nanoparticles and larger particles was found to consist of unburned lubricating oil. A link between lubricating oil and soot formation has also been suggested (Carbone et al., 2019). When using synthetic oil, the PM emissions have been shown to be 19–24% smaller than PM emissions when using mineral oils (Gligorijevic et al., 2006). Canagaratna et al. (2004) found the mass spectra of diesel exhaust nonrefractory components to be dominated by lubricating oil spectral signatures. Furthermore, Vaaraslahti et al. (2005) studied the effect of lubricating oil on nanoparticle formation, and found that the formation of a semi-volatile nucleation mode was highly dependent on the oil, although they were unable to conclude which aspect of the oil in particular was responsible for the differences. The study also found smaller differences (23% at maximum, approximated from a bar graph) in the PM sampled from the raw exhaust (no after-treatment).

Reviews of biodiesel emissions conclude that the effect on PM emissions is generally favorable (Ribeiro et al., 2007; Lapuerta et al., 2008), with an average PM reduction of 20–40% in heavy-duty vehicles due to decreased soot formation and enhanced soot oxidation (Lapuerta et al., 2008). On the other hand, there is a tendency towards increased particle number (Lapuerta et al., 2008). These two trends has been observed for blended fuels as well (Lapuerta et al., 2008). Despite this general consensus, Lapuerta et al. also acknowledged large variability in the results and even some contradicting findings: little to no difference or even increased PM emissions, as well as reductions in particle number emissions. They noted that results may differ depending on engine technologies, the origin of the biofuel or the driving cycle or other differences in measurement methodology. Some of the more recent studies of blended fuels are congruent with the general trends, i.e. increasing the biofuel to fossil fuel ratio reduced PM emissions (Martin et al., 2017; Cheng et al., 2015; Macor et al., 2011; McCaffery et al., 2022) and increased number emissions (Zhu et al., 2010). However, other studies found no difference or even decreased particle number emissions (Book et al., 2015; Young et al., 2012a).

Despite the numerous studies on engine emissions, experimental studies on Indian-manufactured vehicles and fuels are lacking.

Especially blends of fossil fuel and biofuel are of interest, as it is the current direction of the government policies, and previous results have showed high variability in emissions, depending on many parameters. In this experimental study, we measured the particle emissions of an Indian-manufactured heavy-duty diesel vehicle using different fuel and oil combinations, including two different 20% biofuel blends, and two fossil fuels, as well as two lubricants. The fuels and oils were chosen to match Indian traffic characteristics. A driving cycle representative of Indian traffic was also chosen, as driving cycles have a significant influence on emission factors (e.g. Giechaskiel et al., 2015; Pirjola et al., 2019). The measurement setup offers insight to how the emissions of an older diesel engine behave when fueled with newer, low-sulphur fuel and how biodiesel can improve emissions under these driving cycle conditions. The aim of this study is to measure the effect of biofuels on engine particle emissions relevant to India, and to provide emission data in a form which is easy to use by different sectors (e.g., legislative parties, modeling, fuel, and oil technology). For this purpose, we present a detailed set of particle emission indicators: emission factors (PN, PM, eBC, other chemical compounds) and particle size distributions.

2. Methods

The truck (Eicher Pro1059xp, with a 2.956 L E483 CRS diesel engine, BS IV, registered 2017) was operated on a chassis dynamometer. The vehicle was equipped with a diesel oxidation catalyst (DOC). The odometer reading in the beginning of the tests was 60 150 km. The PM limits for this type of truck are 0.02 and 0.03 g/kWh, for European Stationary Cycle and European Transient cycle, respectively. Tests were conducted by driving the Delhi Bus Driving Cycle (DBDC) which includes idling, acceleration, and deceleration periods with a maximum speed of 50 km/h (speed profile visible in Fig. 1). The load profile (vehicle power and tractive force) is visible in the supplement. The first cycle of each measurement day was started with a cold engine (after letting the engine cool overnight) and the rest were driven with a warm engine (steady driving preceded each warm cycle).

All tests were repeated using different combinations of four fuels (or fuel blends) and two lubricating oils. Two fossil fuels fulfilling the emission standards BS IV and BS VI were used as standalone fuels. The BS standard fuel specifications are presented in the supplement (STable 1). The maximum allowed sulphur contents of BS IV and BS VI fuels are 50 and 10 ppm, respectively. Additionally, BS IV fuel was blended with Fatty Acid Methyl Esther -based renewable biofuel (r-FAME) and Renewable Paraffinic Diesel (RPD). Both blends were 80% BS IV fossil fuel. The properties of the fuels are visible in Table 1. The lubricating oils were publicly available market-grade oils, both synthetic, with viscosity grades 15W-40 and 10W-40. From now on, the oils will be referred to as Oil 1 and Oil 2, respectively. More detailed descriptions of the measurement matrix and the lubricating oils are presented in the supplement (STable 2 and STable 3, respectively).

The exhaust aerosol was diluted using Constant Volume Sampling (CVS). From the CVS tunnel, a sample flow was drawn to the measurement instruments. The dilution ratio in the CVS varied between approximately 1.5 and 10, corresponding to high and low exhaust gas flow time periods, respectively. Additional dilution after the CVS yielded a dilution ratio of 4.6–5.6. Dilution air temperature was kept between 25 and 26 °C at all times. A Thermal Denuder (TD) operating at 265 °C was used to remove semi-volatile compounds from the exhaust, enabling the measurement of the non-volatile fraction of particles, or primary particles. The exhaust aerosol also containing the particle mass formed via nucleation and condensation is labeled as “fresh”, referring to the real-world fresh exhaust aerosol that has just undergone the dilution, cooling and subsequent particle formation processes (nucleation and/or condensation) after exiting the tailpipe and entering the surrounding air. In the literature, also other terms such as “solid/non-volatile particles”, and “total particles” are used for the primary- and fresh exhaust particles, respectively. A detailed depiction of the measurement setup is

Table 1
Properties of the fuels used in the tests.

	BS IV fossil fuel	BS VI fossil fuel	r-FAME	RPD
Density (kg/m3)	815–845	810–845	882	770–790
Viscosity (cSt at 40°C)	2–4.5	2–4.5	4.9	2
Flash point (°C)	35	35	>61	>61
Cetane number	51	51	56	>70
Total Sulphur (mg/kg)	32.6	8.3	38.5	<5
Aromatics (% m/m)	8	8	0.7	<1
FAME content (% v/v)	7	7	75.8	0
Ash (%)	0.01	0.01	<0.001	<0.001

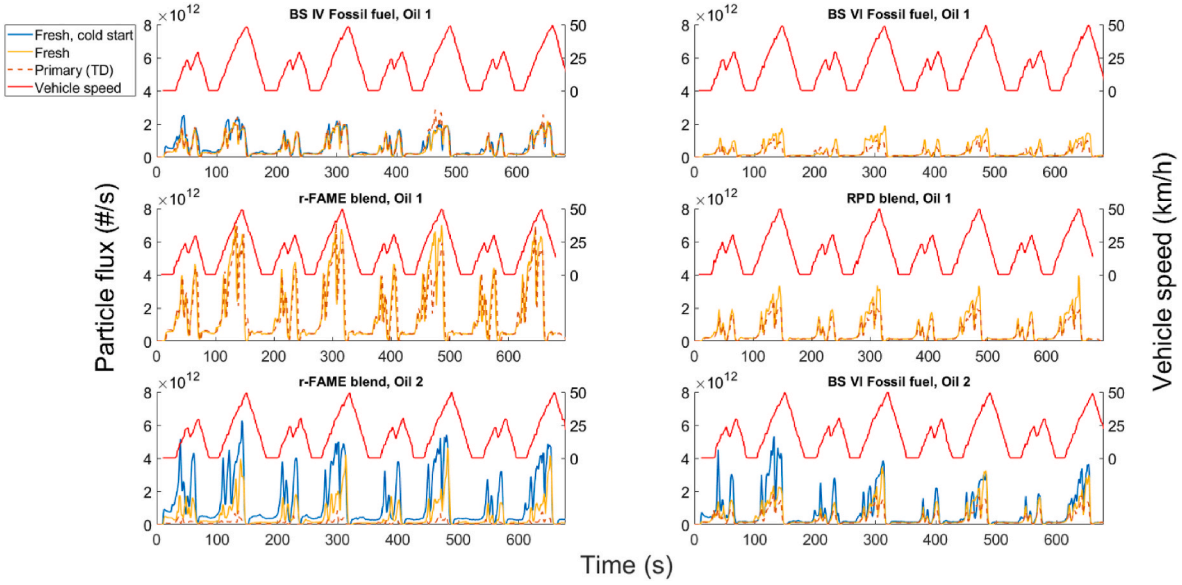

Fig. 1. Time series of fresh and primary particle number flux, measured with a CPC, and vehicle speed with different fuels and lubricating oils and with cold and hot engine. Cold cycle was performed three times (BS IV fossil fuel + Oil 1, r-FAME blend + Oil2 and BS VI fossil fuel + Oil 2).

available in the supplement.

The particle emissions were characterized in terms of particle number concentration, size distribution, chemical composition and eBC (equivalent black carbon) mass concentration using a Condensation Particle Counter (CPC, TSI model 3776, 2.5 nm cutoff), an Electrical Low-Pressure Impactor (ELPI, Dekati ELPI+, size range of 6–9910 nm), a Time-of-Flight Aerosol Chemical Speciation Monitor (ToF-ACSM, Aerodyne) and an aethalometer (Magee Scientific model AE-16), respectively. The filter-loading effect of the aethalometer was corrected according to Virkkula et al. (2007) and the detailed description of the correction is presented in the supplement. It should be noted that the ACSM is limited to measuring non-refractory substances and has a particle measurement range of approximately 40–1000 nm. CO_2-concentration was measured from the tailpipe, CVS and before the measurement instruments for dilution ratio (DR) determination. Other relevant parameters such as vehicle speed, traveled distance, fuel consumption, vehicle power and CVS flow were also recorded. In addition to the previously mentioned measurements with high time resolution, the regulated emissions of CO, HC, NOx, CO_2, and PM were also determined (visible in the supplement).

In the case of measurement points where the TD was used, the particle concentrations were corrected for losses in the TD using the loss curves presented by Heikkilä et al. (2009)a,b (the TD used in the tests was the same unit). The specifics of the loss correction calculation are presented in the supplement. The eBC concentration measured with the aethalometer was corrected with a constant penetration factor of 0.76, which is the maximum level of the denuder penetration curve, plateauing at the particle size of 51 nm.

In some cases, a measurement point was repeated once. The replicate was conducted directly after the first measurement. The differences in calculated results from these repetitions were used in estimating the measurement uncertainty. More details about the data processing and calculation of results and measurement uncertainty are presented in the supplement file.

3. Results and discussion

As many of the general features visible in the results are already established in the research field, they will not be repeated in the discussion of each independent result figure or table. Instead, the observed general features are shortly described in this paragraph. Generally, operating a cold engine has been shown to emit more particles in terms of number and mass (e.g, Fontaras et al., 2009), due to inefficient operation of engine aftertreatment system and sub-optimal engine conditions. As explained in detail in the introduction section, diesel exhaust has been shown to often have potential to form particle mass via nucleation and condensation as the raw exhaust dilutes and cools. Therefore the thermal treatment of the sample (TD) is expected to remove a fraction of the particles.

Fig. 1 presents the time series of particle number emissions (fluxes, in units of #/s). The subfigures show different combinations of fuel and lubricating oil, while the colors indicate whether the sample was conditioned with TD or not (primary and fresh exhaust particles, respectively), and whether the engine was cold or warm before the start of the test cycle.

Some general trends were observed in all the time series seen in Fig. 1. First, the particle emissions increased with acceleration and decreased with deceleration in each case, causing high variation in the instantaneous particle emissions. Overall, it spanned several orders of magnitude, from approximately 10^8 to 10^{12} particles per second. In addition, several separate but smaller emission spikes occurred during the accelerations, likely caused by gear shifts of the vehicle.

The effect of cold start on particle number emissions (increased emission) was substantial in both cases when the engine was used with Oil 2, but not so prevalent with the combination of BS IV fossil fuel and Oil 1. It is possible, however, that the difference in this case lies in particle sizes that the CPC could not detect (<2.5 nm). In the case of BS VI fossil fuel and Oil 2, the difference between the emissions of cold and warm starts was largest in the beginning of the driving cycle, diminishing towards the end, whereas in the case of r-FAME blend the difference was clear throughout the cycle.

The difference between the fresh exhaust particle emissions and primary particle emissions was largest in the case of r-FAME blend with Oil 2. During engine idling after decelerations the fluxes were on the same level: the difference between the two arose during accelerations. The highest peaks were recorded with the r-FAME blend and Oil 1, for both fresh and primary particles.

Fig. 2 presents particle number size distributions averaged over the driving cycles, measured with ELPI+. The subfigures represent different fuel-oil combinations and include measurement points with cold and warm engine at startup and with or without the TD (primary and fresh exhaust particles, respectively). The distributions represent the concentration in the CVS. Overall, Fig. 2 shows that the average particle size distributions were bimodal with a soot mode (60–80 nm) and a small particle mode (<40 nm). The shape of the mode of small particles indicates that the elevated concentrations extended beyond the lower size limit of the ELPI+ in all cases. In general, a bimodal particle size distribution is typical for heavy-duty diesel vehicle exhaust (e.g. Karjalainen et al., 2014; Heikkilä et al., 2009b; Young et al., 2012b).

As was seen in Fig. 1, in two cases (of BS IV fuel + Oil 1 and r-FAME blend + Oil 1) the total number concentrations of fresh exhaust and primary particles were equal. However, as is seen in Fig. 2, the size distributions obtained with r-FAME blend and Oil 1 are not equal (the fresh exhaust particle size distribution is missing for BS IV fuel + Oil1). This discrepancy between the two instruments is likely explained by the particles below the ELPI + lower size limit. A size distribution comparison figure containing the primary particle modes is visible in the supplement and the soot modes will be discussed in detail later in the article.

The effects of fuels and lubricant oils on the average particle size distributions were not straightforward. Comparing the fresh exhaust particle size distributions obtained with Oil 1 in use, the RPD blend produced the smallest sub-40 particle mode whereas the r-FAME blend produced the largest. It should be noted, however, that the fresh exhaust particle size distribution is missing for the test with BS IV fuel and Oil 1. Similarly in the case of primary particles, the r-FAME blend yielded a sub-40 nm mode larger than others, while the smallest mode was produced by the RPD blend. BS IV fuel produced a slightly larger (+55%, within limit of uncertainty) sub-40 nm mode than BS VI. The differences in the sub-40 nm modes can be quantified by looking at the particle number concentrations of ELPI stages 1–3 (corresponding to particle diameters of approximately 10–41 nm), presented in Table 2.

When Oil 2 was in use, using the r-FAME blend resulted in a larger (compared to BS VI fossil fuel) fresh exhaust particle mode in the sub-40 nm region, both with cold (+140%) and warm (+240%) engine. The sub-40 nm primary particle modes of BS VI fossil fuel and r-FAME blend have a dissimilar shape: with BS VI fossil fuel the concentration decreases towards the lower size limit, whereas with r-FAME blend it increases.

Two of the fuels (BS VI and r-FAME blend) were paired with two lubricating oils in order to study how the lubricating oil affects the emissions. In both cases, oil 1 resulted in more (+48% and +50%, respectively, within limit of uncertainty) sub-40 nm fresh exhaust particles. With r-FAME blend, Oil 1 resulted in more (+210%) sub-40 nm primary particles, whereas with BS VI fossil fuel the effect was opposite, although the difference in concentration was not so prominent (−20%, within limit of uncertainty).

Fig. 3 shows emission factors for particle number (particles emitted per kilogram of fuel consumed, $\#/kg_{fuel}$), the error bars representing the measurement uncertainty, the determination of which is described in the supplement. The emission factors presented in Fig. 3, as well as emission factors in units of #/km, #/kWh, are shown in Table 3.

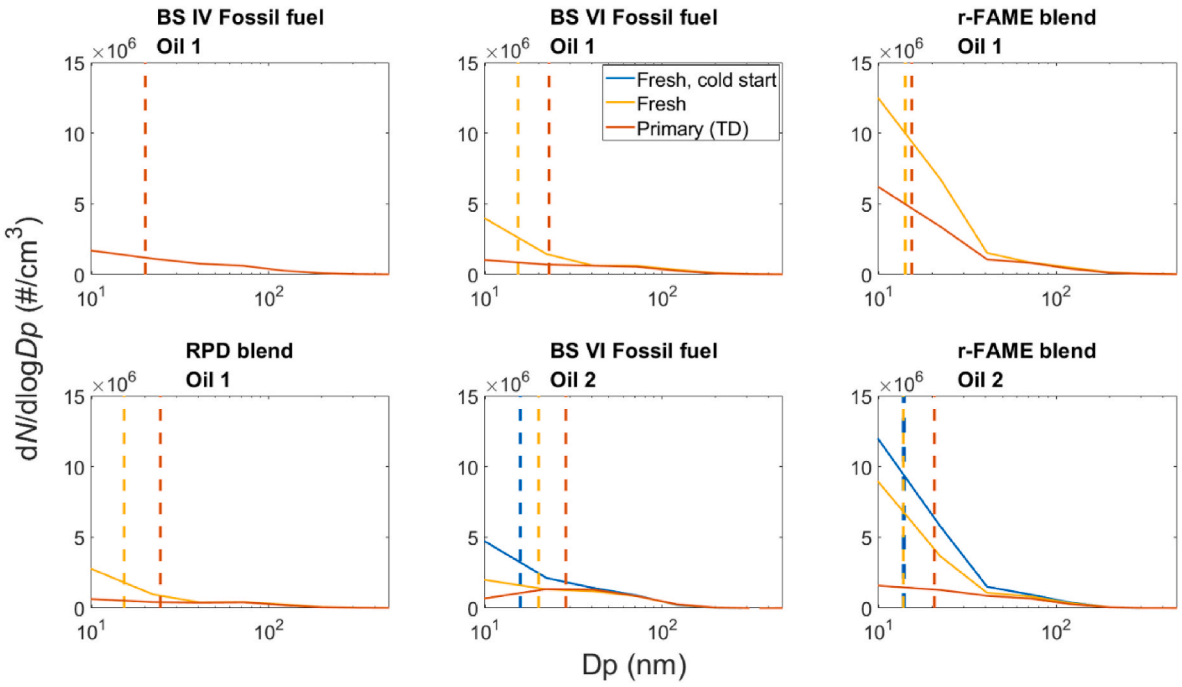

Fig. 2. Exhaust particle number size distributions measured with ELPI+ and averaged over the cycle. Concentrations are corrected for the dilution ratio to the concentrations in the CVS. Dashed lines represent the geometric number mean diameter. A version with a logarithmic y-axis visible in the supplement.

Table 2
Particle number concentrations in ELPI + stages 1–3 (corresponding to particle diameters of approximately 10–41 nm), and in stages 4–14 (corresponding to particle diameters of approximately 72–7300 nm).

	Stages (particle sizes)	Number concentration (10^6 #/cm^3)		
		Fresh, cold start	Fresh	Primary (TD)
BS IV Fossil fuel, Oil 1	1-3 (10–41 nm)	–	–	1.2 ± 0.5
	4-14 (72–7300 nm)	–	–	0.23 ± 0.08
BS VI Fossil fuel, Oil 1	1-3 (10–41 nm)	–	2.3 ± 0.8	0.79 ± 0.27
	4-14 (72–7300 nm)	–	0.25 ± 0.09	0.21 ± 0.08
r-FAME blend, Oil 1	1-3 (10–41 nm)	–	7.6 ± 2.6	3.9 ± 1.4
	4-14 (72–7300 nm)	–	0.35 ± 0.12	0.32 ± 0.11
RPD blend, Oil 1	1-3 (10–41 nm)	–	1.6 ± 0.6	0.48 ± 0.17
	4-14 (72–7300 nm)	–	0.18 ± 0.06	0.16 ± 0.06
BS VI Fossil fuel, Oil 2	1-3 (10–41 nm)	3.0 ± 1.1	1.5 ± 0.6	0.99 ± 0.34
	4-14 (72–7300 nm)	0.26 ± 0.09	0.26 ± 0.09	0.26 ± 0.09
r-FAME blend, Oil 2	1-3 (10–41 nm)	7.1 ± 2.5	5.1 ± 1.8	1.3 ± 0.5
	4-14 (72–7300 nm)	0.32 ± 0.11	0.28 ± 0.10	0.23 ± 0.08

Additional emission factors for PM1 and primary PN (>23 nm), calculated from the ELPI size distributions, and regulated emissions (PM and gaseous compounds), are available in the supplement.

The order of magnitude of the obtained results (0.34–4.8 × 10^{15} for primary particles and 1.1–5.0 × 10^{15} for fresh exhaust particles) is similar to previous studies. In general, particle number emission factors for heavy-duty diesel vehicles have been determined with a multitude of measurement methods and instrumentation. Roadside and chase measurement results presented by Westerdahl et al. (2009), Ban-Weiss et al. (2009), Ban-Weiss et al. (2010) and Saari et al. (2016) fall between 1 × 10^{15} and 11 × 10^{15} #/kg$_{fuel}$ (fresh exhaust aerosol, particles larger than 3–6 nm). Beddows and Harrison (2008) estimate a fleet average fresh exhaust particle number emission factor for heavy-duty vehicles to be 7.06 × 10^{14} #/km. For Euro 4 class heavy-duty vehicles in urban driving, Vouitsis et al. (2017) present emission factors of 0.673 × 10^{14}–2.240 × 10^{14} #/km for fresh exhaust particles and 0.467 × 10^{14}–1.550 × 10^{14} #/km for primary particles, depending on vehicle class (7.5 t to 14 t). Primary particle emission factors as low as PN 6 × 10^{10} #/km have been reached with EURO 6 class HD vehicles in on-road PEMS measurements (Giechaskiel et al., 2015). Thus, significantly lower emission factors are achievable by best available technology than what were measured in our study.

In the cases with Oil 1, the use of stricter-standard fossil fuel (BS VI vs. BS IV) reduced the fresh particle number emissions of the studied vehicle by approximately 40% and primary particle emissions by approximately 60%. Adding RPD to BS IV fuel elevated the number of emitted fresh exhaust particles by approximately 30% and reduced the number of emitted primary particles slightly (<20%), whereas the addition of r-FAME resulted in significantly higher emissions of both fresh exhaust- and primary particles (>190% and >150% increase, respectively). In the cases with Oil 2, r-FAME blend produced more fresh exhaust particles (>15%) but less primary particles (about 70% less) than BS VI fossil fuel. It should be noted, however, that some of the mentioned differences fall withing the determined limits of uncertainty.

Looking at the effect of changing the lubricating oil while keeping the fuel same, with BS VI fuel in use, approximately 30% more fresh exhaust- and primary particles were emitted with Oil 2. When r-FAME blend was in use, switching from Oil 1 to Oil 2 resulted in decreases in the emitted fresh and primary particles (decreases of more than 60% and 90%, respectively). The differences were within the determined limits of uncertainty for BS VI fuel, but not for r-FAME. Interestingly, the effect of changing from Oil 1 to Oil 2 was opposite with r-FAME and BS VI fossil fuel. This is a possible indicator for synergistic effects with the chosen fuels and oils.

The exact mechanisms that govern the effects of how the properties of lubricating oil affect the particle formation (in conjunction with the fuel) cannot be deduced from this data and they remain a topic for future

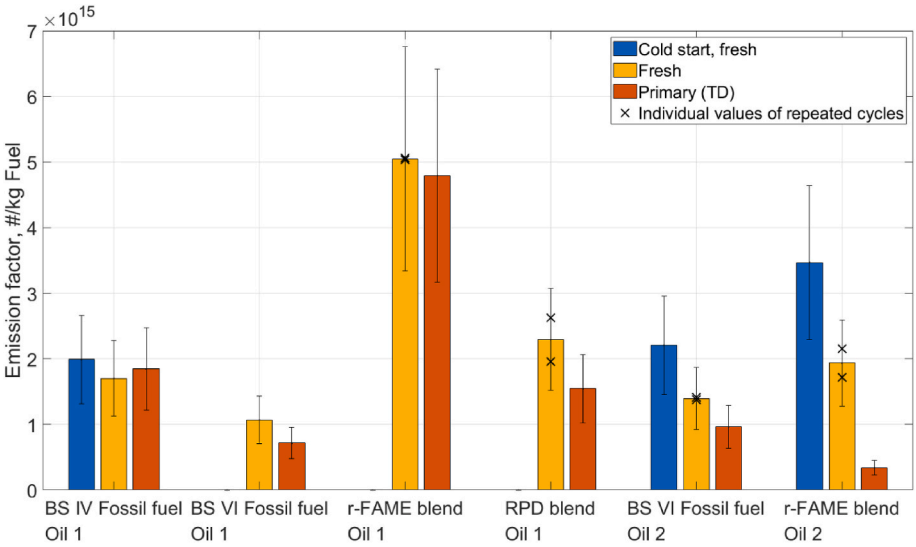

Fig. 3. Particle number emission factors calculated from the particle number concentrations measured by CPC (cut-off size 2.5 nm). Similar figures in units of #/km and #/kWh visible in the supplement.

Table 3
PN (>2.5 nm) emission factors.

Fresh/Primary	Unit	BS IV Fossil fuel, Oil 1	BS VI Fossil fuel, Oil 1	r-FAME blend, Oil 1	RPD blend, Oil 1	BS VI Fossil fuel, Oil 2	r-FAME blend, Oil 2
Fresh (cold start)	10^{14} #/km	1.4 ± 0.49	–	–	–	1.6 ± 0.56	2.8 ± 0.96
	10^{15} #/kg_{fuel}	2.0 ± 0.67	–	–	–	2.2 ± 0.75	3.5 ± 1.2
	10^{15} #/kWh	1.1 ± 0.34	–	–	–	1.1 ± 0.36	1.9 ± 0.61
Fresh	10^{14} #/km	1.2 ± 0.42	0.76 ± 0.27	3.6 ± 1.3	1.5 ± 0.54	1.0 ± 0.36	1.3 ± 0.45
	10^{15} #/kg_{fuel}	1.7 ± 0.58	1.1 ± 0.37	5.0 ± 1.8	2.3 ± 0.78	1.4 ± 0.48	1.9 ± 0.66
	10^{15} #/kWh	0.86 ± 0.27	0.56 ± 0.18	2.6 ± 0.81	1.1 ± 0.35	0.74 ± 0.24	0.89 ± 0.28
Primary (TD)	10^{14} #/km	1.3 ± 0.45	0.53 ± 0.19	3.5 ± 1.3	1.1 ± 0.37	0.70 ± 0.25	0.24 ± 0.083
	10^{15} #/kg_{fuel}	1.9 ± 0.63	0.72 ± 0.25	4.8 ± 1.7	1.5 ± 0.53	0.96 ± 0.33	0.34 ± 0.12
	10^{15} #/kWh	0.95 ± 0.30	0.38 ± 0.12	2.6 ± 0.81	0.81 ± 0.26	0.50 ± 0.16	0.17 ± 0.053

research, partly because we are missing the information of the additives used in the oils. Additionally, the knowledge on how individual oil properties affect particle emissions and what are the processes that govern these effects, is scarce. Oil 1 had a slightly higher viscosity (14.45 vs 13.8 cSt at 100 °C) and a slightly higher sulphate content (1.4 vs 0.93 %w) than Oil 2.

Viscosity of the lubricating oil may affect engine operation and therefore the combustion conditions, affecting the particle formation without participating in the combustion process itself. Additionally, viscosity is the main factor that determines oil consumption and more oil will be present in the combustion chamber when the viscosity is lower (Wang et al., 2017). It has been shown to affect particle morphology and graphitization degree (Wang et al., 2017), as well as the emitted particle size distribution (Dong et al., 2013). Wang et al. (2021) found that the sulphate in the oil affects the molecular composition of the emitted particles. According to Pirjola et al. (2015), oil additives, such as Zn, Mg, P and S, positively correlate with particle number emissions. Additionally, they state that the particle number emissions are especially sensitive to changes in oil properties during transient operation. In their study they report particle emission reductions of up to 99% by just switching the oil. However, the study was conducted on a GDI engine, where the particle formation processes may differ from the engine used in this study. Lähde et al. (2014) suggest that the core particles in the diesel engine exhaust of their study were metallic ash from the lubricant oil, formed in high-temperature in-cylinder processes independent of the soot particles. Also Kim et al. (2020) conclude that the metal and non-metal additives in the oil affect the small particle formation. Kuuluvainen et al. (2020) studied the engine-out exhaust of a non-road diesel engine and found the size distribution of the particles to be trimodal; one mode consisting of lubricating oil -originated primary particles. They suggest that the formation processes of this lubricating oil -originated mode are independent of the fuel-originated mode formation. Our results challenge this view of independent formation processes. Combustion conditions are dependent on the fuel, and the e.g. temperature has been shown to affect the evaporation of oil from the cylinder walls (Tornehed and Olofsson, 2022), which is one way of mixing the fuel and oil effects.

Adding to the complexity of interpreting the results, emissions of nanoparticles (especially number-wise) could be sensitive to changes in the soot mode, due to differences in the condensation- and/or agglomeration sink. For example, a higher concentration of soot mode particles could hinder, or even completely prevent formation of particles via nucleation and decrease the number of primary nanoparticles via agglomeration. Therefore, the emission factors need to be looked at in conjunction with the size distributions. Two cases stand out from the results in terms of changes in the particle emissions: The addition of r-FAME to BS IV diesel and the oil switch combined with r-FAME blend. The addition of r-FAME and switching from Oil 2 to Oil 1 both elevated the number emissions and according to the size distributions, especially in the nanoparticle size range. However, according to size distribution data in Fig. 2 and Table 2 (see also supplement for a better comparison of soot modes), the soot mode concentration was elevated simultaneously, ruling out this mechanism.

Fig. 4 shows the emission factors for the mass of chemical compounds of particles (μg/kg_{fuel}) measured in this study (tabulated data in the supplement). eBC emission factors were calculated from the data obtained with the aethalometer, whereas the rest of the compounds (organics, ammonium, chloride, nitrate, and sulphate) were obtained

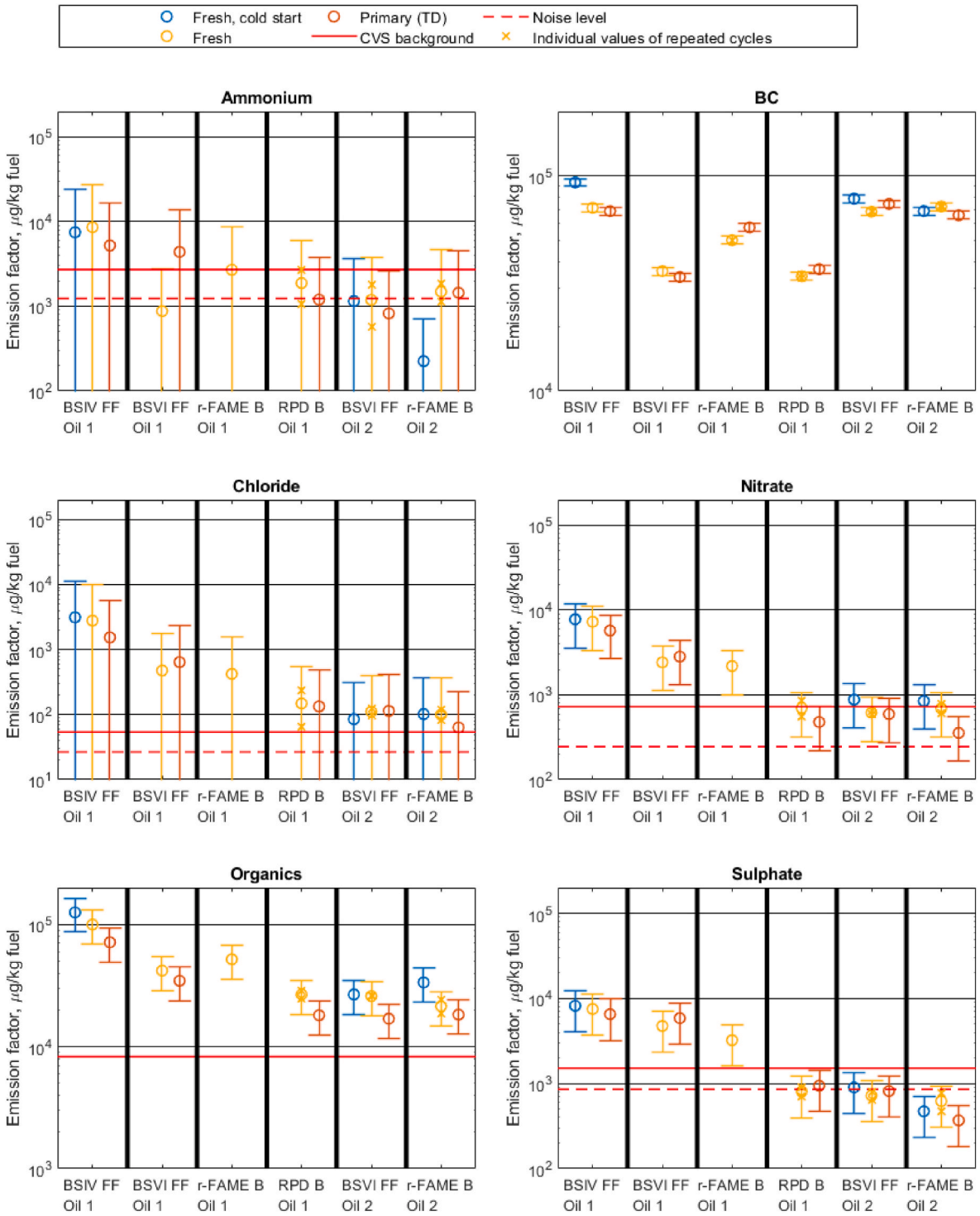

Fig. 4. Emission factors for different chemical components (ACSM and aethalometer (eBC)). Background levels in the CVS and the instrument noise level are also shown (red lines). Emission factors in units of μg/km and μg/kWh visible in the supplement, both as tables and figures. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

from the ACSM measurements. The CVS background levels for each compound are also shown, along with the noise level of the instrument. Results below or at CVS levels are considered contamination from the CVS air. The error bars represent the measurement uncertainty, the determination of which is presented in the supplement. While looking at the effects of fuels and lubricating oils based on the composition data, it should be kept in mind that the largest differences (particle number-wise) were in the nanoparticle size range, which is mostly out of the measurement range of the ACSM. In addition, the mass-based particle composition data emphasize the larger particle size range than particle number-based analyses above. The sub-40 nm nanoparticle contribution to the mass was estimated from the average size distributions and was found to be less than 15% in all cases.

In several cases the measurement uncertainty was substantially high, and the emission factors often fell below the determined CVS background and/or noise levels. Organics and eBC were the only substances

that had a high signal in every case, which is consistent with previous studies of diesel engine emissions (Pirjola et al., 2019). Nitrate and sulphate signals exceeding the background levels were recorded with three combinations of fuel and oil: BS IV fossil fuel, BS VI fossil fuel and r-FAME blend with Oil 1. Some ammonium and chloride were also detected in these cases, but while the mean value was above the background level, the uncertainty for these compounds was high.

The eBC emission factors varied between 33.5 and 93.5 mg/kg_{fuel}, corresponding to 22.5 and 66.0 mg/km. Typically, BC emission factors have been determined as averages for large populations of vehicles and the availability of BC emission factors for vehicles on a specific emission level is poor. The emission factors determined in this study are generally lower than, but on a comparable level to, different fleet average estimations. E.g. S. S. Park et al. (2011) determined fleet average BC emission factors in roadside measurements and present mean values of 170–970 mg/kg_{fuel}, and median values of 70–170 mg/kg_{fuel}, depending on driving conditions. Dallmann et al. (2012) present a fleet average BC emission factor as well (540 mg/kg_{fuel}), emphasizing the contribution (47%) of few vehicles with high emissions ("high emitters").

With Oil 1 in use, the lowest eBC emission factors were obtained with BS VI fossil fuel and RPD blend. In other words, adding RPD to older-grade BS IV fossil fuel lowered the eBC emissions to a level obtained with the stricter-grade BS VI fossil fuel. The lowered aromatics content can be a soot-reducing factor (Arad et al., 2017), but with r-FAME addition the decrease of eBC was not so prevalent, even though the aromatic content was on a similar level to RPD. The highest eBC emissions were obtained with BS IV fossil fuel. In the particle number size distributions (see also SFig. 5 in the supplement for a clearer comparison) the soot modes with BS VI fossil fuel and RPD blend were the smallest, as with the aethalometer results. However, the highest soot mode concentration in the size distribution measurements was with the r-FAME blend, and the BS IV (highest soot concentration in the aethalometer results) was the second highest, as seen in Table 2. The emission factors of organics behave similarly to eBC: RPD blend had the lowest emissions and BS IV fossil fuel the highest.

Switching from Oil 1 to Oil 2 increased the eBC emission factors both with BS VI fossil fuel and r-FAME blend (approximately 80–120% and 5–40%, respectively), the effect being more prominent with the former. Looking at the size distributions, with BS VI fossil fuel Oil 2 resulted in higher concentration of particles in the soot mode size range (about 30% higher) but with r-FAME blend, the effect was the opposite (about 30% reduction). It is noteworthy that the effect of oil on the soot modes and eBC emission factors was of similar magnitude as the effect of fuel. While eBC emissions increased, the emissions of organics were lowered by approximately 40–50% (BS VI fuel) and 60% (r-FAME blend) with the switch from Oil 1 to Oil 2.

4. Conclusions

In Table 4, the relative increases or decreases in PN, eBC and organics caused by switching the fuel or oil are assembled for a quick overview of results. The main results obtained in this study are.

1. **The transition from BS IV grade diesel to BS VI diesel** has significant potential in reducing particle emissions of heavy-duty diesel vehicles in India. The older-grade fuel yielded 59% and 158% higher fresh exhaust- and primary PN emission factors, respectively. The eBC emissions were twice as high and the emissions of organics were 139% and 107% higher (fresh exhaust- and primary particles, respectively). Lowering the sulphur content of the fuel has been shown to decrease the emissions of organic carbon and elemental carbon (e.g. Zhang et al., 2009), as well as the number of nanosized particles (Ristovski et al., 2006).
2. **Blending fossil fuel with r-FAME and RPD** strongly affected particle number emissions, chemical composition, and eBC emissions and the emissions were highly sensitive to biofuel type. Adding r-FAME to BS IV diesel increased the PN emissions of primary- and fresh exhaust particles by 196% and 160%, respectively, whereas the addition of RDP resulted in a 35% increase in fresh exhaust particles and a decrease of 16% in primary particles. Both of the blends yielded lower emissions of organics (48–75% decrease) and eBC (16–52% decrease) than standalone BS IV fuel, following the overall trend of reduced PM emissions as showed by Lapuerta et al. (2008). According to Lapuerta et al. (2008), the majority of studies have reported increases in the number of small particle emissions with biodiesel. The results reported by e.g. Young et al. (2012a) and Book et al. (2015) deviate from this trend. Soot emissions have been shown to decrease with biodiesel use (Ribeiro et al., 2007; Pirjola et al.,

Table 4
Changes in the obtained emission factors (PN, eBC and organics) when changing the fuel or oil. Green cell colour (or ratio smaller than 1) refers to a decrease and red (or ratio greater than 1) to an increase.

	Instrument →		CPC	ACSM	Aethalometer
	Particle emission property →		$PN_{>2.5\ nm}$	Organics	eBC
Description of change	Fuel/Oil switch	Fresh/Primary	Ratio of emission factors		
Changing the fuel while using Oil 1	BS VI→BS IV Oil 1	Fresh	1.59 ± 0.77	2.39 ± 1.05	1.97 ± 0.12
		Primary	2.58 ± 1.24	2.07 ± 0.91	2.03 ± 0.13
	BS VI→r-FAME blend Oil 1	Fresh	4.73 ± 2.27	1.24 ± 0.55	1.39 ± 0.09
		Primary	6.70 ± 3.22	-	1.70 ± 0.11
	BS VI→RPD blend Oil 1	Fresh	2.15 ± 1.03	0.64 ± 0.28	0.95 ± 0.06
		Primary	2.16 ± 1.04	0.52 ± 0.23	1.09 ± 0.07
Changing the fuel while using Oil 2	BS VI→r-FAME blend Oil 2	Fresh (cold start)	1.57 ± 0.76	1.25 ± 0.55	0.88 ± 0.06
		Fresh	1.39 ± 0.67	0.83 ± 0.37	1.06 ± 0.07
		Primary	0.35 ± 0.17	1.08 ± 0.48	0.89 ± 0.06
Changing the oil	BS VI Oil 1→Oil 2	Fresh	1.31 ± 0.63	0.62 ± 0.28	1.89 ± 0.12
		Primary	1.35 ± 0.65	0.49 ± 0.22	2.19 ± 0.14
	r-FAME blend Oil 1→Oil 2	Fresh	0.38 ± 0.19	0.41 ± 0.19	1.43 ± 0.09
		Primary	0.07 ± 0.04	-	1.14 ± 0.07
Adding biodiesel to BS IV fuel	BS IV→r-FAME blend Oil 1	Fresh	2.96 ± 1.43	0.52 ± 0.23	0.71 ± 0.05
		Primary	2.60 ± 1.25	-	0.84 ± 0.06
	BS IV→RPD blend Oil 1	Fresh	1.35 ± 0.65	0.27 ± 0.12	0.48 ± 0.03
		Primary	0.84 ± 0.41	0.25 ± 0.12	0.54 ± 0.04

2019). Cheng et al. (2015) and Martin et al. (2017) report lower emissions of organic carbon with biofuel blends. As a noteworthy detail, the addition of RPD to older-grade BS IV fossil fuel lowered the eBC emissions to a level obtained with the stricter-grade BS VI fossil fuel.

3. **Changing the lubricating oil** had a comparable magnitude of effect as changing the fuel. With r-FAME blend, the oil switch (Oil 1 to Oil 2) decreased the number of emitted fresh exhaust- and primary particles by 62% and 93%, respectively. The emission of organics was decreased by 59% (fresh exhaust particles), whereas the eBC emissions were increased (14–43%). With BS VI fuel, the same oil switch increased the emitted PN by 31% and 35% (fresh exhaust- and primary particles, respectively). Like with r-FAME blend, the emissions of organics were decreased (38–51%). eBC emissions increased more (89–119%) with r-FAME than with BS VI fuel. Lubricating oil has been shown to affect PM emissions (Vaaraslahti et al., 2005; Gligorijevic et al., 2006), the number of fresh exhaust particles (Vaaraslahti et al., 2005; Ovaska et al., 2020) and chemical composition (Canagaratna et al., 2004) of particles. Kuuluvainen et al. (2020) found a lubricating-oil-only primary particle mode in the exhaust of a diesel engine.
4. In addition to the individual effects, **the fuels and oils may also have had synergistic effects.** We arrived at this conclusion by looking at the effect of changing the fuel from BS VI fuel to r-FAME blend with the two tested lubricating oils. With Oil 1, the fuel change resulted in increases of eBC (39–70%), organics (24%) and most notably PN (373–570%). However, the same fuel change with Oil 2 resulted in a significant decrease (65%) of primary PN and a smaller increase (or even decrease) of fresh exhaust PN, eBC and organics. To our knowledge, such phenomenon has not been previously reported.

This data set is not complete enough to examine the particle formation processes behind this phenomenon. However, just knowing that the particle emissions can be this sensitive to the combination of fuel and lubricating oil would mean that regarding particle emissions, a combination of fuel and lubricating oil should be chosen, instead of choosing them separately.

5. **We provide a detailed set of particle emission indicators** (emission factors of PN, eBC, organics and other chemical compounds, as well as size distributions, for both fresh exhaust- and primary particles), which can be useful in any field of work related to air quality (e.g., modeling, legislation and fuel and oil technology). The knowledge of particle emission characteristics and the effects of changing any relevant parameters (such as fuel or lubricating oil) is crucial when optimized strategies for reducing emissions are devised.

We suggest further investigation of the effects of biofuels on emissions, for example, the possible high nanoparticle emissions linked here with the addition of r-FAME to fossil diesel. The mechanisms of the synergistic effects of fuels and lubricating oils fall beyond the scope of this study and should also be examined in the future. We would like to once again emphasize the effect the lubricating oil might have on particle emissions: with r-FAME blend as fuel, the fresh exhaust- and primary particle number emissions were reduced by 62% and 93%, respectively. As a concluding remark, the air quality impact of heavy-duty traffic in India could be significantly lowered with already available technologies, that are simply not implemented yet on a large scale, and as the vehicle fleet slowly renews and better after-treatment systems become more common, simple changes to fuel and lubricating oil can make quick and significant improvements to air quality right now, with the existing fleet.

CRediT authorship contribution statement

Sampsa Martikainen: Formal analysis, Investigation, Writing – original draft, Visualization. **Laura Salo:** Investigation, Writing – original draft, Visualization. **Heino Kuuluvainen:** Investigation, Writing – original draft. **Kimmo Teinilä:** Investigation, Writing – review & editing. **Rakesh K. Hooda:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Arindam Datta:** Project administration, Writing – original draft, Writing – review & editing. **Ved Prakash Sharma:** Project administration, Resources, Writing – review & editing. **Hafizur Rahman:** Project administration, Resources, Writing – review & editing. **Sanjukta Subudhi:** Conceptualization, Resources, Writing – review & editing. **Prashant Kumar:** Project administration, Resources, Writing – review & editing. **Panu Karjalainen:** Supervision, Writing – review & editing. **Jorma Keskinen:** Supervision, Writing – review & editing. **Hilkka Timonen:** Conceptualization, Writing – review & editing, Funding acquisition. **Antti Hyvärinen:** Conceptualization, Funding acquisition, Writing – review & editing. **Topi Rönkkö:** Conceptualization, Methodology, Resources, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Parts of the data used in the study might be classified as confidential by the project partners.

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Appendix A. Supplementary data

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