

Supporting Information: “Strategies to Diminish the Emissions of Particles and Secondary Aerosol Formation from Diesel Engines”

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EXPERIMENTAL SETUP

As shown in Fig. S1, particle sampling was conveyed by a partial exhaust sampling system¹ downstream the aftertreatment to be tested. The sampling system consisted of a porous tube diluter (PTD) (primary dilution ratio (DR) 12, dilution air temperature 30 °C), residence time tube (2.5 s) and secondary dilution conducted by Dekati diluter (DR 5). The dilution air for the primary and secondary dilution was synthetic air (purity 5.0) to avoid secondary aerosol formation from the dilution air. The CO₂ concentration was measured after all dilution steps with a CO₂ analyzer (Sidor, Sick Maihak) to determine the exact dilution ratios.

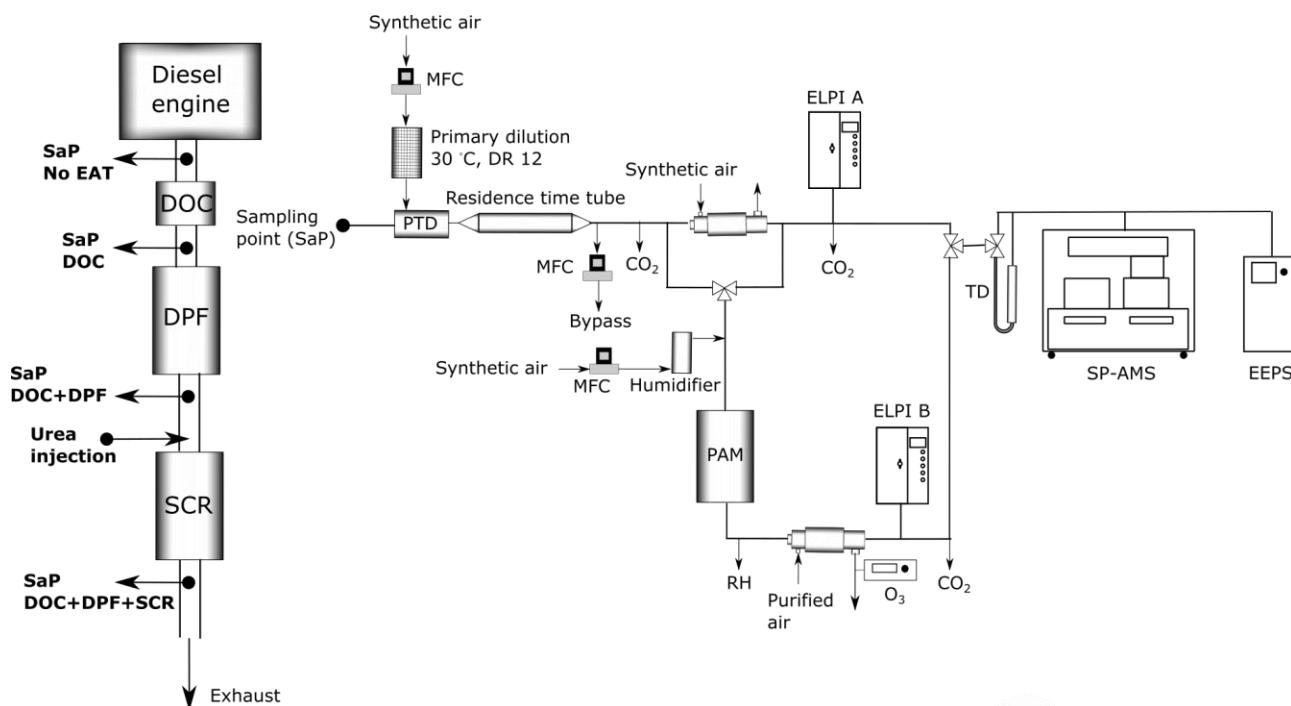


Figure S1. Detailed experimental setup.

A potential aerosol mass (PAM) reactor was used to study secondary aerosol processes downstream of the dilution. The diluted exhaust was led into the PAM either after the PTD (total

DR 12) or after the the secondary dilution (total DR 60), depending on the EAT setup. The higher dilution was used when the exhaust line was not equipped with SCR because of the higher NO_x concentrations in those cases. The NO_x concentration has a significant effect on PAM OH exposure², and thus the additional dilution was necessary to achieve similar OH exposures in all cases. Humidified air was mixed with the sample in prior to the PAM, producing an additional DR of 1.3. The absolute humidity in the PAM reactor was $9.4 \pm 1.6 \text{ g m}^{-3}$. The sample flow through the PAM reactor was set to $\sim 5.5 \text{ l min}^{-1}$ resulting in average residence time of 140 s. The ozone concentration was measured downstream of the PAM with a Model 205 analyzer (2B Technologies).

DESCRIPTION OF THE SP-AMS

The Soot Particle Aerosol Mass Spectrometer^{3,4} (SP-AMS, Aerodyne Research Inc., US) was used to measure the detailed chemical composition of PM. The SP-AMS is described in detail by Onasch et al., 2013⁵. Shortly, the SP-AMS is a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) with an added laser (intracavity Nd:YAG, 1064 nm) vaporizer. Thus, particles are vaporized either by the tungsten vaporizer (600 °C) to analyze non-refractory inorganic species and organics and/or with the laser (intracavity Nd:YAG, 1064 nm) vaporizer in order to analyze refractory black carbon (rBC) and metals in addition to inorganics and organics attached to these particles. In this study, both vaporizers were utilized. The vaporized compounds are ionized using electron impact (EI, 70 eV) ionization. Ions are guided to the time-of-flight chamber and to the multi-channel plate (MCP) detector. AMS measurements were performed in the single-reflection configuration (V-mode) that offers higher sensitivity but lower resolving power (up to ~ 2100 at m/z 200). A one minute averaging time was used in the measurements. For

the SP-AMS the 3σ detection limit for the rBC mass concentration measurements is $0.03 \mu\text{g m}^{-3}$ for 1 min time resolution measurements. For organic and sulfate, detection limits are 1.8 times higher and 0.2 times lower than for rBC, respectively^{4,5}.

CE OF THE SP-AMS

The collection efficiency (CE) of the AMS is a factor applied to the data due to the fact that not all the particles in the sample air are detected by the AMS. There are several reasons why the CE can be lower than unity, e.g. particle lost during the transit through inlet and aerodynamic lens, particles miss the tungsten vaporizer due to the beam divergence and particle bounce at the tungsten vaporizer, and additionally in the SP-AMS a part of the particles can miss the laser vaporizer due to non-optimal particle beam-laser beam overlap. With the tungsten vaporizer the CE depends on the chemical composition and acidity of aerosol as well as on relative humidity of the sample air the CE being typically close to 0.5⁶. For the SP-AMS the CE can be significantly larger than 0.5 due to the laser vaporizer and it has also been noticed that the amount of coating on the rBC particles has an impact on the CE. The factors affecting the CE in the SP-AMS have been discussed e.g. in Onasch et al. (2012)⁵, Willis et al. (2014)⁷ and Ahern et al. (2016)⁸. Since it is not easy to estimate an accurate CE for the fresh combustion particles, the default value of 0.5 was used for the CE in this study.

AMS CALIBRATION

The ionization efficiency of the SP-AMS was determined using monodisperse ammonium nitrate particles. Ammonium nitrate standard was dissolved in purified water (Milli-Q) and the particles were produced with the Constant Output Atomizer (TSI model 3076) operated at a

constant pressure (~2 bar). Particles were dried with the silica gel drier (TOPAS) and a narrow range of electrical mobility particle diameters were selected by using the Differential Mobility Analyzer (DMA, TSI model 3080; set to 300 nm). The mass of monodisperse ammonium nitrate particles was measured by the SP-AMS and the number of particles was determined by the Condensation Particle Counter (CPC, TSI model 3772) in parallel with the SP-AMS. The SP-AMS was calibrated only with ammonium nitrate particles in this study and the concentration of other species were calculated by using the default relative ionization efficiencies⁹ (RIEs; organics 1.4, nitrate 1.1, sulfate 1.2, ammonium 4 and chloride 1.3; Alfarrar et al., 2004). The ions used for the calculation of organics and inorganic species were obtained from the high-resolution peak fitting⁴ by using the ToF-AMS HR Analysis 1.16I Software^{4,10,11}.

Organics in the SP-AMS was corrected for gas-phase CO₂. CO₂ was measured in realtime by the non-dispersive infrared CO₂ analyzer (SickMaihak, SIDOR, range 0-3000 ppm) connected to the same inlet with the SP-AMS.

For refractory black carbon a default RIE of 0.20 was applied⁵. Refractory black carbon was calculated by summing up the carbon fragments from C1 to C8. To account for C1+ that may result from the fragmentation of non-refractory organics, C1+ signal attributed to rBC was constrained to $0.625 \times C3+$ that is the ratio observed for the SP-AMS earlier⁵.

QUALITY CONTROL IN THE MEASUREMENTS

First thing in the morning, the instruments were measuring pure dilution air (PTD operated at flush mode). This was done to make sure that there are no leakages to the sample lines from the lab or that the dilution air does not cause particle concentrations. This “zero background” particle concentration was always practically 0 particles/cc independent of the instrument. This “zero

background” was also performed to verify that dilution air did not contain any CO₂ and at the same time check the baseline for the CO₂ analyzer.

So called “PAM background” was measured every morning prior the experiments, after the “zero background” measurement. During this “PAM background” measurement, the PAM chamber was run exactly the same way as in the normal experiment only difference being that the PTD was run in flush mode that only dilution air related components were measured. The PAM background mass results (previously presented) were used in order to determine the aged PM results during each day.

For SP-AMS, in order to ensure that the measurement results were accurate and reliable there were parameters that were checked during the measurements. Single ion and mass-to-charge calibrations were done on daily basis and also baseline and threshold were set daily. The SP-AMS airbeam value and flow rate were monitored continuously during the measurements. The used airbeam was $\sim 1.20 \times 10^{-7}$ and the sample flow rate was $\sim 1.3 \text{ cc min}^{-1}$.

PAM BACKGROUND MASS

The PAM reactor produces aerosol particles even from pure synthetic air, since it is impossible to reach absolute purity in the dilution air and sampling line surfaces. Thus, a blank experiment is a standard protocol both in oxidation flow reactor studies as well as when using environmental batch reactors (e.g. ^{12–14}) to account for the background aerosol formation. In this study, the background mass (M_{BG}) was measured each day and subtracted from the measured aged mass concentration using the following equation:

$$M_{aged} = \left(M_{measured} - \left(1 - \frac{1}{DR_{PAM}} \right) \times M_{BG} \right) \times DR_{TOT} , \quad (S1)$$

where M_{aged} is the background-corrected aged mass concentration, $M_{measured}$ is the measured mass concentration after PAM, DR_{PAM} is the dilution ratio of the sample entering the PAM reactor and DR_{TOT} is the total dilution ratio. As a clarification, $\left(1 - \frac{1}{DR_{PAM}}\right)$ is the proportion of the dilution air in the sample entering the PAM reactor. The total aerosol mass concentration in the PAM reactor during blank experiments was 1.8-18.0 $\mu\text{g m}^{-3}$ (11.5 $\mu\text{g m}^{-3}$ on average).

PAM OH EXPOSURE AND LVOC LOSSES

The OH exposure is defined as $[\text{OH}] \times t$, where $[\text{OH}]$ is the mean OH radical concentration and t is the mean residence time of the sample in the reactor. Two samples oxidized in the PAM are comparable when their exposure to OH radicals is on the same level. Since no online OH exposure monitoring was performed, the PAM UV lamp intensity was kept constant and the OH exposure was modeled afterwards.

The PAM reactor used here has been characterized earlier, and a photochemical model can relatively well predict the OH exposure¹⁵. The input parameters for the model are PAM relative humidity and temperature, PAM inlet concentrations of NO, NO₂ and CO, and the OH reactivity (OHR) of the hydrocarbons at PAM inlet. The OHR is calculated from the measured total hydrocarbon concentration by assuming a similar exhaust VOC profile as Jathar et al.¹⁴. In addition, the model requires the 254 nm photon flux as an input parameter. The PAM UV lamps were set to full intensity, so the photon flux is approximately 3.4×10^{15} photons $\text{cm}^{-2} \text{s}^{-1}$ according to the earlier characterization. However, using this photon flux here does not always result in a good agreement between the modeled and the measured values of ozone concentrations. Thus, either the maximum photon flux varies between the measurements or some of the assumptions are not satisfied (e.g. the assumption of the VOC profile). As an uncertainty analysis, we varied the

model photon flux for each measurement so that the modeled ozone concentration agrees with the measured one.

Figure S2 shows the modeled OH exposures as equivalent days (assuming average ambient OH concentration of $1.5 \times 10^6 \text{ cm}^{-3}$) and associated uncertainties regarding the photon flux and overall model uncertainty of 30%. For example, the model gives OH exposure of 4.17 days for the case “No EAT, Fossil (Mode 7)” when using 254 nm photon flux of $3.4 \times 10^{15} \text{ photons cm}^{-2} \text{ s}^{-1}$. The 30 % uncertainty gives the lower limit of the error bar, (2.92 days). The modeled and measured ozone concentrations are equal when photon flux of $4.04 \times 10^{15} \text{ photons cm}^{-2} \text{ s}^{-1}$ is used. Then the modeled OH exposure is 5.39 days, and applying the 30% uncertainty gives the upper limit of the error bar (7.01 days).

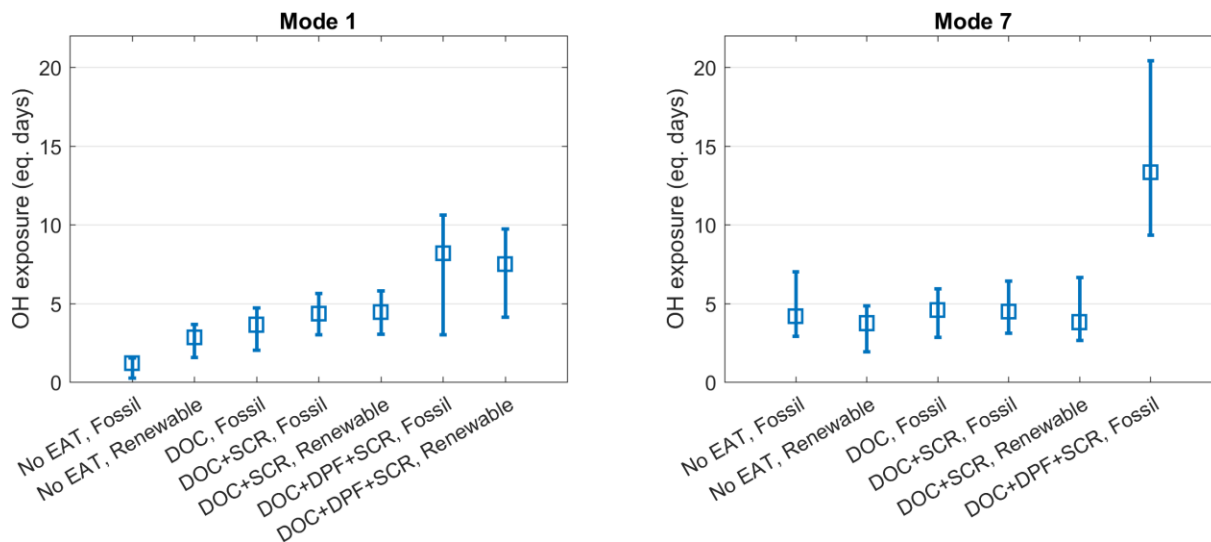


Figure S2. Modeled OH exposures for each measurement. The squares show values obtained by using 254 nm photon flux of $3.4 \times 10^{15} \text{ photons cm}^{-2} \text{ s}^{-1}$ as an input parameter in the model. The error bars represent the uncertainty related to estimated 254 nm photon flux and additional 30% uncertainty in model results.

According to previous research, the magnitude of SOA formation from vehicle exhaust is usually rather insensitive to OH exposure between OH exposures of approximately 3 days and one week^{14,16}. Thus, we expect that most of the cases are comparable despite some differences in the modeled OH exposures. The only exceptions are the “No EAT, Fossil” case in Mode 1, where the secondary aerosol formation potential may be underestimated due to low OH exposure achieved, and the “DOC+DPF+SCR, Fossil” case in Mode 7, where the OH exposure is significantly higher than in the other cases.

As a sensitivity test, we also tested a lower PAM UV lamp intensity for two cases (both fuels in Mode 1, DOC+DPF+SCR), and did not observe significant changes in the outcome (Fig. S3). The negative values in the other case are a consequence of subtracting a background value which is very close to the actual measured value (Eq. (S1)).

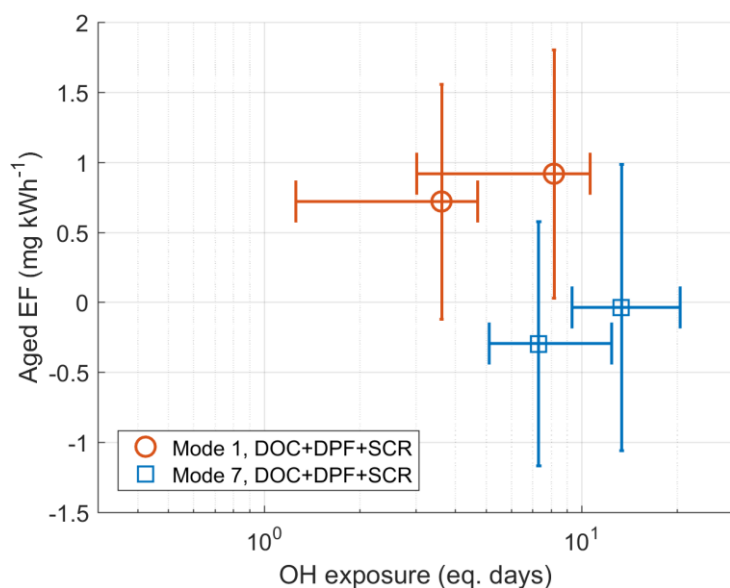


Figure S3. The aged mass as a function of OH exposure for two different cases.

The modeled OH exposures are further used to estimate the losses of low volatile organic compounds (LVOC) formed by oxidation of the VOCs. To measure the total amount of SOA formed in the PAM reactor, all LVOCs should condense to particle phase. However, they can also

condense on PAM walls, fragment before condensing, or exit the reactor before condensing¹⁷. These losses are modeled here by the equations and parameters presented by Palm et al.¹⁷ and assuming an accommodation coefficient of 1.0. The total LVOC losses (i.e. the fraction of LVOCs that do not condense on particles) for each measurement are presented in Fig. S4.

The total LVOC loss is approximately 10% or less in most cases, which means that at least 90% of the formed SOA is measured. When the DPF is installed in the exhaust line, the LVOC loss in the PAM reactor increases up to ~10-55 % because of the decreased condensation sink and high OH exposure. However, in these cases the measured aged mass is almost equal to the fresh aerosol mass, so that it is impossible to correct the data with the estimated LVOC loss. In any case, the amount of secondary aerosol mass in these cases is negligible, so even a high correction factor would not change the conclusions.

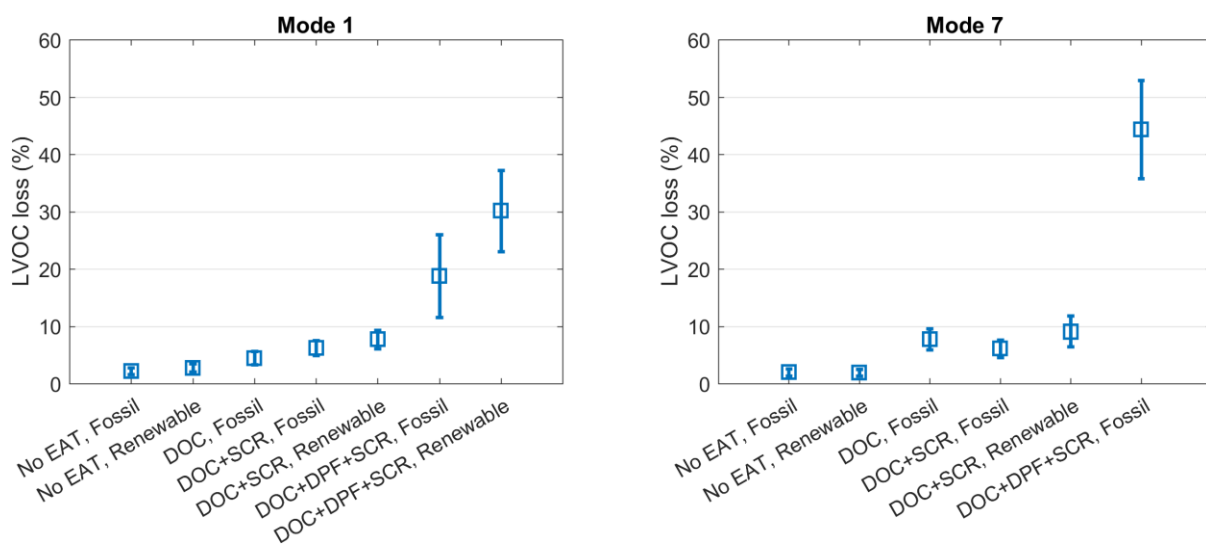


Figure S4. Estimated LVOC loss in the PAM reactor. The error bars reflect the uncertainty in the OH exposure estimation.

MASS CONCENTRATION DETERMINATION

To obtain the mass concentration of particles, the ELPI current measurement was first converted to particle number mobility size distribution. The ELPI charging efficiency and the secondary collection efficiencies of each ELPI stage depend on the mobility size of the particles¹⁸. Thus, it is necessary to convert the ELPI aerodynamic current distribution to mobility current distribution by using the effective density of the particles. The effective density is also needed when converting the EEPS and ELPI number distributions to mass distributions.

Most fresh aerosol number distributions were clearly bimodal according to the EEPS, consisting of a nucleation mode and a soot mode. If the nucleation mode particles are spherical and their composition does not depend on the particle size, their effective density is constant. On the other hand, the effective density of the soot particles depends on the mobility diameter. Thus, we use a constant effective density for the nucleation mode and size-dependent density for the soot mode.

We fitted a log-normal distribution for the EEPS nucleation mode (when bimodal distribution was clearly observed). Then, we estimated the nucleation mode effective density by simulating the ELPI currents of the fitted distribution and varying the density until a good agreement between the simulated ELPI currents and the measured ELPI currents was found^{19,20}. However, this method works only when the soot mode has a minor effect on the ELPI currents, i.e., when the soot mode concentration is low. We found only two measurement points where this condition is fulfilled, and the obtained effective densities of the nucleation mode (ρ_{nucl}) in these measurements were 1.24 g cm⁻³ and 1.57 g cm⁻³. The average of these two densities were applied for all the nucleation modes of the fresh aerosol.

The effective density of the soot particles was assumed to be size-dependent similarly as the coated soot particles²¹, so that

$$\rho_{soot}(D_p) = \left(10.468 \times \left(\frac{D_p}{nm}\right)^{2.55-3}\right) \frac{g}{cm^3}. \quad (S2)$$

The total particle size-dependent effective density is calculated as follows: the soot particle number distribution is obtained by subtracting the fitted nucleation mode from the measured EEPS number distribution, then the density for each particle mobility size is

$$\rho_{eff}(D_p) = \frac{N_{nuc}(D_p) \times \rho_{nuc} + N_{soot}(D_p) \times \rho_{soot}(D_p)}{N_{nuc}(D_p) + N_{soot}(D_p)}, \quad (S3)$$

where N_{nuc} is the concentration of nucleation mode particles and N_{soot} is the concentration of soot mode particles. However, the maximum value for the effective density is limited to that of the nucleation mode.

The particle size distribution of the aged aerosol is always close to unimodal, so it is possible to estimate the effective density for the fitted distribution by simulating ELPI currents with the fitted log-normal distribution for each of the measurement cases. Again, Eq. (S3) is used to calculate the effective density of largest particles that do not belong to the fitted distribution. As an example, we show the measured EEPS distributions for the case “DOC, fossil (Mode 7)”, along with the fitted nucleation modes and calculated size-dependent effective densities in Fig. S5a (fresh aerosol) and Fig. S5b (aged aerosol).

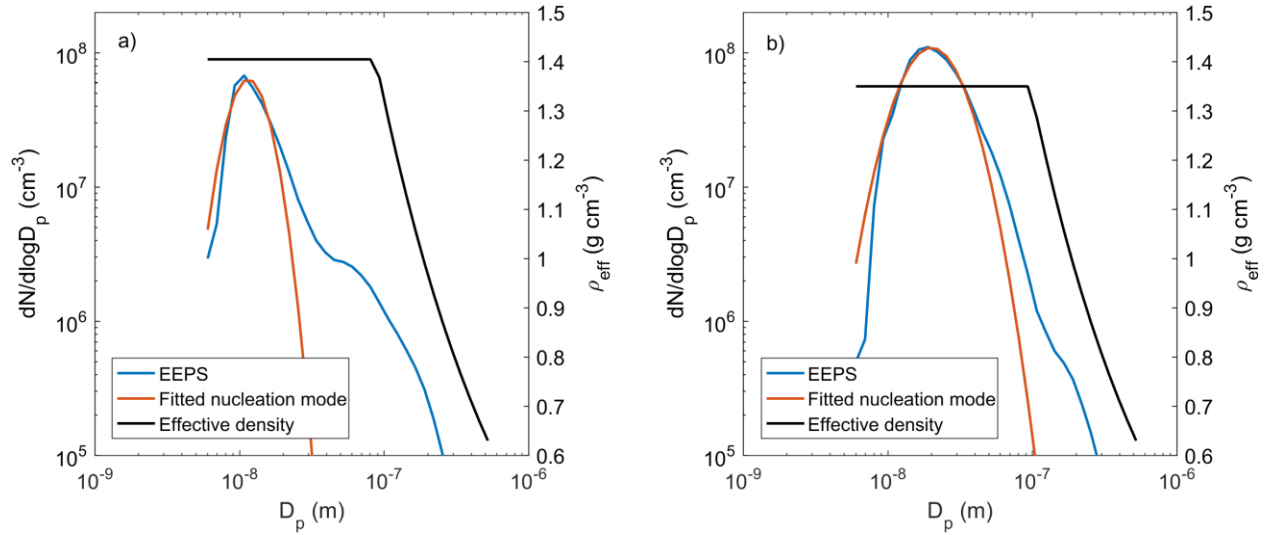


Figure S5. a) Measured EEPS size distribution for the fresh aerosol in case “DOC, Fossil (Mode 7)” and the fitted nucleation mode. Constant effective density of 1.40 g cm^{-3} is assumed for the nucleation mode particles. For the soot mode particles, the effective density is calculated with Eq. (S3), but limited to 1.4 g cm^{-3} . **b)** Measured EEPS size distribution for the aged aerosol in the same case, along with the fitted nucleation mode. Here, the nucleation mode effective density (1.35 g cm^{-3}) is obtained by simulating the ELPI currents for the fitted nucleation mode.

The obtained effective densities are applied into ELPI inversion to convert the current distribution into number distribution. If the corrected current for any stage is less than 2.35% of the total current measured, the data for that stage is neglected. Also, the data from stages 11 and 12 (cut points $1.6 \text{ }\mu\text{m}$ and $4 \text{ }\mu\text{m}$, respectively) are neglected. These small residual currents result from inaccuracies in the secondary collection correction, but they can have a high impact on the total mass if the cut point of the corresponding stage is large. For the same reason, the EEPS mass distribution is only calculated for the range from 5.6 nm to 340 nm , neglecting the two highest size-channels, where a small number concentration can result in a very high mass.

After converting the EEPS and ELPI number distributions into mass distributions by using the effective densities, we get a good agreement between ELPI and EEPS mass concentration measurements, as shown in Fig. S6. The EEPS mass in all cases but one is within 30% of the ELPI mass. Thus, a 30% uncertainty in determining the mass from ELPI current measurement is used for the error bars in Fig. 2.

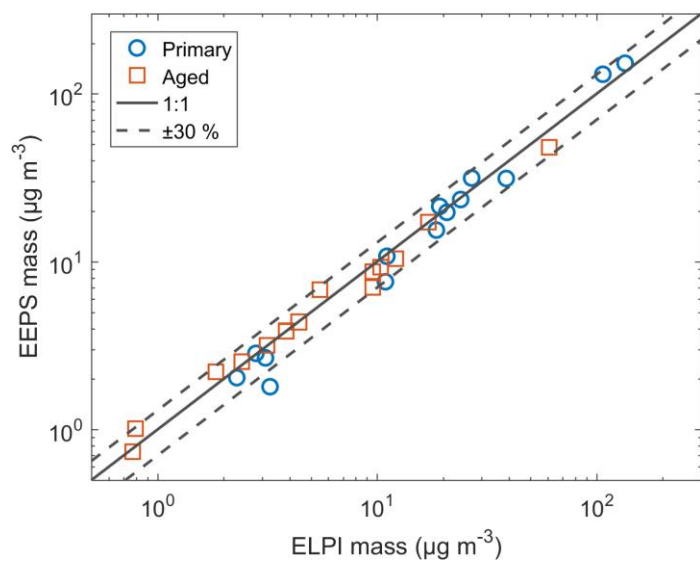


Figure S6. Comparison between ELPI and EEPS mass concentrations.

Figure S7 shows the comparison between EEPS and ELPI when the PAM background aerosol formation from dilution air is measured. The agreement is not as good in these cases as in the other measurements. Thus, an additional 50% uncertainty in PAM background mass is used for the error bars of the aged mass EF in Fig. 2.

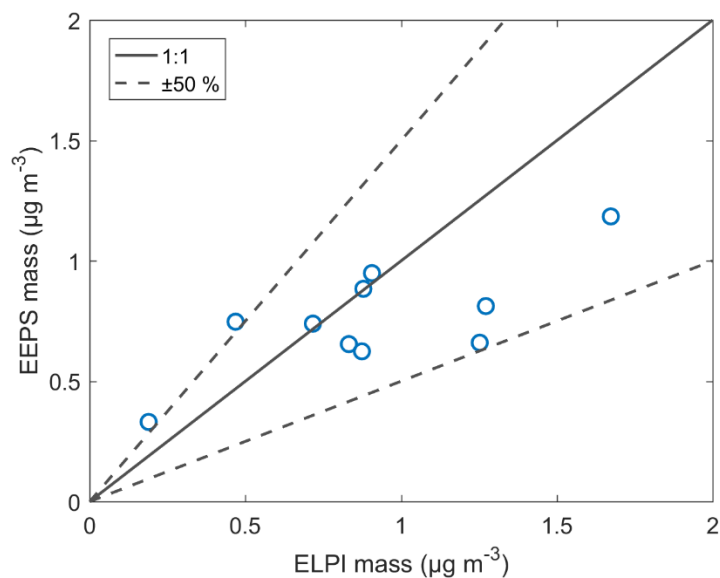


Figure S7. Comparison between ELPI and EEPS mass when PAM background is measured.

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