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## Strategies To Diminish the Emissions of Particles and Secondary Aerosol Formation from Diesel Engines

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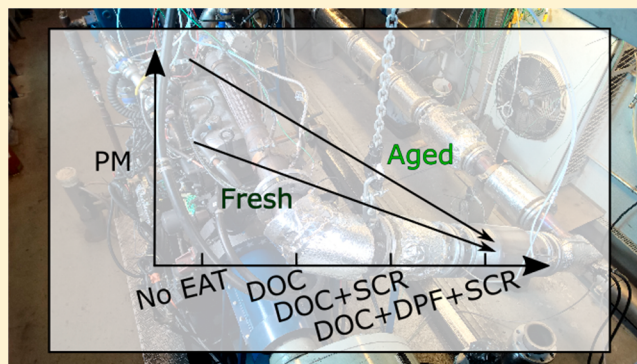
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### S Supporting Information

**ABSTRACT:** Particle emissions and secondary aerosol formation from internal combustion engines deteriorate air quality and significantly affect human wellbeing and health. Both the direct particle emissions and the emissions of compounds contributing to secondary aerosol formation depend on choices made in selecting fuels, engine technologies, and exhaust aftertreatment (EAT). Here we study how catalytic EATs, particle filtration, and fuel choices affect these emissions concerning heavy-duty diesel engine. We observed that the most advanced EAT decreased the emissions of fresh exhaust particle mass as much as 98% (from 44.7 to 0.73 mg/kWh) and the formation of aged exhaust particle mass ~100% (from 106.2 to ~0 mg/kWh). The composition of emitted particles depended significantly on the EAT and oxidative aging. While black carbon typically dominated the composition of fresh exhaust particles, aged particles contained more sulfates and organics. The fuel choices had minor effects on the secondary aerosol formation, implicating that, in diesel engines, either the lubricant is a significant source of secondary aerosol precursors or the precursors are formed in the combustion process. Results indicate that the utilization of EAT in diesel engines would produce benefits with respect to exhaust burden on air quality, and thus their utilization should be promoted especially in geographical areas suffering from poor air quality.



## INTRODUCTION

Health studies have revealed the association of untreated diesel exhaust with short-term and long-term adverse health effects in humans.<sup>1</sup> Due to that, the collective efforts of scientists together with the solutions achieved in technology development and policies to diminish the emissions have brought significant improvements to exhaust emission control. The breakthrough in the control of diesel exhaust particle emissions has been achieved with the enforcement of the usage of diesel particle filters (DPFs) in many diesel applications starting from diesel passenger cars and later extending to trucks and buses. The DPF effectively filters the exhaust gas before it is emitted to the atmosphere and have been shown to lead to very efficient real-world reductions in the emissions of PM mass and in total particle number (PN),<sup>2,3</sup> compared to typical pre-DPF levels. Heavy-duty engines equipped with DPFs and complying

with the US2010 standards were reported to emit 2 orders of magnitude less mass and number of total particles compared to the pre-DPF ones in a comprehensive laboratory study.<sup>4</sup> DPFs have also been shown to result very low nonvolatile particle (mostly soot and metallic ash) emissions.

Vehicles contribute to atmospheric PM concentrations not just through their direct (primary) PM emissions, measured at the tailpipe but, even more significantly, through photo-oxidation and gas-to-particle processes of initially gaseous exhaust components (secondary PM).<sup>5</sup> Primary particulate matter refers to particles directly emitted, e.g., from engine, fuel combustion process or brakes, and not yet experienced any

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significant chemical transformation in the atmosphere. Depending on engine and fuel type, primary exhaust PM emissions from vehicles consist mainly of soot and different fuel and lubricating oil components.<sup>6–8</sup> In addition to primary PM, the combustion process in the engine cylinder produces so-called delayed primary aerosol species (see Rönkkö et al.<sup>9</sup>) like sulfuric acid which are in gaseous phase under tailpipe conditions but will condense or nucleate immediately when the exhaust is cooled and diluted, without any significant chemical transformation in the atmosphere.<sup>10–12</sup> In particle number size distribution, the exhaust PM formed by different processes are frequently seen as separate modes with different concentrations and particle size ranges.<sup>11,13</sup> In addition to the primary and delayed primary PM, large amounts of secondary particulate matter forms after the exhaust gases are released into the atmosphere.<sup>14–17</sup>

Both batch chambers (such as smog chambers) and flow through chambers have been applied to study vehicular secondary aerosol emissions under both laboratory and ambient conditions. In general, a batch chamber is good for detailed oxidation process studies<sup>14,17</sup> but cannot be used, e.g., to differentiate the influence of rapidly changing driving conditions to emissions during a test cycle. To overcome this challenge, oxidation flow reactors (OFRs) are designed to simulate secondary aerosol mass formation potential on a close to real-time basis.<sup>18–20</sup> Recent chamber studies have shown that secondary particulate matter from combustion engines consists mainly of organic compounds and ammonium nitrate<sup>14–16</sup> and that the secondary PM formation can be significantly larger than primary PM emissions.<sup>16,17</sup>

The emissions of secondary PM precursors from internal combustion engines have been observed to depend on fuel properties; e.g. Timonen et al.<sup>21</sup> reported significant decrease of these emissions when the fuel of a gasoline direct injection (GDI) passenger car was changed from the mixture of gasoline (90%) and ethanol (10%) to 100% ethanol. However, also the advantages of the oxidative exhaust aftertreatment (EAT) and especially the use DPF<sup>22,23</sup> in terms of secondary aerosol formation from diesel exhaust have been reported. A common way of making the conclusions is based on carrying out smog chamber or OFR experiments for different individual vehicles equipped with their specific EAT systems. It should be noted that in these cases, also the engine-out emission of SOA precursors may differ from an engine to another and thus the conclusions regarding the effects of EAT on secondary PM precursors and also on PM in fresh exhaust are not straightforward.

In this study, we used a comprehensive set of controlled measurements to characterize both primary, delayed primary, and secondary particulate emissions of a modern heavy-duty diesel engine. The measurements were conducted under laboratory conditions with a nonroad diesel engine by varying the EAT and fuel; the effects of a diesel oxidation catalyst (DOC), a selective catalytic reduction (SCR) system, and a diesel particulate filter (DPF) and the fuel change from a conventional fossil diesel fuel to a renewable paraffinic fuel were studied. All measurements were performed with the same engine in order to consistently evaluate the effects of each EAT-fuel combination. This is in contrast with the study by Gordon et al.<sup>23</sup> where the effect of EAT was studied by measuring vehicles with different EATs. In addition, we cover all relevant exhaust catalyst types in a stepwise manner, as opposed to the study by Jathar et al.<sup>22</sup> where one EAT

combination was tested. According to our knowledge, this is the first time when secondary aerosol formation from paraffinic diesel fuel is compared to a common fossil fuel.

The engine and the EAT systems used in the study represent modern technologies; hence the results of the study can be seen to describe the diesel engine applications in general. Current emission limits in several regions still do not require the use of DPFs in diesel engine applications, and even in countries with the most advanced emission standards, DPF equipped vehicles only correspond to a fraction of the fleet. However, the engine and EAT systems used in this study also describe the emission situation of nonroad mobile machinery and forthcoming changes in their particle emissions. The new Stage V for nonroad mobile machinery (NRMM) was introduced with Regulation (EU) 2016/1628. Stage V calls for the introduction of diesel particle filters (DPFs) on most diesel engine categories as a result of the decision to regulate particle number on top of particle mass and thus goes forward from Stage IV, for which NO<sub>x</sub> specific EAT like selective catalytic reduction (SCR), seemed adequate to reach demanded emission levels.

## ■ EXPERIMENTAL SECTION

The test engine was a prototype high-speed 4.4 L turbocharged intercooled nonroad diesel engine producing at maximum 100 kW. This is a prototype of an engine designed to fulfill Stage IV emission levels, when combined with a DOC and an SCR system with proper calibration. For detailed characterization two load points of ISO 8178 C1 (Non-Road Steady Cycle, NRSC) were selected; “Mode 1” (2100 rpm, 100% load) and “Mode 7” (1500 rpm, 50% load). These points represent two different operation conditions for the engine and especially for the EAT systems.

To compare different vehicle technologies, particle emissions were studied for four different EAT combinations and two fuels under laboratory conditions. The studied EAT combinations were: no exhaust aftertreatment (No EAT), a diesel oxidation catalyst (DOC), a combination of diesel oxidation catalyst and an SCR system (DOC+SCR), and a combination of a diesel oxidation catalyst, a diesel particulate filter and an SCR system (DOC+DPF+SCR). Details of the EAT components used are provided in Table 1. The DPF was

**Table 1. Properties of Exhaust Aftertreatment (EAT) Components Used in the Study**

	DOC	DPF	SCR
volume (l)	3	10.4	11
cells/in <sup>2</sup>	200	200	350
coatings	Pt–Pd/Al <sub>2</sub> O <sub>3</sub> , 35 g/cft	high porous SiC 58%, Pt 5 g/cft	Cu/ zeolite

a platinum catalyzed wall-flow filter that operates with passive regeneration. SCR dosing rate was selected based on test runs prior to the actual experiments to have constant AdBlue (diesel exhaust fluid) injection rates enabling maximum NO<sub>x</sub> conversion below 10 ppm of NH<sub>3</sub> slip. Thus, NO<sub>x</sub> stoichiometric ratios (a.k.a. ammonium to NO<sub>x</sub> ratios) were selected to be 0.95 and 1.1, in Modes 1 and 7, respectively, in order to reach typical NO<sub>x</sub> reductions.

The engine operated sequentially on two fuels, one being a typical pump-grade fossil diesel fuel and the other a renewable

paraffinic diesel fuel (Table 2). This paraffinic 100% renewable fuel is made primarily from waste and residues in a hydro

**Table 2. Analysis Results of Fossil and Paraffinic Diesel Fuels**

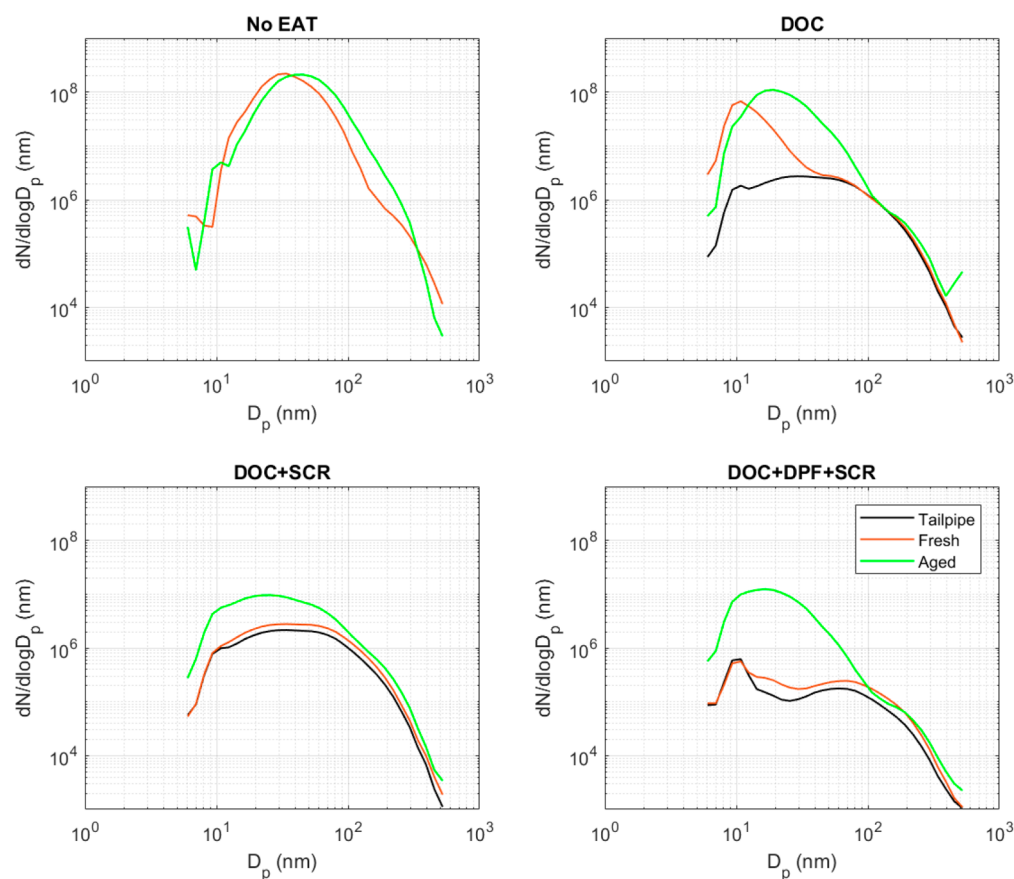
property	analysis method	fossil diesel	paraffinic diesel
density at 15 °C (kg/m <sup>3</sup> )	ENISO12185	841.7	780.0
ash (wt %)	ENISO6245	0.001	<0.001
cetane number	<sup>a</sup> ENISO5165; <sup>b</sup> ASTMD6890	52.4 <sup>a</sup>	77.0 <sup>b</sup>
monoaromatics (wt %)	EN12916	25.1	0.3
polyaromatics (wt %)	EN12916	3.5	<0.1
total aromatics (wt %)	EN12916	28.6	0.3
higher heating value (MJ/kg)	ASTMD4809	45.7	47.3
lower heating value (MJ/kg)	ASTMD4809	42.8	44.0
carbon content (wt %)	ASTMD5291	86.1	84.8
hydrogen content (wt %)	ASTMD5291	13.3	15.2
sulfur content (mg/kg)	NM380	7.0	11.0

treatment process. The major difference in chemical composition between the two fuels relates to their aromatic content. Fossil fuel aromatics content was 28.6 wt %, of which 3.5 wt % was polyaromatics. The paraffinic fuel only contained 0.3 wt % aromatics, which practically contained species with

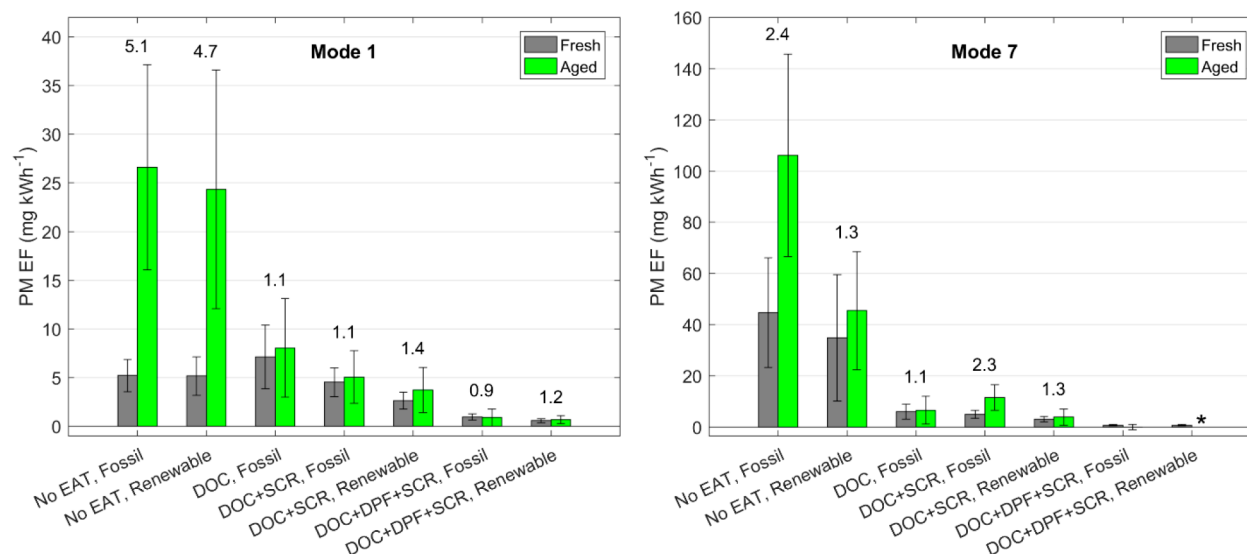
one aromatic ring. Combining density and mass-specific heat enthalpy, the volumetric flow of paraffinic fuel was somewhat higher than the fossil one for the same load conditions.

The exhaust particle sampling and measurement was designed to produce information on tailpipe exhaust aerosol, fresh exhaust aerosol and aged exhaust aerosol.<sup>24</sup> Tailpipe exhaust aerosol includes the primary nonvolatile particles that are found in the particle phase in the hot exhaust gas, and which are counted in the PMP protocol. Fresh exhaust aerosol contains not only the tailpipe aerosol but also the delayed primary PM or PN that are formed in the dilution process with cool air or right after it; in other words, this is the particle population after seconds or minutes from the time of emission. Aged exhaust aerosol is the sum of fresh PM and secondary PM formed due to oxidation processes of gaseous precursors. Secondary aerosol formation typically takes hours or days under atmospheric conditions.

Fresh exhaust aerosol particles were measured downstream of a partial flow dilution system enabling nucleation and condensation processes to take place. In terms of nucleation particle formation, the sampling system mimics atmospheric dilution and the resulting particle size distribution.<sup>25–27</sup> As shown in Figure S1, particle sampling was conducted downstream of each EAT configuration under test. The sampling system consisted of a porous tube diluter (PTD) (primary dilution ratio (DR) 12, dilution air temperature 30 °C), a residence time chamber (2.5 s) to let nanoparticles and grow in size and stabilize their concentration,<sup>28</sup> and a Dekati ejector diluter (DR 5) for secondary dilution. Primary particles



**Figure 1.** Measured particle size distributions of tailpipe, fresh and aged diesel exhaust aerosol with different EAT combinations at the Mode 7 test point with fossil fuel. “Tailpipe” dataserries in panel “No EAT” is not available.



**Figure 2.** PM emission factors of fresh and aged diesel exhaust measured during Mode 1 (high load) and Mode 7 (medium load). Error bars indicate the uncertainty in the mass EF determination (see SI). Numbers above the bars are the calculated aged/fresh PM EF -ratios. \* indicates missing data point.

(nonvolatile particle fraction) were measured downstream of a thermodenuder connected to the dilution system.<sup>29</sup> Secondary aerosol formation was studied with a potential aerosol mass (PAM) oxidation flow reactor (OFR).<sup>18,30</sup> The PAM reactor used here was a 13 l aluminum cylinder equipped with two low-pressure Hg lamps. The reactor was operated in OFR185 mode,<sup>31</sup> which means that the sample is exposed both to 185 and 254 nm UV radiation. The estimated photochemical ages achieved with the PAM reactor were between 2.8 and 4.6 equiv days in the atmosphere (assuming average ambient OH concentration of  $1.5 \times 10^6 \text{ cm}^{-3}$ ) with a few exceptions: for the “no EAT” case at Mode 1, the estimated photochemical age was only 1.2 equiv days, whereas for the cases with all of the EATs (DOC+DPF+SCR), the age was 7.5–13.3 days (see SI for more information). The dilution air for the primary and secondary dilution steps was synthetic air (99.999% purity in gas bottles) to minimize any secondary aerosol formation interference from any impurities in the dilution air.

After sampling and dilution, exhaust particle size distribution (PSD) was characterized by an engine exhaust particle sizer<sup>32</sup> (EEPS; TSI Inc.) with soot inversion matrix<sup>35</sup> in data post processing and two identical electrical low pressure impactors<sup>34</sup> (ELPIs; Dekati Inc.) with improved nanoparticle resolution<sup>35,36</sup> similar as in the state-of-the-art ELPI+ (Dekati Inc.) impactor. A soot particle aerosol mass spectrometer<sup>37</sup> (SP-AMS; Aerodyne Research Inc.) provided the chemical composition of exhaust particles. One ELPI measured downstream of the secondary dilution, whereas the other measured downstream of the PAM (and ejector), thus enabling simultaneous measurement of the fresh and aged exhaust aerosol. Additional details of the measurement setup and associated data analysis are provided in the Supporting Information (SI).

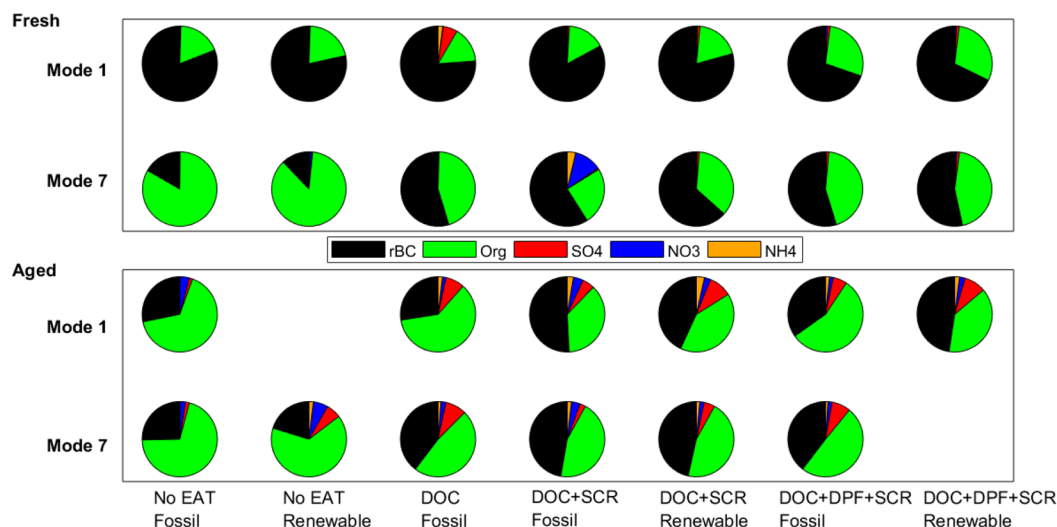
## RESULTS

**Particle Size Distributions (PSDs).** Exhaust number PSDs were measured downstream of all EAT configurations, separately in three sampling branches regarding tailpipe, fresh, and aged aerosol (Figure 1). Without EAT (Figure 1a), the

fresh and aged particle size distributions were fairly similar in shape and dominated by nucleation mode particles, showing also particle growth in the PAM reactor. The tailpipe aerosol particle size distribution was not measured for the case “No EAT”. Downstream of the DOC (Figure 1b), the tailpipe aerosol mainly comprised a soot (accumulation<sup>38</sup>) mode, peaking at sizes of 40–50 nm. There were also some particles around ~10 nm but the EEPS sensitivity and resolution are fairly weak in that size range. In contrast, nucleation mode particles were clearly detected in fresh exhaust that further grew in size and volume as the aerosol aged in the PAM.

When the SCR was fitted downstream of the DOC (Figure 1c), the clearly distinctive nucleation mode disappeared and fresh and tailpipe PSDs were similar. When exhaust aged in the PAM reactor, PSD hardly changed but particle concentration increased. This would indicate either new particle formation in the PAM reactor or particle growth from a size range earlier found below the EEPS measurement range. Fitting the DPF between the DOC and the SCR (Figure 1d) removed most of the soot mode of tailpipe and fresh aerosol but left traces of particles in the nucleation mode size range. This nucleation mode substantially grew in both concentration and size in the PAM reactor, again a possible result of new particle formation or growth from sizes not detected by EEPS.

**PM Emissions Factors.** Emission factors (EFs) for both fresh and aged exhaust particulate matter (PM) are shown in Figure 2. These have been calculated on the basis of size distributions measured by ELPIs applying effective particle density obtained from simultaneous ELPI and EEPS measurement. The detailed procedure for determining the effective density is described in the SI. We note that in Mode 1 with no EAT, the aged mass may be underestimated due to significantly lower OH exposure compared to other cases. Likewise, in Mode 7, the aged mass in the case with DOC +DPF+SCR is not directly comparable to other cases because of very high OH exposure (see the SI for details and sensitivity analysis). Emission factors are presented for both the high (Mode 1) and medium load (Mode 7) modes. In general, both the fresh and aged exhaust PM decreased with the use EATs; e.g., during Mode 1, the fresh and aged exhaust PM decreased



**Figure 3.** Chemical composition of diesel engine exhaust PM during Modes 1 and 7, determined for both the fresh and aged PM. The composition was measured downstream different EAT systems and with both fuels (fossil/paraffinic). The PM composition was measured with SP-AMS.

from 5.2 mg/kWh and 24.4–26.6 mg/kWh, respectively, to about 1 mg/kWh when the most advanced EAT (DOC+DPF+SCR) was implemented. At medium load, PM levels without EAT were in the range of 35–106 mg/kWh, depending on aerosol condition (fresh, aged) and the fuel used (fossil, renewable). Although this is much higher level than at high engine load, implementation of advanced EAT again decreased these levels down to 1 mg/kWh or less.

Use of DOC with fossil diesel seemed to increase fresh PM over no EAT levels in Mode 1 but not in Mode 7. The increase in Mode 1 was associated with high nucleation mode concentration in fresh exhaust and is likely caused by continuous SO<sub>2</sub> oxidation in DOC and/or sulfuric acid release from DOC. The storage and release of sulfur compounds in EAT systems have been observed to significantly affect the exhaust aerosol PSD and concentration.<sup>39</sup>

Despite the inconsistency in the DOC effect on fresh aerosol, overall the use of the DOC decreased the aged PM mass by a factor of 3–16. The addition of the SCR downstream of the DOC seemed to have an inconsistent impact on aged PM mass, whereas the use of the DPF substantially decreased both fresh and aged PM mass. The decrease of fresh PM emissions by the DPF has been well established before. However, our results show that the positive impacts of the DPF are even larger when aged PM is considered. Aged-to-fresh PM ratios in Mode 1 were at maximum 5.1 without EAT (Figure 2) but close to unity with the DOC+DPF+SCR system. With the same EAT configuration, aged PM levels dropped below detection limit in Mode 7. In few cases, the aged-to-fresh PM ratios dropped below one, possibly due to particle losses in the PAM reactor, uncertainties in defining the PAM background aerosol formation and the reduced sensitivity of ELPI due to the additional dilution steps when measuring the aged mass.

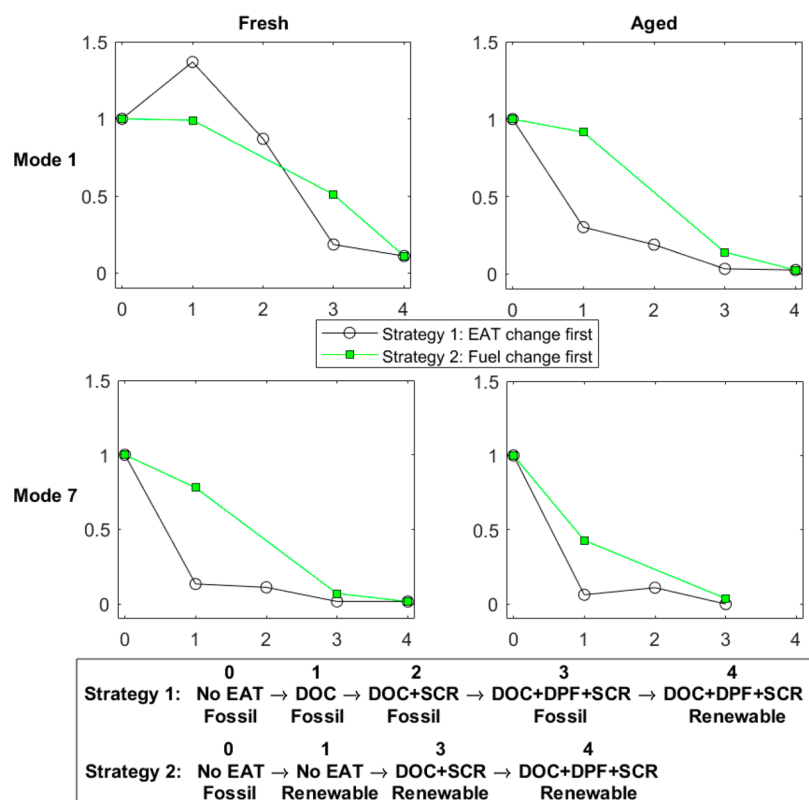
Less impact of fuel specifications on PM could be seen, compared to the corresponding impact of EAT. The fresh exhaust PM EFs were at the same level in Mode 1 for both fuels, but the fresh PM EF was about 20% lower with the renewable paraffinic fuel in Mode 7. For all the EAT systems, the use of paraffinic fuel always resulted in lower aged PM EF.

**Chemical Composition, Fresh Aerosol.** The chemical composition of fresh exhaust aerosol is shown in Figure 3,

grouped in the main chemical groups provided by the SP-AMS, including organics (Org), refractory black carbon (rBC), sulfate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>), and ammonium (NH<sub>4</sub>). The higher fresh PM concentration at medium than at high engine load operation observed earlier was also associated with differences in PM composition. Fresh PM was dominated by rBC at high load (Mode 1), but the addition of organic material significantly increased the total mass in Mode 7 especially when no EAT was used. The high PM reduction in the DOC (see Figure 2, Mode 7) seems to relate to the efficient removal of organics in Mode 7 (Figure 3). Instead, in Mode 1, the level of organics was low, and the relative reduction (change in composition) was not directly observed in the SP-AMS data. The choice of fuel was seen in the amount of rBC in Mode 7, where lower levels were detected with the renewable paraffinic fuel. In general, the concentrations of sulfates, nitrates and ammonium were low in fresh exhaust PM, so that the levels that the SP-AMS actually measured were close to the limit of detection.

Regarding the above-mentioned, relatively high fresh exhaust PM in Mode 1 downstream the DOC was observed. The composition measurement support the hypothesis on the role of sulfate release and its effects on PM, since this was the only measurement situation where the sulfates had elevated concentrations in fresh exhaust PM. Simultaneously also the ammonium had higher contribution to the PM, indicating that the PM partly comprised of ammonium sulfate. One difference in composition was also detected in Mode 7 with DOC+SCR and fossil fuel, where almost 20% of PM was composed of ammonium nitrate.

**Chemical Composition, Aged Aerosol.** The chemical composition of aged exhaust PM is also shown in Figure 3 (bottom panel). Except the measurements during Mode 7 without exhaust aftertreatment (No EAT), the photochemical aging simulated in the PAM chamber increased the fraction of organic compounds in PM. Connecting with results in Figure 2, a clear observation is that the EAT systems reduced the concentrations of SOA precursors (and thus observed SOA), so that organics reduced gradually. The combination DOC+SCR+DPF was extremely efficient with respect to the reduction of SOA formation; even though the DPF filtered majority (~90%) of the soot particles, the average particle



**Figure 4.** Effects of individual technology steps (*x*-axis) in the reduction of fresh or aged PM (normalized *y*-axis). The difference between strategies 1 and 2 is whether fuel or EAT is changed as the first step.

composition was not entirely changed compared to the DOC+SCR situation, indicating that the DPF was almost as effective in removing soot and SOA precursors. It should be noted that the DPF in this study had catalytic coating (see Table 1).

The photochemical aging of exhaust sample in the PAM chamber increased also the fractions of sulfates, nitrates, and ammonium in the aged PM so that they reached clearly elevated and detectable concentrations (Figure 3). This was observed with both low sulfur fuels, indicating that also the lubricant oil may contribute to secondary sulfate. In addition, the sulfate concentrations were so high that it should be taken into account when the total secondary aerosol mass from diesel engine is evaluated. In contrast, it can be seen that the ammonium concentration was not remarkable, even when measured downstream the SCR, indicating low levels of ammonia slip in the SCR.

## DISCUSSION

The study of Timonen et al.,<sup>21</sup> made with GDI passenger car, indicates that the aromatic content of fuel may even determine the emissions of SOA precursors from gasoline-fueled internal combustion engines. According to Gentner et al.,<sup>40</sup> approximately 53% of SOA from diesel engines originate from the aromatics in the fuel. In this study, the experiments were made with two significantly different diesel fuels in composition, one containing 28.6% of aromatics and the other being nearly aromatic-free. As a result, we did not observe as high exhaust SOA formation reduction as expected. We think that this can indicate a higher role of lubricant oil in SOA precursor formation for diesel engine when compared to GDI passenger cars. This is supported by the result that elevated concentrations of sulfates were observed in the aged PM

with both of the low-sulfur tested fuels. However, since fuel compounds can chemically transform during combustion,<sup>41</sup> it is possible that SOA precursors originate from fuel.

Relatively the role of EAT systems was greater than the role of fuel. The installed additional exhaust aftertreatment systems reduced aged PM concentrations step-by-step, finally downstream the DPF the aged PM levels were roughly the same whether the PAM chamber measurement was performed from the diluted exhaust gas or just from the dilution air.

As a strategy to achieve lower fresh or aged PM formation from diesel engines as well as other climate and air quality benefits one can consider to change the fuel (from fossil to renewable (paraffinic)) or add more EAT components (DOC, SCR, and DPF). The fuel can be regulated from the supplier level being quite easy to monitor but the catalyst are owned by users and their functioning is more difficult to quantify for the entire vehicle fleet. Based on our data, the stepwise reduction in emissions by the implementation of advanced technology is shown in Figure 4 where either fuel or aftertreatment is changed as a first step and later additional EATs are applied according to the description in the figure. Apart from Mode 1, fresh, where additional sulfate formation was detected, introduction of the DOC reduced the PM concentrations effectively. The addition of the SCR was also effective but finally the PM was practically removed by adding the DPF. Overall, the fuel change as a first step had only decreasing fresh/aged PM effects.

Based on this study and the literature, gasoline engines have greater secondary aerosol precursor emissions than diesel engines.<sup>16,21–23</sup> In addition, gasoline particle filter (GPF) has not shown similar reduction potential against SOA as DPF.<sup>42</sup> This might be due to several reasons, e.g., that gasoline engines

do not need as high filtration efficiencies or precious metal loading in the filter.

The DPFs have already been used for years in all road diesels in Europe. In the emission regulations of NRMM, the next step in 2019 will be the introduction of nonvolatile particle number limit, which will enforce the DPFs systematically in Europe. Based on the findings of this study and the others, the introduction of DPFs will not only create benefit in terms of primary (or fresh) particle emission but also secondary organic aerosol formation in the atmosphere. Actually, the DPF appears to offer additional benefits in decreasing secondary aerosol formation, over the DOC and DOC+SCR systems. The SOA precursor removal mechanisms in the DPF may include adsorption of precursors on the DPF washcoat or the accumulated soot cake and subsequent oxidation. The contribution of oxidation should be dominant, since we observed no outbursts of SOA emissions during high-load high-exhaust temperature events that are known to lead to desorption of semivolatile compounds. Therefore, the DPF seems to eliminate and not just filter out most of these species. Catalyzed DPFs on road vehicles were also demonstrated to yield very low SOA over transient operation and even some SOA formed during regeneration was attributed to the surplus of fuel used to initiate regeneration rather than the desorption of species from the DPF.<sup>23</sup> So far, the effects of these special occasions like the DPF regeneration events or potential ammonia slip events are beyond the scope of this study, and also the real-world events like highly transient driving conditions<sup>43</sup> and engine cold starts may cause higher primary and secondary PM emission levels.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: [10.1021/acs.est.9b04073](https://doi.org/10.1021/acs.est.9b04073).

Elaborated description of the exhaust sampling system, measurement protocol, and steps in data analysis (PDF)

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### Author Contributions

The research was conducted and the manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

### Notes

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