Supplemental Information for

Changes in CCN activity of ship exhaust particles induced by fuel sulfur content reduction and wet scrubbing

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List of Acronyms

Table S1: List of acronyms used within the manuscript.

Acronym	Description
PN	Particle number
PM	Particulate matter
FSC	Fuel sulfur content
CCN	Cloud condensation nuclei
SMPS	Scanning mobility particle sizer
CCNc	Cloud condensation nuclei counter
SFCA	Scanning flow CCN analysis (operation mode for the CCNc)
STXM	Scanning transmission X-ray microscopy
NEXAFS	Near edge X-ray absorption fine structure
UVSOR	Synchrotron Facility in Okazaki, Japan used for STXM/NEXAFS measurements
MAX IV	Synchrotron Facility in Lund, Sweden used for STXM/NEXAFS measurements
IMO	International maritime organisation
SECA	Sulfur emission control area
HGO	Heavy gas oil (distillate fuel with FSC = 0.86%)
MGO	Marine gas oil (distillate fuel with FSC $<$ 0.03%)
HVO	Hydrotreated vegetable oil (distillate fuel with FSC $<$ 0.03%)
WS	Wet Scrubber (only operated in conjunction with HGO)
SWS	Wet scrubber operated with seawater
FWS	Wet scrubber operated with freshwater
HFO	Heavy fuel oil (residual fuel oil used in commercial shipping)
MK1/MK3	Swedish environmental class distillate fuels
PSD	Particle size distribution
EF_{PN}	Particle number emission factors
EF_{CCN}	CCN emission factors
SS	Supersaturation
SS_c	Critical supersaturation (50% of activated fraction)
κ	Dimensionless hygroscopicity parameter
AF	Activated fraction
AS	Ammonium sulfate

Wet Scrubber PSD Fits

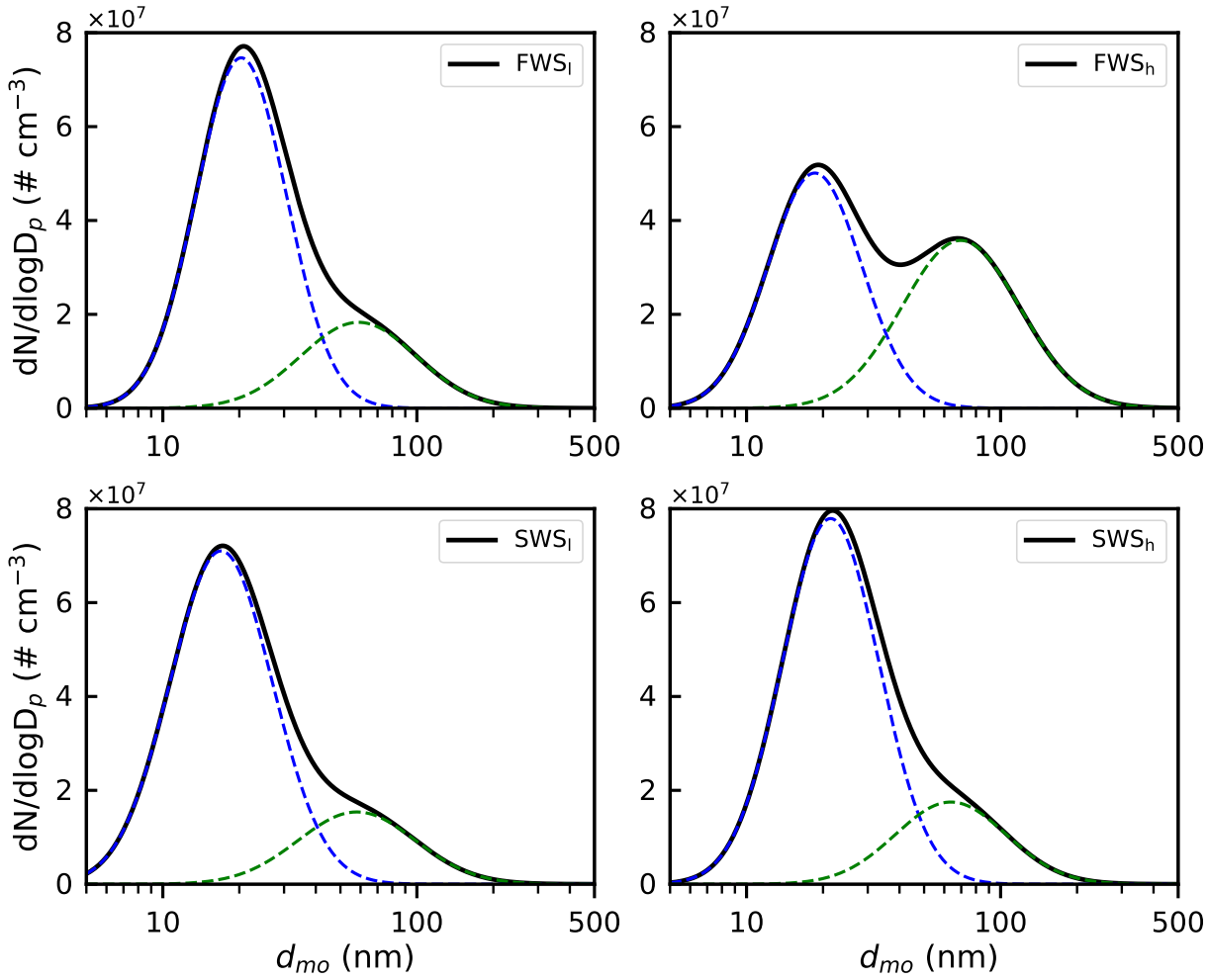

Figure S1: Bimodal lognormal fits of particle size distributions (PSDs) for all four wet scrubber cases from Figure 2 are replotted to illustrate the underlying unimodal distributions.

Particle Number Emission Factors

Particle Number Emission Factors (EF_{PN}) shown in Figure 3 are replotted in Figure S2.

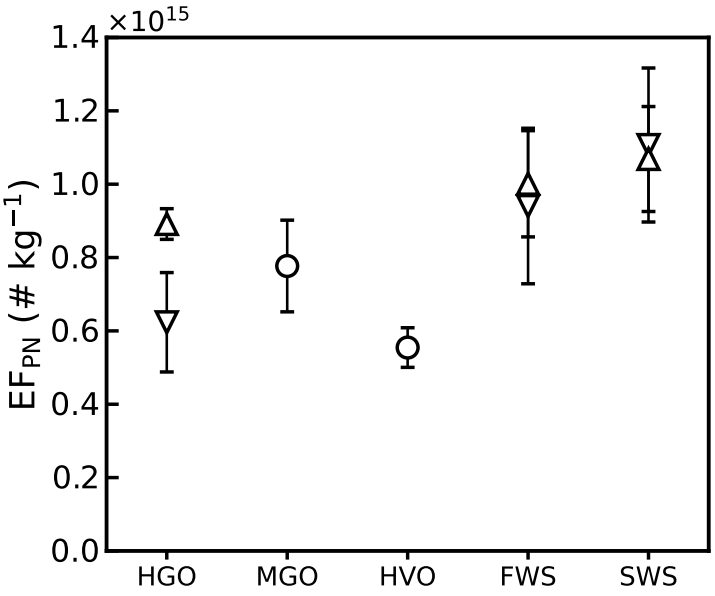

Figure S2: Particle number emission factors (EF_{PN}) that were calculated from lognormal least-squares fits (Figure 3) are replotted for better visibility. Error bars represent ± two standard deviations. Observed differences in combustion conditions for HGO, FWS and SWS are denoted by downward triangles for relatively low CO_2 concentrations, e.g. HGO_l, and upward triangles for relatively high CO_2 concentrations, e.g. HGO_h.

CCNc Calibration

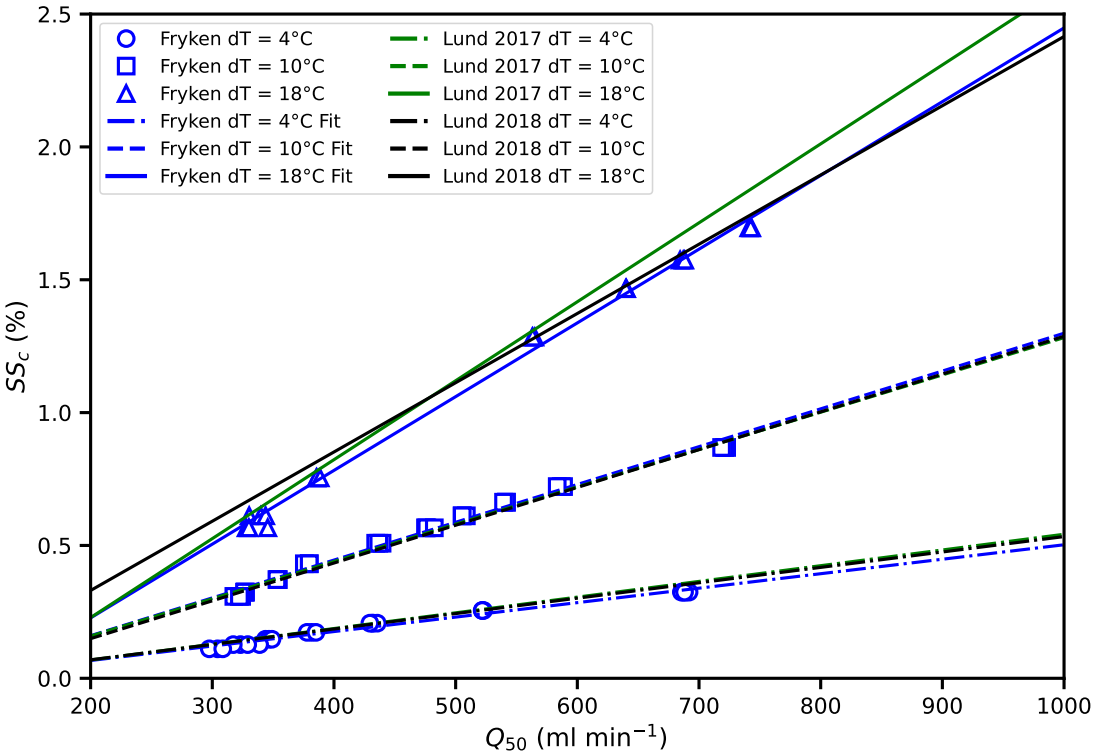

Figure S3: CCNc calibration measurements performed with ammonium sulfate (AS) at ΔT=4 K, 10 K and 18 K. The calibration data is compared to two previous calibrations conducted at the aerosol science lab at Lund University, Sweden.

The DMT CCN counter (CCNc) was calibrated as described in Korhonen et al.[1] Polydisperse $(NH_4)_2SO_4$ (AS) particles were generated using a TSI model 3079A aerosol generator and size-selected with an electrostatic classifier (Model 3080L, TSI Inc., USA) prior to entering the CCNc. The results are shown in Figure S3. Fit parameters for each ΔT setting as well as the functions used to calculate SS are given below:

$$SS(Q)_{4°C} = 0.000545 * Q - 0.0419$$

$$SS(Q)_{10°C} = 0.001422 * Q - 0.1242$$

$$SS(Q)_{18°C} = 0.00278 * Q - 0.3278$$

Hygroscopicity parameter (κ) fits

To calculate CCN emission factors (EF_{CCN}) for given particle size distributions, a simple model was applied where the hygroscopicity parameter κ was estimated for mobility diameters (d_{mo}) between 50 and 150 nm using an exponential fit function (Equation S1),

$$\kappa(d_{mo}) = a * e^{-d_{mo}*b} + c \ . \tag{S1}$$

Fit parameters a, b and c were estimated using a least-squares minimization function in Python. The following equations were used to calculate κ for all particle sizes,

$$\kappa_{fit}(d_{mo}) = \begin{cases} \kappa(50 \text{ nm}) & \text{for } d_{mo} < 50 \text{ nm} \ , \\ \kappa(d_{mo}) & \text{for } 50 \text{ nm} \leq d_{mo} \leq 150 \text{ nm} \ , \\ \kappa(150 \text{ nm}) & \text{for } d_{mo} > 150 \text{ nm} \ , \end{cases} \tag{S2}$$

where constant κ values equal to the respective threshold values were assumed for d_{mo} smaller than 50 nm and larger than 150 nm, respectively. Estimated κ values based on Equation S2 as well as average κ values derived from CCNc measurements are shown in Figure S4 for HGO, MGO, FWS and SWS. For HVO no CCN activity could be measured, hence the absence of this case in Figure S4.

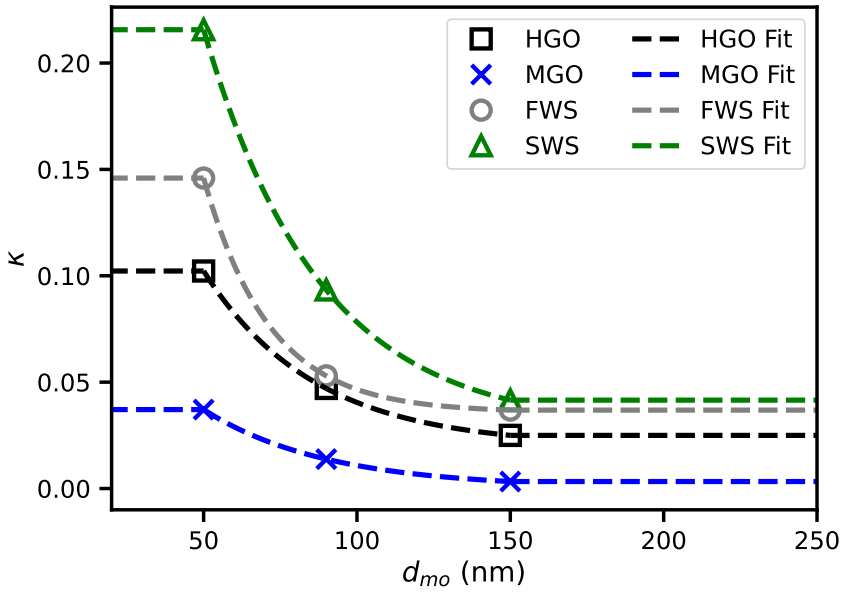

Figure S4: Calculated average κ values for HGO, MGO, FWS and SWS for size-selected particles at $d_{mo} = 50$, 90 and 150 nm represented by markers. Data points were interpolated using least-squares exponential fit functions (Equation S1). For d_{mo} smaller than 50 nm and larger than 150 nm, constant κ values equal to the respective threshold values were assumed. Fitted and extrapolated data are represented by dashed lines.

Images and morphology of exhaust particles

Scanning transmission X-ray microscopy (STXM) images of MGO, HGO and SWS particles are shown in Figure S5. Images indicate that wet scrubbing leads to generally smaller and denser, more compact particles.

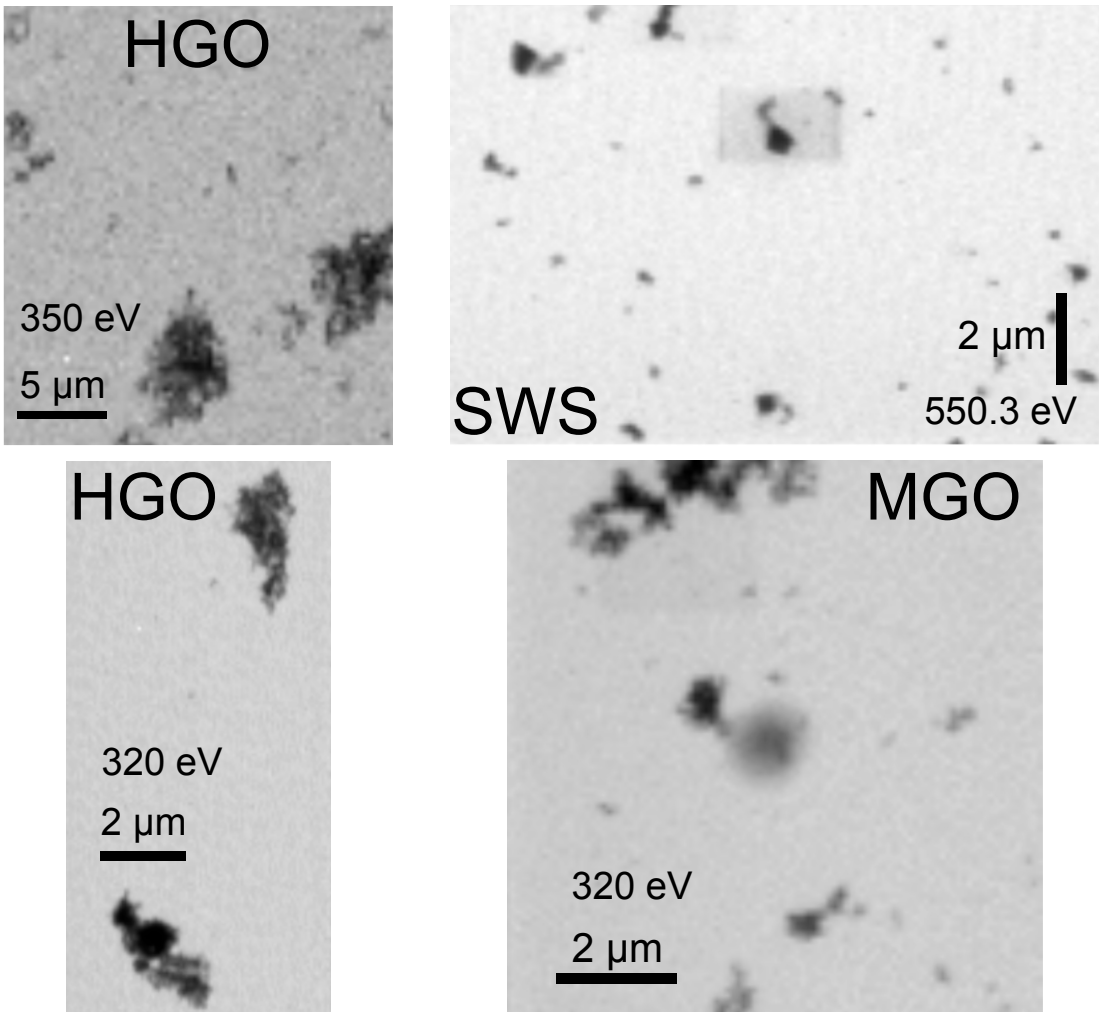

Figure S5: STXM images of exhaust particles for MGO, HGO and SWS.

Near-edge X-ray absorption fine structure (NEXAFS) spectra of soot particles

Near-edge X-ray absorption fine structure (NEXAFS) spectra for oxygen, nitrogen, sodium and chlorine for typical soot particles are shown in Figure S6.

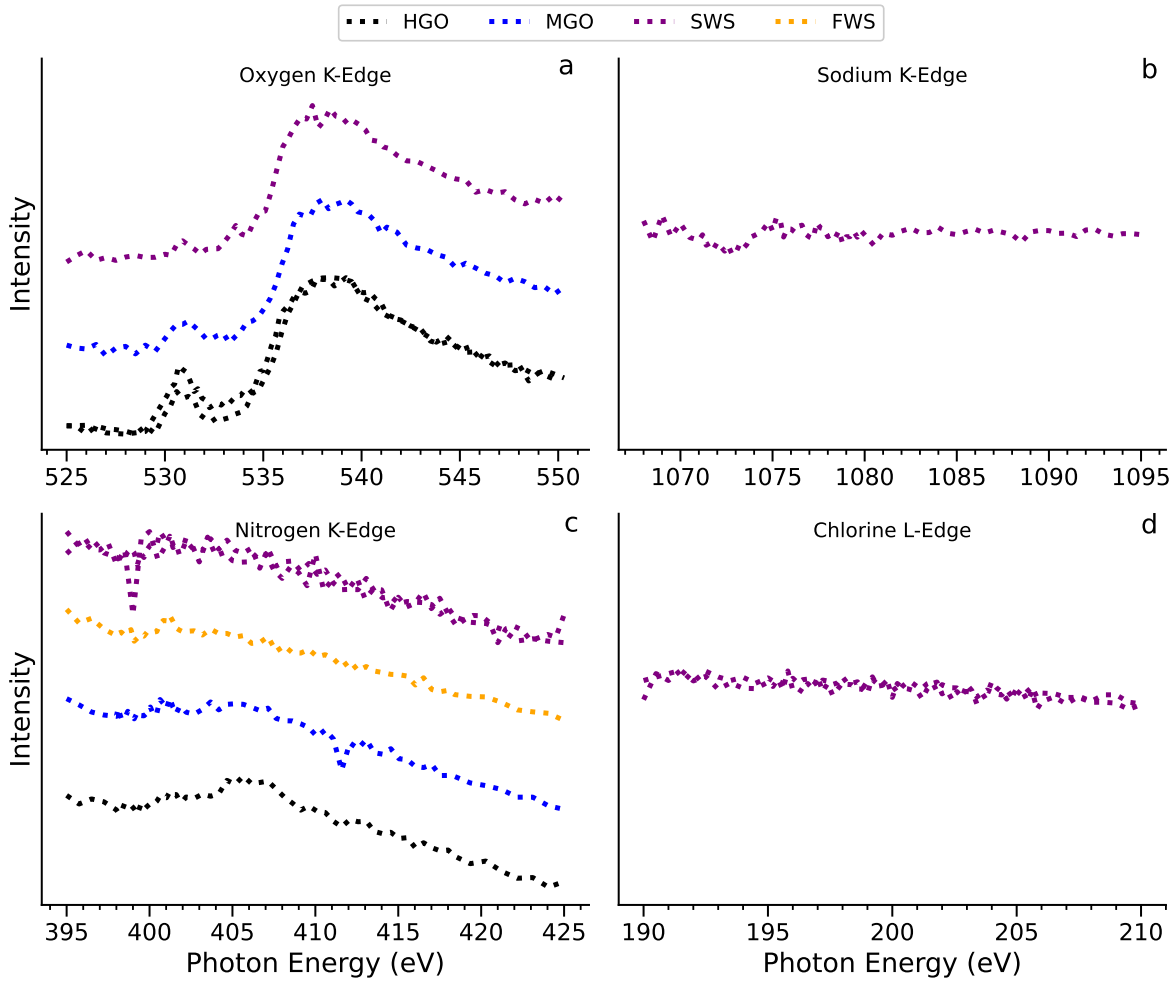

Figure S6: Near-edge X-ray absorption fine structure (NEXAFS) spectra of (a) oxygen, (b) chlorine, (c) nitrogen and (d) chlorine for HGO, MGO, FWS and SWS particles. In panels with more than one case, case-specific spectra have been shifted in their y-positions.

Inhomogeneous SWS particle

Figure S7 shows an example of an inhomogeneous SWS particle consisting of a cubic-shaped salt containing particle marked by the blue rectangle, a column-shaped mineral particle (orange rectangle) and soot (black rectangle). The carbon and calcium spectra of the orange region, or column-like particle, agree well with those of carbonate mineral particles.[2]

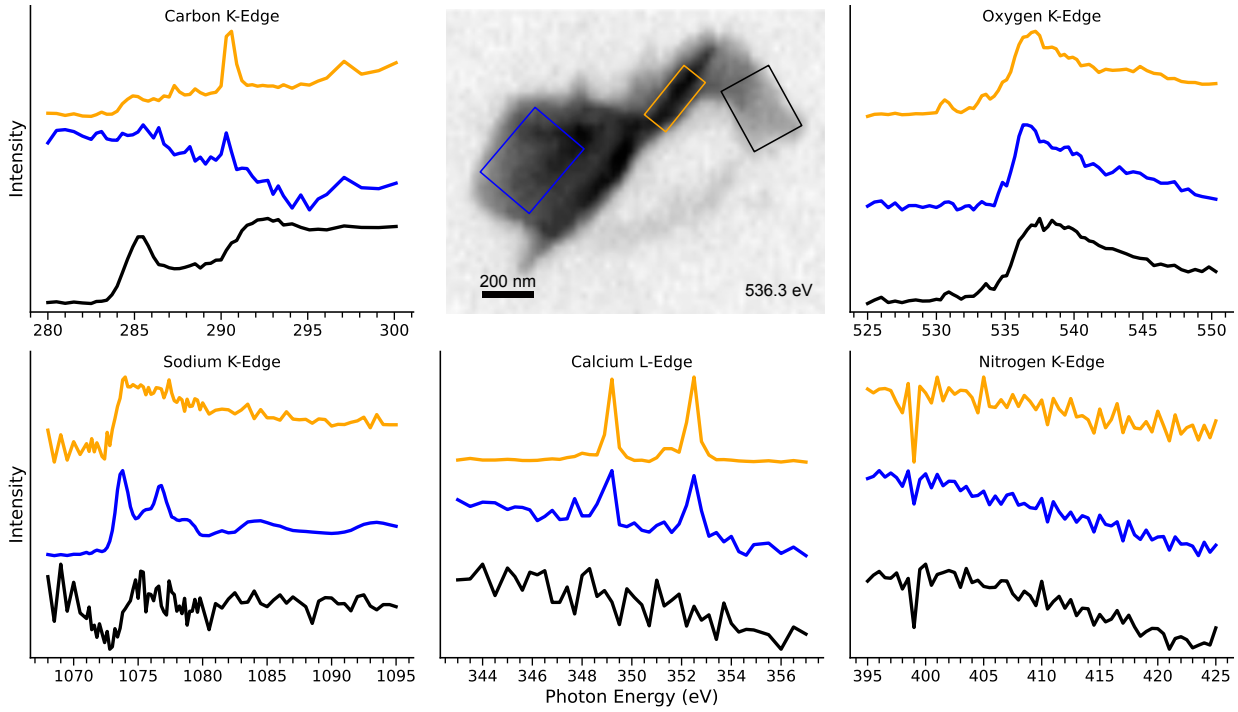

Figure S7: Carbon, oxygen, sodium, calcium and nitrogen Near-edge X-ray absorption fine structure (NEXAFS) spectra of an inhomogeneous SWS particle displayed in the top center panel. Individual spectra correspond to the region marked on the particle image with the respective color. Spectra in each panel have been shifted in their y-positions.

References

(1) Korhonen, K. et al. Particle emissions from a modern heavy-duty diesel engine as ice nuclei in immersion freezing mode: a laboratory study on fossil and renewable fuels. *Atmospheric Chemistry and Physics* **2022**, *22*, 1615–1631.

(2) Brandes, J. A.; Wirick, S.; Jacobsen, C. Carbon K -edge spectra of carbonate minerals. *Journal of Synchrotron Radiation* **2010**, *17*, 676–682.