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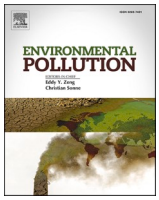

Towards zero pollution vehicles by advanced fuels and exhaust aftertreatment technologies☆

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ABSTRACT

Vehicular emissions deteriorate air quality in urban areas notably. The aim of this study was to conduct an in-depth characterization of gaseous and particle emissions, and their potential to form secondary aerosol emissions, of the cars meeting the most recent emission Euro 6d standards, and to investigate the impact of fuel as well as engine and aftertreatment technologies on pollutants at warm and cold ambient temperatures. Studied vehicles were a diesel car with a diesel particulate filter (DPF), two gasoline cars (with and without a gasoline particulate filter (GPF)), and a car using compressed natural gas (CNG). The impact of fuel aromatic content was examined for the diesel car and the gasoline car without the GPF. The results showed that the utilization of exhaust particulate filter was important both in diesel and gasoline cars. The gasoline car without the GPF emitted relatively high concentrations of particles compared to the other technologies but the implementation of the GPF decreased particle emissions, and the potential to form secondary aerosols in atmospheric processes. The diesel car equipped with the DPF emitted low particle number concentrations except during the DPF regeneration events. Aromatic-free gasoline and diesel fuel efficiently reduced exhaust particles. Since the renewal of vehicle fleet is a relatively slow process, changing the fuel composition can be seen as a faster way to affect traffic emissions.

1. Introduction

Vehicular emissions are known to be a major source of air quality deterioration in urban areas (Fenger, 1999; Wang et al., 2009; Harrison et al., 2021b). Vehicle exhaust emissions contain gaseous compounds carbon monoxide (CO), nitrogen oxides (NO_x), ammonia (NH_3), nitrous oxide (N_2O) and volatile organic compounds (VOCs) as well as semi-volatile compounds, and particulate matter (PM). Primary exhaust PM is emitted directly from the vehicle and has not yet transformed in the atmosphere (Maricq, 2007). Fresh PM contains primary PM and species condensed on it in tailpipe when the raw exhaust cools down. Additionally, fresh PM consists of compounds that are in the gaseous phase under tailpipe conditions, like sulfuric acid, but condense or nucleate immediately when the exhaust is cooled and diluted with no significant chemical transformation (Rönkkö et al., 2013; Pirjola et al., 2015; Rönkkö et al., 2017). Secondary PM forms from exhaust gases via gas-to-particle conversion after they are released into the atmosphere as the oxidation processes typically decrease the volatility of the gaseous compounds (Gentner et al., 2017). Besides exhaust emissions, vehicle emissions contain pollution from non-exhaust sources such as tires, brakes and road abrasion. Their contribution cannot be overlooked, since it has been estimated that over 50% of $PM_{2.5}$ from road transport comes from the non-exhaust sources (Harrison et al., 2021a). In Euro 7, the new measures to limit particle emissions from tires and brakes will be implemented (https://www.europarl.europa.eu/news/en/press-room/20231207IPR15740/euro-7-deal-on-new-eu-rules-to-reduce-road-transport-emissions).

To diminish the harmful impacts of vehicular pollution, emission

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standards for vehicle PM emissions have been tightening globally. Currently, some exhaust pollutants are efficiently regulated in many regions, e.g. NO_x, PM, and number of non-volatile, solid particles with the size of >23 nm in European regulations (Commission Regulation (EU) 2017/1151). Instead, the emissions of small (<23 nm) and volatile particles, the formation potential of secondary PM, and the non-exhaust emissions are not directly under control, although it is not apparent why exhaust particles should be limited to this size and volatility fraction as the vehicle exhaust contains also volatile and semi-volatile particles (Biswas et al., 2007; Wang et al., 2017), as well as particles much smaller than 23 nm in size (Kittelson, 1998). In future, smaller particles will also be included in the regulations as in Euro 7 the size limit for solid particle number will be decreased to 10 nm. Furthermore, as previous studies have demonstrated (e.g. Chirico et al., 2010; Nordin et al., 2013; Platt et al., 2013; Timonen et al., 2017), secondary PM can be significantly greater than primary PM indicating that the emission limits may not regulate the total PM emissions measured in ambient air.

Exhaust emissions from combustion engines depend on several factors such as engine type, exhaust aftertreatment systems (ATS), fuel and lubricating oil properties as well as driving and environmental conditions (Kittelson et al., 2008; Lähde et al., 2009). To reduce air pollution caused by vehicles, these factors can be affected. For example, ATSs typically decrease solid particle number and mass concentrations (Tzamkiozis et al., 2010; Giechaskiel, 2018). A substantial decrease of soot has been obtained by using a diesel particulate filter (DPF;Robinson et al., 2015; Karjalainen et al., 2019; Rönkkö et al., 2023) drastically decreasing the soot emissions from heavy duty vehicles with the newest Euro standards (Dallmann and Jin, 2020). In gasoline engines particle number (PN) emissions can be reduced by equipping vehicle with a gasoline particle filter (GPF;Jang et al., 2018). At the same time ATSs can also change the chemical composition of emission particles. For instance, oxidative exhaust after-treatment like diesel oxidation catalyst (DOC) and DPF with catalytic coating may enhance particulate phase sulfur (Maricq et al., 2002; Arnold et al., 2012; Rönkkö et al., 2013). In addition to primary and fresh PM, the use of ATS can also affect the formation of secondary aerosol from the engine emissions. The presence of DOC and DPF has been shown to reduce the production of secondary organic aerosol in diesel vehicles (Chirico et al., 2010; Novakovic et al., 2023).

In terms of fuels, there are novel improved fuels on the market that produce less pollutants than conventional gasoline or diesel fuel. Aromatic compounds are known to be a significant source of exhaust PM (e.g. Peng et al., 2017; Chen et al., 2019), and therefore several aromatic-free fuels have been introduced. For example, alkylate gasoline is aromatic-free fuel that contains mostly isooctane. Zardini et al. (2014) have tested alkylate fuel for the 2-stroke and the 4-stroke scooters and noticed an emission reduction for several gaseous and particulate phase species. Similarly in small gasoline engines, typically used in gardening and forestry operation, alkylate fuel resulted in the reduction of aromatics and soot content of the emitted particles (Zardini et al., 2019). Regarding diesel fuel, Hydrotreated Vegetable Oil (HVO) fuel is an aromatic-free, paraffinic renewable diesel that has been noticed to decrease PM and NO_x emission whereas the effect on PN was not consistent (Bortel et al., 2019). In another study, HVO reduced PN significantly at the upstream of the ATS (DOC and DPF) but the PN emissions were not much different from standard diesel at the downstream of the ATS due to the low PN concentrations (Wu et al., 2017). Regarding PM emissions, in the study of Karavalakis et al. (2016) the PM mass emissions were larger for HVO than for ultra-low sulfur diesel fuel. In terms of secondary organic aerosol (SOA), HVO has been noticed to produce less SOA than petroleum diesel (Gren et al., 2021) or ultra-low sulfur diesel fuel (Ghadimi et al., 2022). However, it has been noticed that the use of biofuels can have some unwanted effects, for example on the engine lubricating oil performance (Stepien et al., 2014).

Compressed natural gas (CNG)-fueled cars have shown low particle emissions. Compared to the gasoline vehicles, the solid PN emissions of >23 nm particles were an order of magnitude lower for the CNG vehicle (Toumasatos et al., 2021). However, when the particle size limit was decreased to 10 nm or 2.5 nm, the emissions were similar for both fuels. Aakko-Saksa et al. (2020) have investigated the cars at Euro 2–6a level and noticed that the CNG cars emitted the smallest amounts of pollutants of all the cars with only the methane (CH_4) emissions elevated. In another study, higher NO_x emission compared to using gasoline has been measured for the CNG vehicles (Lv et al., 2023).

The aim of this study was to investigate gaseous and particulate exhaust emissions, and their potential to form secondary aerosols, of the cars meeting the most recent emission Euro 6d standards. Cars represented various fuels (gasoline, alkylate, diesel, HVO, CNG) and aftertreatment technologies (GPF, DOC, SCR, DPF, three-way catalyst (TWC)). Additionally, the cars were tested in cold and warm ambient temperatures in laboratory conditions. Measurement species included both regulated and unregulated pollutants (nitrogen dioxide (NO_2), N_2O, NH_3, CH_4, formaldehyde (HCHO), ethanol, particle number size distribution, PM_1 chemical composition). In terms of particles, measurements covered non-volatile primary, fresh and aged particle emissions. This study provides novel results on gaseous and particle emissions, and their potential to form secondary aerosols, from state-of-the art cars as well as illustrates the impact of ATS, aromatic content of fuel and ambient temperature on emissions. This study also discusses the importance of measuring specific unregulated pollutants (PN < 23 nm, chemical composition, secondary aerosol formation potential) in terms of their effect on air quality. The information obtained in this study is highly required by the authorities when the passenger car emission regulations are revised in near future. The results of this study can help the decision makers to determine what are the pollutants that need to be regulated in terms of urban air quality and what are the required limit values.

2. Materials and methods

2.1. Cars and test procedures

All the studied cars were relatively new, the model year of the oldest car was 2019 (Table 1), and they all met the latest Euro 6 limits according to their type approval. The cars were selected to represent different engine technologies. The cars included a compression-ignition (CI) diesel car with a diesel particulate filter (named Diesel DPF), two spark-ignition (SI) gasoline cars, one with multipoint fuel injection (MPFI) and a three-way catalyst (named Gasoline), and one with turbocharged stratified injection (TSI), a TWC and a gasoline particulate filter (named Gasoline GPF), and a natural gas vehicle (NGV) fueled with CNG (named CNG). This car was equipped with a TWC and the injection

Table 1
Studied Euro 6d cars and fuels.

	Gasoline	Gasoline GPF	Diesel DPF	CNG[a]
Model year	2021	2021	2019	2020
Odometer (km)	2500	9400	38600	9800
Aftertreatment	TWC	TWC, GPF	DOC, DPF, SCR	TWC
Displacement (cm^3)	1199	999	1598	1498
Power (kW)	55	81	85	96
Torque (Nm)	110	200	250	200
Mass (kg)	1055	1298	1385	1389
Ignition system	SI	SI	CI	SI
Injection system	MPFI	TSI	TDI	TSI
Emission standard	Euro 6d	Euro 6d	Euro 6d-TEMP	Euro 6d
Fuels	EN228 (ArH) Alkylate (Ar0)	EN228 (ArH)	EN590 (Ar20) HVO (Ar0)	CNG (EN228[a])

[a] Equipped also with gasoline tank to back-up CNG in certain conditions.

system was TSI.

The gasoline car was tested with normal EN228 gasoline fuel containing 34% of aromatic compounds (ArH) and with alkylate gasoline that was free of aromatics (Ar0) (Table S1). The gasoline car with the GPF was tested only with EN228 fuel. The diesel car was examined with normal EN590 diesel fuel containing 19% of aromatics (Ar20) and with HVO type fuel free of aromatics (Ar0). The CNG car was driven with CNG from the Finnish natural gas pipeline but it also consumed small amounts of EN228 gasoline for instance during start-ups. Lubricant oil properties were not analyzed in this study.

Measurements were conducted in two different temperatures: warm (between +22 and + 24 °C) and cold (between −8 and +2 °C) to represent summer and winter temperatures in the northern countries, respectively. Cars were preconditioned in the temperature-controlled dynamometer room overnight and kept in the room before they were tested to maintain warm/cold conditions. Before the measurement run, cars were moved on the light-duty chassis dynamometer (Froude Consine) and fastened without starting the engine. Thus, the engine experienced a cold start in all the tests. The driving cycle was a modification of the Worldwide Harmonized Light Vehicles Test Cycle (WLTC). Two consecutive WLTC cycles were combined resulting in the test point with 1 h duration.

The regeneration of the DPF occurred twice during the measurements. Both events took place when the diesel car was driven at cold temperature and with HVO fuel. The first DPF regeneration occurred at the end of the WLTC cycle (~3400 s after the start of the cycle) and it lasted until the end of the cycle. The second DPF regeneration took place 600–800 s after the start of the WLTC cycle and was likely to be the continuation of the first regeneration event.

2.2. Measurement set-up

The measurement setup is presented in Supporting information in Fig. S1. Measurements were conducted from two different sampling points in the system. First sampling point was from the heated transfer line connected to the tailpipe and the second sampling point was after the exhaust dilution in a constant volume sampler (CVS) system.

Gaseous components were measured from the first sampling point with a Fourier transformation infrared (FTIR) spectrometer (Mega-1, Rowaco AB, Sweden). A large set of aerosol instruments measured also from the first sampling point through a sampling system where the primary dilution was performed with a Porous Tube Diluter (PTD) with a dilution ratio (DR) of 12, followed by residence time tube and secondary dilution with an ejector diluter (DR = 11). With primary DR 12, the system has been reported to reproduce the semivolatile particle formation processes of fresh exhaust and enable the comparison of trends in fresh exhaust aerosols (see e.g. Keskinen and Rönkkö, 2010). Particles measured from this sampling point are considered as fresh particles as the aerosol measured after the dilution system contains both primary particles and species that would condense on particles after they exit the tailpipe. Particle number concentrations and size distributions were measured by a Condensation Particle Counter (CPC) battery consisting of CPCs at the cut points of 1.2 nm (A10, Airmodus Oy, Finland), 10 nm (A20, Airmodus Oy, Finland), and 23 nm (A20, Airmodus, Finland) as well as with an Electrical Low-Pressure Impactor (ELPI+, Dekati Ltd, Finland). Chemical composition of the submicron particles (<1 μm) was determined with a Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne Research Inc., US). Black carbon (BC) was determined with the aethalometer (AE33, Magee scientific, Berkeley, US).

Aging of particles was mimicked by using an oxidation flow reactor (OFR; Potential Aerosol Mass chamber, Aerodyne Research Inc, US). The photochemical age in the ORF was calculated by using two methods. In the first method, the photochemical age was obtained from the decay of CO. CO was continuously injected into the sample line before the OFR and measured by the CO analyzer (Environnement S.A. CO12) after setting the OFR UV lights on/off, similarly to Karjalainen et al. (2022) and Lambe et al. (2011). In the second method, the difference in the CO concentration between the fresh and aged measurements was utilized to obtain the photochemical age. Based on these calculations, the photochemical age varied between 6.1 and 10.4 days depending on the vehicle and calculation method (Table S2). It should be noted here that the calculation of the photochemical age with these methods has a lot of uncertainties, e.g. due to the different dilution ratios. Also, definition and features of SOA from exhaust is ambiguous as there are a several different OFRs used in the emission measurements (e.g. Lambe et al., 2011; Simonen et al., 2017; Ihalainen et al., 2019; Wu et al., 2023) that often operate with different parameters and therefore produce SOA with different oxidation states. Furthermore, the aging process in the OFRs may differ from that in the atmosphere even though the atmospheric oxidant ratios are simulated in the OFRs as closely as possible. The measured SOA yield can be compared with the predicted SOA yield calculated based on the measured SOA precursors (Odum et al., 1996; Timonen et al., 2017) but in this study predicted SOA could not be calculated as the individual precursors were not measured.

Standard emission measurements with some additional instruments were conducted after the CVS-dilution (i60, AVL List GmbH, Austria). For the non-volatile primary particle measurement, the CVS-diluted exhaust gas was additionally diluted with a PMP-type dilution system (DEED-100, Dekati Ltd, Finland), which removes volatile compounds from the particles in a high-temperature dilution process. The DEED dilutes exhaust at 350 °C in the first stage, evaporates volatile compounds at 450 °C and dilutes with room temperature air in the second stage. The dilution ratio (particle reduction factor) of the DEED was 89. The number of non-volatile particles was measured with a CPC (A23, Airmodus Oy, Finland) with a 23 nm cut off and the size distribution with an ELPI (Dekati Ltd., Finland). Also the gravimetric PM sampling was conducted from the CVS-diluted sample. The PM samples were collected on polytetrafluoroethylene membrane filters (Fluoropore, Merck KgaA).

Results for gases, PM and non-volatile PN for >23 nm particles (nvPN23) are presented as emissions (g/km or #/km) while the number concentrations for fresh particles (1.2–10 nm, 10–23 nm and >23 nm) and the number size distributions are presented as concentrations (#/cm^3). Also the chemical species and mass for fresh and aged PM_1 are presented as concentrations (mg/m^3).

3. Results

3.1. Gaseous emissions, PM and non-volatile primary PN > 23 nm

NO_x emissions were low, below 10 mg/km, for all the cars except for the diesel car with the DPF at cold ambient temperature that had the NO_x emissions above 30 mg/km (Fig. 1a). The diesel car with the DPF had also the largest N_2O emissions in the range of 7–10 mg/km (Fig. S2a). In contrast, the CNG car produced the largest NH_3 emissions of 9–11 mg/km (Fig. S2a). Ammonia and nitrous oxide are known to be induced by some exhaust aftertreatment technologies in certain conditions (Heeb et al., 2006; Jung et al., 2022). In this study, ammonia was observed especially from the TWC in the CNG-fueled car while nitrous oxide was detected from the DPF in the diesel car.

Total hydrocarbon (THC) emissions were below 5 mg/km for all the other cars except for the CNG car (Fig. S2b). The THC emission measured for the CNG car at cold ambient temperature (~60 mg/km) can be regarded as a relatively high emission. Hydrocarbon emissions originate typically from unburned or partly combusted fuel and lubricating oil. That was also the case here as the CNG car emitted unburned methane, which represented a major part of the THC emissions of this car (Table S3). Formaldehyde emissions were extremely low for all the cars being below 2 mg/km (Fig. S2b), and in most cases below the detection limit of the instrument. Formaldehyde emissions indicate also partly combusted fuel and lubricating oil.

In terms of particle emissions, PM and nvPN23 emissions were

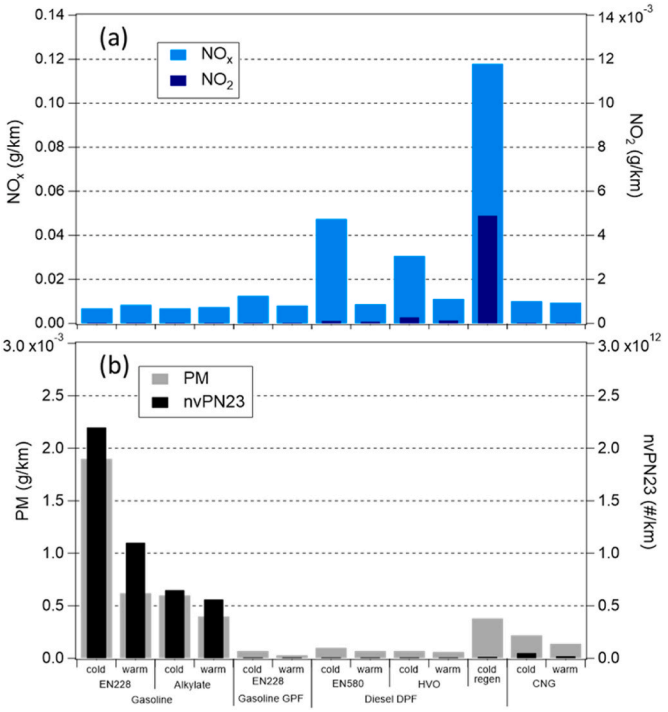

Fig. 1. NO_x and NO_2 (a), and PM and nvPN23 emissions from the tested cars over two consecutive WLTC cycles. The DPF regeneration events occurred during the measurements with the Diesel DPF, HVO fuel and cold ambient temperature (shown separately).

clearly larger for the gasoline car without the GPF than for the other tested cars (Fig. 1b). When using EN228 fuel for the gasoline car, the nvPN23 emission was larger than the Euro 6d limit at both test temperatures (Table S3), however, with aromatic-free alkylate fuel the nvPN23 emission decreased below the Euro 6d limit value. PM emissions were in all cases below 4.5 mg/km (Euro 6d limit value) even for the gasoline car without the GPF. With some exceptions (see Fig. 1, Table S3), gaseous, PM and nvPN23 emissions were generally larger at cold than at warm ambient temperature. That is in line with the study of Liu et al. (2023) that found a strong and positive impact of low ambient temperatures on vehicular emissions. Also Wang et al. (2022) have noticed six times larger PN emission factor at −10 °C compared to that at 23 °C for the gasoline vehicles.

The regeneration of the DPF increased the nvPN23 emissions for the diesel car for a short period of time (at ~600–800 s; Fig. S3b), but the increase was not substantial when the results were calculated over the whole test point. However, the DPF regeneration was divided into two tests, each consisting of two WLTC cycles. Hence, the actual effect of the regeneration on the PM and nvPN23 emissions would be higher than that shown in Fig. 1b and Table S3. The DPF regeneration was most clearly seen in NO_x and especially NO_2 emissions.

3.2. Size distributions and size-segregated number concentrations

The gasoline car without the GPF emitted non-volatile primary particles in a wide size range (Fig. S4a). Peak of the number size distribution was at 20–30 nm but significant particle concentrations existed also in the larger sizes up to 300 nm, however, the concentration of larger particles was smaller in warm ambient temperature than in cold temperature for EN228. With aromatic-free alkylate fuel, significantly smaller non-volatile particle concentrations were measured over the whole size range. For alkylate fuel, the non-volatile particle emissions were higher in cold conditions, but the difference was not as obvious as with aromatic-containing fuel. Like for gasoline, aromatic-containing diesel fuel (EN590) generated more particles than aromatic-free (HVO) fuel, however, the non-volatile particle concentrations from the diesel car were small and close to the detection limit of the measurement system. Particle concentrations from the CNG car and the gasoline car with the GPF were small being comparable with the emissions from the diesel car. The effect of temperature on the size distributions was not clearly seen for these cars.

In terms of fresh particles, the gasoline car without the GPF emitted also relatively high concentrations of fresh particles (Fig. S4b). For aromatic-containing fuel, cold conditions increased particle concentrations compared to warm temperature, but in case of alkylate fuel, temperature did not affect the size distribution of fresh particles. The diesel car emitted small concentrations of fresh particles with the particle number being too small for the accurate number size distribution measurements. However, during the DPF regeneration events with HVO fuel, high particle concentrations were produced, both non-volatile and fresh particles. Result indicates that regeneration events are important even with aromatic-free hydrocarbon fuels, e.g. due to the storage and release phenomena of semivolatile compounds (Karjalainen et al., 2014).

The size of exhaust particles was also investigated by measuring three different PN fractions, 1.2–10 nm, 10–23 nm and >23 nm, for fresh particles. The largest contribution (~53–63%) of 1.2–10 nm particles was measured for the CNG car with the fraction being larger in warm than in cold ambient temperature (Fig. 2). That is in line with the study of Lähde and Giechaskiel (2021) that noticed a larger fraction of solid 4–10 nm particles at warm temperature than at subzero ambient temperature for the CNG vehicle. Also, the gasoline car without the GPF with aromatic-containing fuel had relatively large fraction of 1.2–10 nm particles (~38–46%) at both temperatures with higher fraction in cold ambient temperature whereas the same vehicle with the alkylate fuel had significantly lower fraction of particles between 1.2 and 10 nm (<20%) with both ambient temperatures. The gasoline vehicle with the GPF had ~30% of particles in that size range. The diesel car had also rather small fraction of 1.2–10 nm particles (<30%) except during the DPF regeneration event when the fraction increased to > 80% (Fig. 3 and S5). Regarding the fraction of <23 nm particles, that fraction was largest for the CNG car in both temperatures (~80%), gasoline car with the GPF in warm ambient temperature (~60%) and the diesel car with the DPF with HVO (~60%). Notably, with the diesel vehicle both <23 nm fractions increased with the HVO fuel (DPF regeneration not included) compared to the EN590 fuel. With the gasoline vehicle, alkylate fuel had smaller fraction of <23 nm particles compared to the EN228 fuel. It has been shown in earlier studies that the use of aromatic-free fuel may not decrease particle number emission consistently (Bortel et al., 2019) that may be related to the measured particle size and volatility-fraction.

3.3. Mass and chemical composition of fresh PM_1

The mass and chemical composition of PM_1 were measured for the fresh particles by the SP-AMS and aethalometer. The largest fresh PM_1 concentrations were measured for the gasoline car without the GPF, the concentrations being larger in cold ambient temperature and with aromatic-containing (EN228) fuel (Fig. 4). The same trend was noticed for the diesel car, the PM_1 emissions being larger with aromatic-containing (EN590) fuel than with aromatic-free (HVO) fuel. PM_1 particles consisted mostly of organic matter or BC (Fig. 4a and S6a). PM_1 from the CNG car had the largest contribution of organic matter (72–77% of PM_1) whereas the gasoline car without the GPF emitted mostly BC (83–87% of PM_1) with both fuels and temperatures. For the other cars, the mass contribution of BC was much smaller (17–36% of PM_1). A large fraction of BC has been measured earlier in Peng et al. (2022) who measured the chemical composition of PM for the gasoline car equipped with a TWC (no GPF).

The composition of organic matter differed between the cars. Organic matter comprised mostly of hydrocarbons for the gasoline car

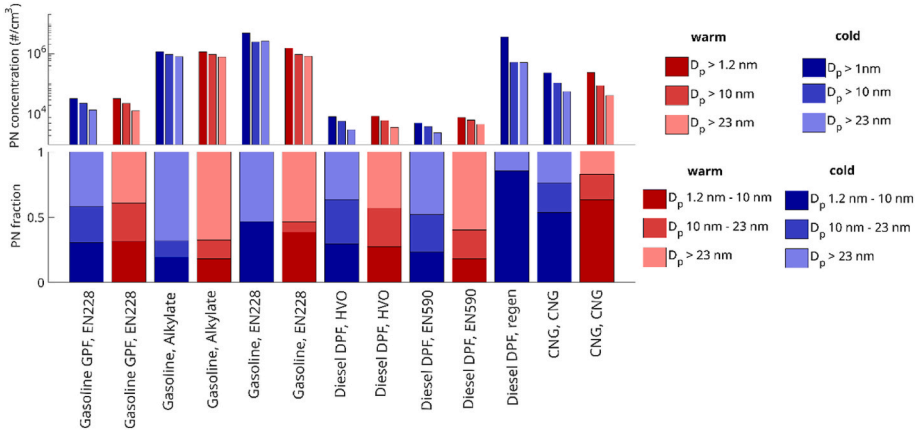

Fig. 2. Fresh particle number concentrations and size fractions for the tested cars over two consecutive WLTC cycles measured with the CPCs with the cut-offs of 23 nm and 10 nm and PSM with the cut-off of 1.2 nm. The DPF regeneration events occurred during the measurements with the Diesel DPF, HVO fuel and cold ambient temperature (shown separately).

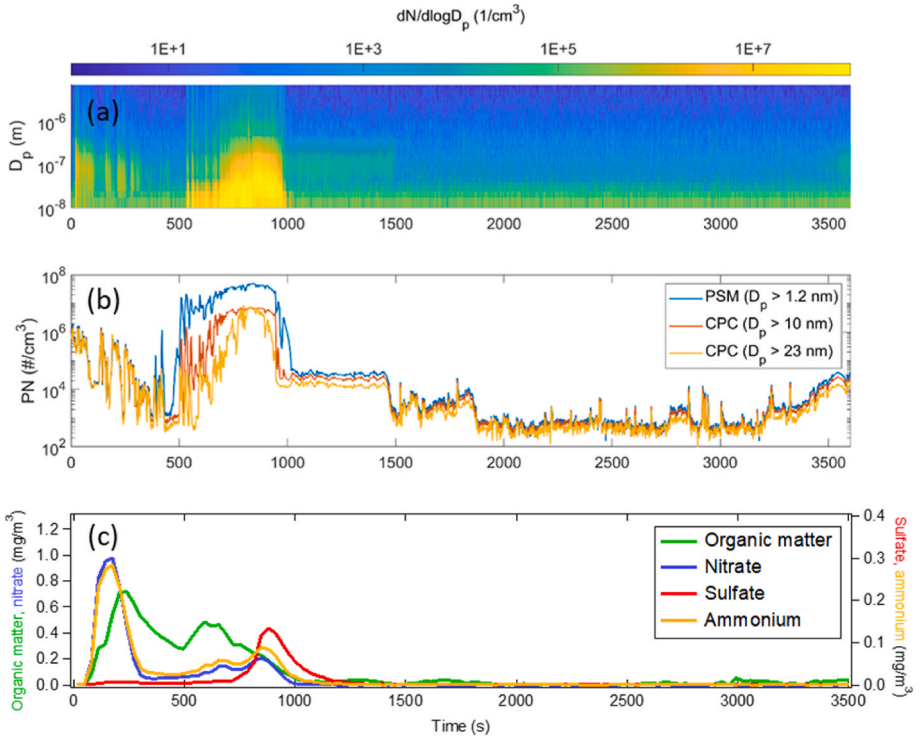

Fig. 3. DPF regeneration event from WLTC-cycle for Diesel DPF, HVO and cold ambient temperature. Number size distribution of fresh particles (a), size fraction of fresh particles measured with the CPCs with the cut-offs of 23 nm and 10 nm and PSM with the cut-off of 1.2 nm (b) and the concentrations of organic matter, sulfate, nitrate and ammonium in aged PM_1 (c).

without the GPF (61–65%) and the CNG car (39–48%) whereas for the gasoline car with the GPF and diesel car with the DPF it consisted mainly of oxygenated organic matter (70–88%; Fig. S7a). PM_1 included also inorganic species, sulfate and nitrate, whereas ammonium and chloride were below the detection limits of the SP-AMS. Nitrate and sulfate concentrations were largest for the gasoline car without the GPF, especially at cold ambient temperature, but in terms of mass fractions they were highest for the gasoline car with the GPF and the diesel car with the DPF. These inorganic species were likely to be related to lubricant oil or they were formed in the ATS and had a detectable contribution since the exhaust emissions from fuel were small. DPF regeneration events did not occur during the fresh PM_1 measurements with the SP-AMS.

3.4. Mass and chemical composition of aged PM_1

Aged PM_1 was measured after the OFR including both fresh particles and secondary material. The largest concentrations of aged PM_1 were detected for the gasoline car without the GPF its concentration being clearly largest for EN228 fuel at cold ambient temperature, even close to 100 mg/m^3 (Fig. 4). The aromatic-free alkylate fuel substantially reduced aged PM_1 to below 4 mg/m^3. For the gasoline car equipped with the GPF, aged PM_1 was higher at cold ambient temperature, above 10 mg/m^3, than at warm temperature. For the diesel car equipped with the DPF, aged PM_1 varied from 0.18 to 2.9 mg/m^3 without a clear effect of the fuel aromatic content, however, the regeneration events with aromatic-free fuel may bias this conclusion. For the CNG car, aged PM_1 was 2.9 mg/m^3 at warm ambient temperature, while it was below 1 mg/

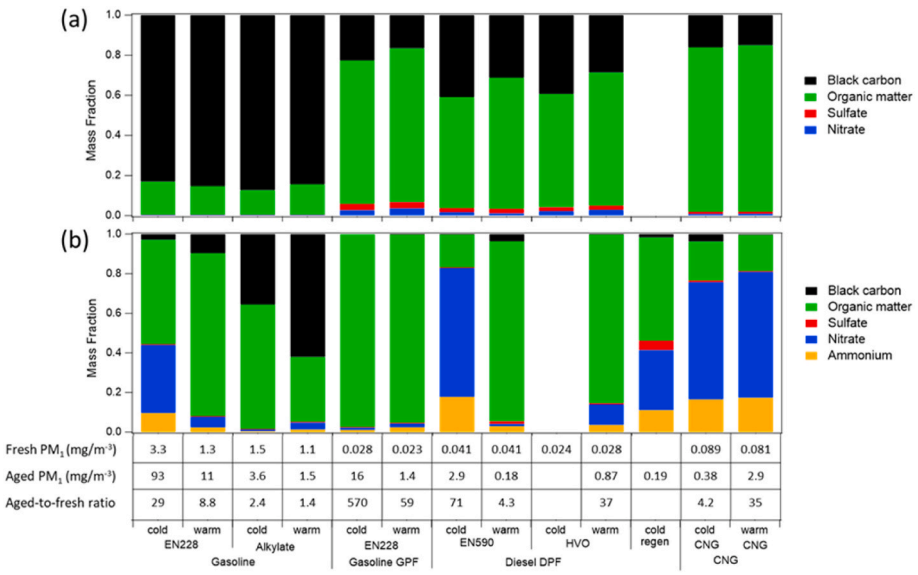

Fig. 4. Chemical composition of fresh (a) and aged (b) PM_1, and the mass concentrations of fresh and aged PM_1. The DPF regeneration events occurred during the aged particle measurement of Diesel DPF, HVO fuel and cold ambient temperature (shown separately).

m^3 at cold ambient temperature. The formation of aged PM_1 with the CNG-fueled car can be explained by the precursors for the secondary aerosol formation, especially ammonia (15–16 mg/m^3) and NO_X (14–15 mg/m^3) were observed at both test temperatures. Additionally, aromatics may be present in the exhaust from the CNG car due to gasoline use at cold ambient temperature. Furthermore, lubricating oil may affect the formation of aged PM_1.

Aged PM_1 consisted mostly of organic matter, BC, nitrate and ammonium (Fig. 4b and S6b). Organic matter dominated the composition of all cars with some exceptions, for example for the CNG car aged PM_1 comprised mostly of nitrate and ammonium. A large share of ammonium nitrate in aged PM_1 for the CNG engine has been presented earlier e.g. in Alanen et al. (2017) and Ghadimi et al. (2023). Notably, when switching from aromatic-containing gasoline to aromatic-free alkylate fuel, especially at warm conditions, the concentration of organic matter decreased, and also its relative share in aged PM_1. Additionally, nitrate and ammonium reduced in aged PM_1 when moving from aromatic-containing gasoline to aromatic-free alkylate. In the case of alkylate, aged PM_1 was dominated by BC because aged PM_1 consisted mostly of fresh particles. Regarding the diesel car with the DPF, organic matter governed aged PM_1 in other conditions than at cold ambient temperature with aromatic EN590 fuel, for which aged PM_1 was made largely of nitrate and also ammonium was present. For the CNG car, the contribution of ammonia in the exhaust induced ammonium in aged PM_1, and nitrate was also present. The contribution of exhaust NO_x in aged PM_1 was not straightforward. For all cars and fuels studied, NO_x in the exhaust was sufficient to form nitrate in aged PM_1, however, nitrate was present in combination with ammonium, potentially in the form of ammonium nitrate. The formation of ammonium nitrate depends on the driving conditions; it has been shown that the formation of ammonium nitrate in secondary aerosol increases in the hot-running phases of the cycles for the gasoline cars with a TWC (Kuittinen et al., 2021). As expected, organic matter in aged particles was highly oxygenated and comprised mostly of oxygenated ions with only 11–38% fraction of hydrocarbon ions (Fig. S7b).

The ratio of aged-to-fresh PM_1 was largest for the gasoline car with the GPF in cold conditions being approximately 600, whereas the corresponding ratio was 29 for the gasoline car without the GPF at same temperature and same fuel (Fig. 4). The smallest ratios (1.4–2.4) were measured for the gasoline car without the GPF with alkylate fuel indicating a small potential for the formation of secondary material in PM_1. In general, the difference between cold and warm ambient temperature was larger for aged PM_1 than for fresh PM_1. Also the reduction in PM_1 due to aromatic-free fuel was more noticeable for aged PM_1 than for fresh PM_1 for the gasoline car without the GPF. For the diesel car with the DPF, the impact of fuel aromatic content could not be investigated due to the DPF regeneration events during the aged PM_1 measurements of HVO fuel.

The DPF regenerations were clearly seen in the emissions of aged PM_1 sulfate and also at some extent in the concentration of organic matter, nitrate and ammonium (Fig. 3and S5b). However, the increase of sulfate was not that substantial when the results were calculated over the whole test point as the concentration of sulfate was only double of that at warm conditions of the same car and fuel. Larger sulfate concentration was likely to be related to the storage of sulfur in the DPF or catalyst, sulfur being released from the DPF during the regeneration (Hirano et al., 2015). The regeneration was also noticed in the nitrate and ammonium concentrations, in the first regeneration event more clearly than in the second one.

4. Discussion

This study shows that gas and particle emissions varied considerably between the investigated vehicles although they all met the most recent Euro 6d emission standards. The environmental conditions where the cars are used can significantly affect both the exhaust particles and the potential of exhaust to form secondary aerosol when it is aged in the atmosphere. In this study, the decrease of ambient temperature generally increased the car emissions. The increase was seen in the particle number concentration as well as in the secondary aerosol formation potential of exhaust. However, it was observed that the effect of ambient temperature was not equal with all the cars; the decrease of ambient temperature was observed to increase the fresh particle number emission of the gasoline car without the GPF and fueled with EN228, but e.g. for the diesel car with the DPF the effect was not equally significant. The study highlights the role of atmospheric conditions and processes also regarding the total air quality impacts of emissions, seen e.g. in relatively high potential of exhaust from certain cars to form secondary aerosol in the atmosphere and especially in high variation in the ratios of aged and fresh exhaust particulate mass, seen in Fig. 4.

Utilization of exhaust particle filtration techniques was found to be important both in diesel and gasoline cars. The effect of filtration was

seen here related to the gasoline car in terms of BC particles, particle number of fresh exhaust and also in the potential of exhaust to generate secondary aerosol. For fresh exhaust particle number, the decrease was even two orders of magnitude whereas the PM_1 mass reduction was mostly related to the decrease of BC both in warm (88% reduction) and cold (82% reduction) ambient temperatures. It can be seen that the emission regulations aiming at reduced impacts of emissions on human health by the exhaust cleaning systems are efficient, e.g. in reduction of BC particles. However, it should be noted that the filtration does not fully remove organic particulate matter emissions, and it seems to increase the relative importance of secondary aerosol formation potential of exhaust. Also the regeneration of particle filters should be taken into account. In this study it was observed that the DPF regeneration increased the concentration of small particles, especially 1.2–10 nm particles of fresh exhaust. For the non-volatile >23 nm particles, the impact was smaller. Furthermore, after the regeneration, the particle number concentration remained at slightly elevated level for some time. The DPF regeneration increased the concentration of secondary sulfate, nitrate and ammonium. Increased sulfate concentration indicated that sulfur was released from the DPF or catalyst (Karjalainen et al., 2014). However, further development of the DPF and GPF technologies are expected to reduce the regeneration emissions (Boger et al., 2021; Rose et al., 2021).

The most traditional and widely used exhaust aftertreatment technology for spark-ignition cars is the TWC, however, addressed with ammonia or nitrous oxide formation in certain conditions (Aakko-Saksa et al., 2020). These emission species are known to be induced by also other exhaust aftertreatment technologies (Dimaratos et al., 2022), which emphasizes the importance of careful consideration of all exhaust species. We observed significant ammonia emissions from several cars, especially from the CNG-fueled car, with elevated potential to form secondary emissions, for example ammonium nitrates.

This study indicates relatively strongly that the emissions can be significantly diminished by affecting the composition of fuel. Especially the reduction of aromatic compounds of gasoline and diesel fuels is efficient, both for fresh and aged exhaust PM, particularly at low ambient temperature. That is line with the earlier results (Zardini et al., 2014) showing a reduction in PM, aromatic compounds and SOA with alkylate fuel. Also a clear decrease in the secondary aerosol formation from the exhaust was observed when the ethanol content of fuel was increased (Timonen et al., 2017) explained by smaller emission of potential aerosol precursors as ethanol fuel contains less aromatic compounds.

Current emission standards regulate the particle number emissions of non-volatile particles larger than 23 nm, but the European Council has decided to decrease the particle size to 10 nm in Euro 7. However, the real fresh exhaust is significantly contributed by 1.2–10 nm particles, as seen with all the cars used here and especially with the CNG-fueled car emitting the largest fraction of 1.2–10 nm particles. Thus, in the development of future emission standards also the sub-10 nm particle size range should be kept in mind, especially if future investigations confirm that those tiny particles have negative impacts on human population health. For example, Maher et al. (2016) has proposed that inhaled nanoparticles originated from combustion processes can penetrate to the human brain through the olfactory bulb and cause Alzheimer's disease and other brain diseases.

5. Conclusions

This study shows that the understanding of all effects of vehicle technologies on particle emissions requires experiments that reveal extensively fresh and aged exhaust emissions in addition to regulated non-volatile particle and gas emissions of cars. Additionally, the whole size range of the exhaust particles needs to be measured as well their chemical composition. In this study the benefits of using aromatic-free gasoline and diesel fuels were perceived, and also the potential secondary aerosol formation from ammonia induced by ATS.

Application of exhaust particle filtration techniques was shown to be efficient both in diesel and gasoline cars. However, the impact of filtration was not similar for all the particle components as the filtration did not fully remove organic matter emissions, and it seemed to increase the relative importance of secondary aerosol formation potential of the exhaust. This should be considered when evaluating the air quality and health related impacts of the newest vehicles, and in future emission regulations. Also the regeneration of the particle filters should not be overlooked as the DPF regeneration increased the concentration of the smallest measured particle fraction (1.2–10 nm particles) of fresh exhaust.

While the renewal of vehicle fleet and thus the reduction of vehicle fleet emissions are relatively slow, fuel changes can be seen as an alternative way to affect faster the traffic emissions. This study demonstrated the emissions can be significantly reduced by affecting the composition of fuel. Notably, aromatic-free alkylate-type gasoline efficiently reduced the aged PM_1 emissions, hence providing a solution to reduce particle emissions from the gasoline cars without the GPF. Aromatic-free fuel can also alleviate the potential to form secondary emissions from dual-fuel cars, which are using gasoline or diesel as a back-up. This opportunity is not fully used in current political actions regarding air quality related policies and actions. Furthermore, fuel-based emission reduction actions can largely be made at the levels of societies and nations, while the vehicular level decisions are more typically made even at individuals' level, which can make fuel changes more feasible way to rapidly reduce atmospheric emissions.

CRediT authorship contribution statement

Sanna Saarikoski: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Funding acquisition, Formal analysis, Conceptualization. **Anssi Järvinen:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Lassi Markkula:** Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation, Formal analysis, Data curation. **Minna Aurela:** Writing – review & editing, Investigation, Data curation. **Niina Kuittinen:** Writing – review & editing, Project administration, Methodology, Investigation. **Jussi Hoivala:** Writing – review & editing, Investigation. **Luis M.F. Barreira:** Writing – review & editing, Investigation, Data curation. **Päivi Aakko-Saksa:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Teemu Lepistö:** Writing – review & editing, Investigation. **Petteri Marjanen:** Writing – review & editing, Investigation. **Hilkka Timonen:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Henri Hakkarainen:** Writing – review & editing, Investigation. **Pasi Jalava:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization. **Topi Rönkkö:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

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

Data will be made available on request.

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Appendix A. Supplementary data

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