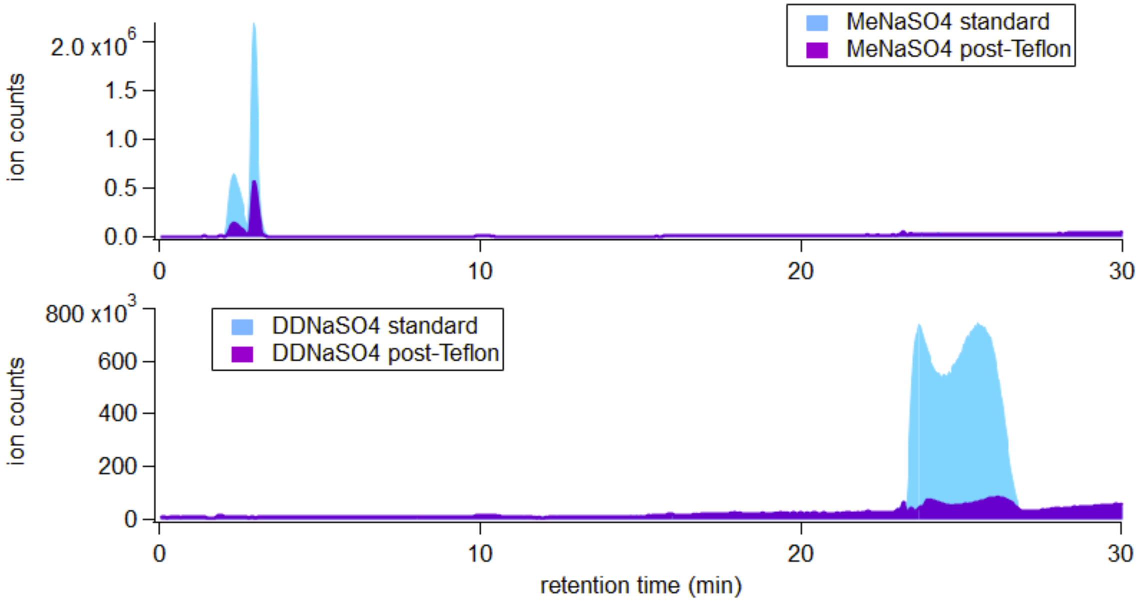

Figure S1: Base peak chromatograms (BPCs) comparing standards and atomized samples of methyl sodium sulfate (top) and dodecyl sodium sulfate (bottom). Samples have a lower signal intensity compared to background, but no major additional or missing peaks.

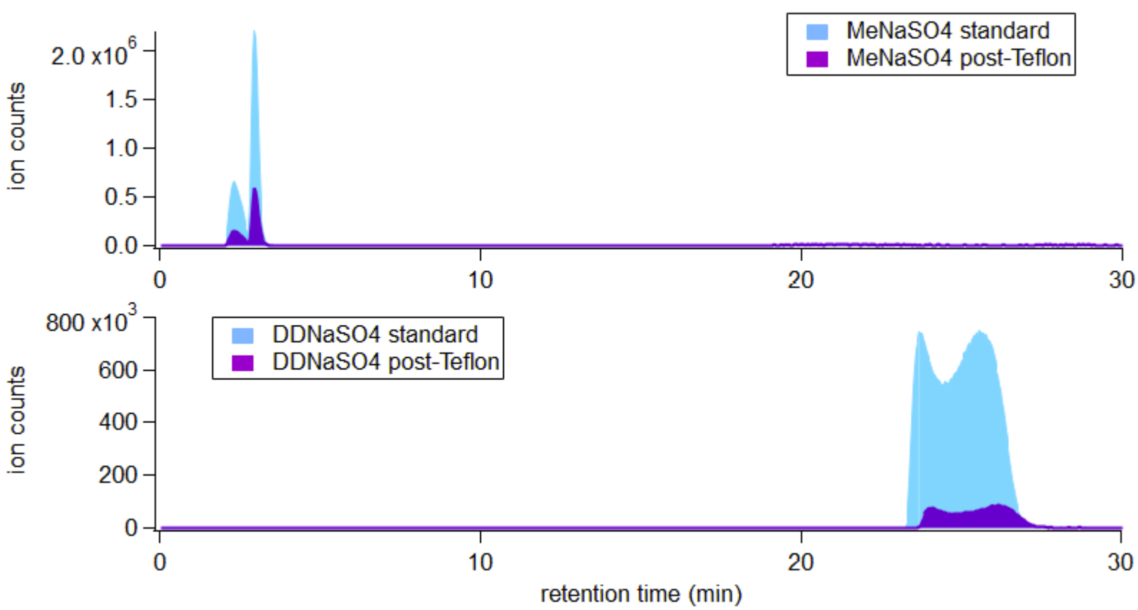

Figure S2: Extracted ion chromatographs (EICs) comparing standards and atomized samples of methyl sodium sulfate (top) and dodecyl sodium sulfate (bottom). Samples have a lower signal intensity compared to background, but no major additional or missing peaks.

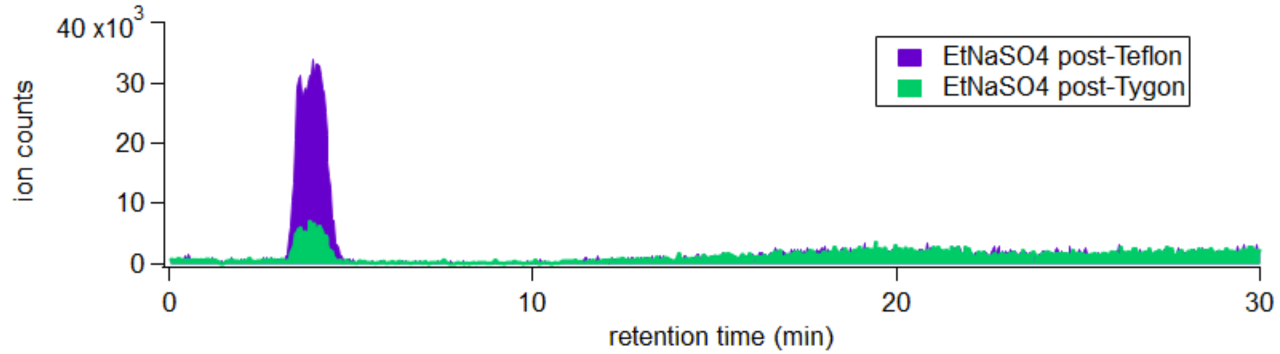

Figure S3: EICs comparing Teflon- and Tygon-atomized samples of ethyl sodium sulfate with similar masses collected on filters.

RPLC/ESI-HR-QTOFMS Filter Analysis

To ensure that standards were not being degraded during atomization in MeOH, we collected filter samples and compared them to the corresponding pure standards by using reverse-phase liquid chromatography coupled to electrospray ionization high resolution-quadrupole time-of-flight mass spectrometry (RPLC/ESI-HR-QTOFMS). We collected dried atomized sample on Teflon filters (47-mm diameter, 0.2-µm pore size; PALL Corp.) for 40 min at a flowrate of 0.3 L min^{-1} to obtain a minimum sample mass of 100 µg and stored at -20 °C until analysis. Filters were extracted by sonicating in MeOH for 45 minutes, after which the extracts were dried and reconstituted in 50:50 MeOH:water. For RPLC/ESI-HR-QTOFMS analysis we used a Waters ACQUITY UPLC HSS T3 column (2.1 × 100 mm, 1.8 µm particle size) and an Agilent 6520 Series LCMS system, at a skimmer voltage of 130 V and a fragmentor voltage of 65 V. Mobile phase A was water and mobile phase B was MeOH, both with 0.05% v/v acetic acid. The 30-min method ran at 0.2 mL min^{-1}: from 0-5 min the solvent ratio held at 0% B, after which it rose to 90% B for the next 10.5 min and then to 100% B for the next 4.5 min, where it held for 9.5 min and then dropped to 0% B in 0.5 min.

Methyl sodium sulfate and dodecyl sodium sulfate samples showed no significant degradation compared to the standards (Figures S1 and S2), though the standards and samples had different concentrations. While an authentic standard for ethyl sodium sulfate was not available at the time of analysis, a comparison of Teflon and Tygon atomization tubing showed that at approximately the same sample concentration Teflon tubing resulted in significantly less degradation (Figure S3).

Table S1: Summary of SPIN OPC scattering parameters used in preliminary classification of particles to initialize machine learning model test dataset. For all columns, x represents the averaged particle-by-particle value for that 1 sec interval from the SPIN OPC. In the case of that droplet class, all data must be above or equal to water saturation.

Class	δ_{SPIN}	$\log_{10}(I_{size})$	S_{liq}
Aerosol	$x < 0.16$	$x \leq 0.125$	-
Droplet	$0.16 \leq x \leq 0.4$	$0.4 \leq x$	$1 \leq x$
Ice	$0.4 \leq x$	$0.4 \leq x$	-
Water Uptake	$x < 0.16$	$0.125 \leq x \leq 0.4$	-

Table S2: Summary of citric acid experiments performed. Columns from left to right indicate the following: tested compound, generation method, glass transition temperature of the dry organic-water mixture (if applicable), PCU chamber temperature, determined class, activation onset ice supersaturation for assigned class of particles (droplet breakthrough, ice formation, or water uptake), onset temperature for assigned class of particles, PCU chamber RH, inlet of PCU RH, geometric mean diameter of size distribution, geometric standard deviation of size distribution, and total particle concentration entering the SPIN.

Compound	Generation Method	$T_{g,org}$ (°C)	PCU Temperature (°C)	Class	Onset S_{ice}	Onset Temperature (°C)	PCU RH (%)	PCU Inlet RH (%)	D_{pg} (μm)	σ_g	CPC (n cm^{-3})
ammonium bisulfate	Atomizer	-	24.3 ± 1	Ice	1.37 ± 0.1	-44.38 ± 0.46	0 ± 5	0 ± 5	0.274	1.58	4572
ammonium bisulfate	Atomizer	-	23.6 ± 1	Ice	1.39 ± 0.11	-39.61 ± 0.51	0 ± 5	0 ± 5	0.278	1.56	4301
ammonium bisulfate	Atomizer	-	23.5 ± 1	Ice	1.44 ± 0.13	-34.46 ± 0.54	0 ± 5	1 ± 5	0.277	1.63	4151
ammonium bisulfate	Atomizer	-	23.5 ± 1	Ice	1.31 ± 0.09	-44.67 ± 0.4	0 ± 5	0 ± 5	0.269	1.52	15559
ammonium bisulfate	Atomizer	-	23.5 ± 1	Ice	1.35 ± 0.09	-39.86 ± 0.44	0 ± 5	0 ± 5	0.267	1.58	15552
ammonium bisulfate	Atomizer	-	23.5 ± 1	Droplet	1.44 ± 0.13	-34.48 ± 0.56	0 ± 5	1 ± 5	0.28	1.57	4093
ammonium bisulfate	Atomizer	-	23.5 ± 1	Droplet	1.52 ± 0.14	-34.58 ± 0.52	0 ± 5	0 ± 5	0.267	1.62	16240
ammonium bisulfate	Atomizer	-	24.2 ± 1	Water Uptake	1 ± 0.03	-44.77 ± 0.15	0 ± 5	0 ± 5	0.275	1.56	4550
ammonium bisulfate	Atomizer	-	23.6 ± 1	Water Uptake	1 ± 0.02	-39.9 ± 0.12	0 ± 5	0 ± 5	0.277	1.56	4103
ammonium bisulfate	Atomizer	-	23.5 ± 1	Water Uptake	1 ± 0.02	-34.82 ± 0.11	0 ± 5	1 ± 5	0.279	1.57	4055
ammonium bisulfate	Atomizer	-	23.5 ± 1	Water Uptake	1.02 ± 0.03	-44.95 ± 0.17	0 ± 5	0 ± 5	0.271	1.51	15527
ammonium bisulfate	Atomizer	-	23.6 ± 1	Water Uptake	1.01 ± 0.05	-39.95 ± 0.31	0 ± 5	0 ± 5	0.269	1.55	15643
ammonium bisulfate	Atomizer	-	23.5 ± 1	Water Uptake	1.01 ± 0.05	-34.94 ± 0.3	0 ± 5	0 ± 5	0.266	1.61	15505
ammonium sulfate	Atomizer	-	24.2 ± 1	Ice	1.26 ± 0.08	-44.89 ± 0.4	0 ± 5	0 ± 5	0.275	1.66	7200
ammonium sulfate	Atomizer	-	24.7 ± 1	Ice	1.27 ± 0.09	-39.87 ± 0.43	0 ± 5	0 ± 5	0.27	1.71	7016
ammonium sulfate	Atomizer	-	24.8 ± 1	Ice	1.41 ± 0.1	-34.94 ± 0.48	0 ± 5	0 ± 5	0.267	1.77	7750
ammonium sulfate	Atomizer	-	24.8 ± 1	Droplet	1.44 ± 0.11	-34.72 ± 0.48	0 ± 5	0 ± 5	0.267	1.76	7617
ammonium sulfate	Atomizer	-	24.6 ± 1	Water Uptake	1 ± 0.02	-44.76 ± 0.14	0 ± 5	0 ± 5	0.269	1.77	7098
ammonium sulfate	Atomizer	-	24.7 ± 1	Water Uptake	1 ± 0.04	-39.77 ± 0.24	0 ± 5	0 ± 5	0.27	1.76	7406

ammonium sulfate	Atomizer	-	24.8 ± 1	Water Uptake	1.05 ± 0.04	-34.85 ± 0.27	0 ± 5	0 ± 5	0.271	1.71	8138
citric acid	Atomizer	-13 ± 10	-65.7 ± 1.1	Ice	1.39 ± 0.1	-44.77 ± 0.42	-	0 ± 5	0.222	1.26	8692
citric acid	Atomizer	-13 ± 10	-65.7 ± 1.1	Ice	1.38 ± 0.1	-39.74 ± 0.45	-	0 ± 5	0.221	1.25	9572
citric acid	Atomizer	-13 ± 10	23.2 ± 1	Droplet	1.57 ± 0.13	-45.43 ± 0.54	0 ± 5	0 ± 5	0.202	1.3	12048
citric acid	Atomizer	-13 ± 10	23.4 ± 1	Droplet	1.5 ± 0.11	-40.05 ± 0.45	0 ± 5	0 ± 5	0.199	1.3	8970
citric acid	Atomizer	-13 ± 10	23.3 ± 1	Droplet	1.44 ± 0.13	-35.06 ± 0.51	0 ± 5	0 ± 5	0.204	1.3	14547
citric acid	Atomizer	-13 ± 10	-65.9 ± 1.1	Droplet	1.52 ± 0.15	-34.63 ± 0.53	-	0 ± 5	0.22	1.24	12842
citric acid	Atomizer	-13 ± 10	23.2 ± 1	Water Uptake	1.22 ± 0.07	-45.1 ± 0.39	0 ± 5	0 ± 5	0.202	1.3	11542
citric acid	Atomizer	-13 ± 10	23.4 ± 1	Water Uptake	1.3 ± 0.09	-40 ± 0.41	0 ± 5	0 ± 5	0.2	1.3	8631
citric acid	Atomizer	-13 ± 10	23.2 ± 1	Water Uptake	1.08 ± 0.07	-35.06 ± 0.38	0 ± 5	0 ± 5	0.203	1.3	12986
citric acid	Atomizer	-13 ± 10	-30.4 ± 1.1	Water Uptake	1.18 ± 0.07	-44.79 ± 0.36	-	0 ± 5	0.201	1.31	10517
citric acid	Atomizer	-13 ± 10	-30.3 ± 1.1	Water Uptake	1.26 ± 0.09	-34.85 ± 0.45	-	0 ± 5	0.205	1.3	10053
citric acid	Atomizer	-13 ± 10	-65.8 ± 1.1	Water Uptake	1.03 ± 0.05	-44.87 ± 0.31	-	0 ± 5	0.223	1.25	9378
citric acid	Atomizer	-13 ± 10	-65.6 ± 1.1	Water Uptake	1.34 ± 0.1	-34.72 ± 0.46	-	0 ± 5	0.219	1.24	11177
citric acid, anhydrous	145 (°C), 0.1 L min-1	-13 ± 10	-70.4 ± 1.1	Ice	1.23 ± 0.08	-44.83 ± 0.37	-	0 ± 5	0.123	1.64	16909
citric acid, anhydrous	145 (°C), 0.1 L min-1	-13 ± 10	-70 ± 1.1	Ice	1.26 ± 0.08	-39.82 ± 0.42	-	0 ± 5	0.14	1.68	13586
citric acid, anhydrous	140 (°C), 0.1 L min-1	-13 ± 10	-30 ± 1.1	Droplet	1.49 ± 0.13	-34.64 ± 0.5	-	0 ± 5	0.321	2	5806
citric acid, anhydrous	145 (°C), 0.1 L min-1	-13 ± 10	-70.3 ± 1.1	Droplet	1.48 ± 0.13	-34.81 ± 0.52	-	0 ± 5	0.167	1.73	11284
citric acid, anhydrous	140 (°C), 0.1 L min-1	-13 ± 10	-30.3 ± 1.1	Water Uptake	1 ± 0.02	-44.97 ± 0.12	-	0 ± 5	0.34	1.89	6942
citric acid, anhydrous	140 (°C), 0.1 L min-1	-13 ± 10	-30.1 ± 1.1	Water Uptake	1 ± 0.02	-40.08 ± 0.1	-	0 ± 5	0.305	1.94	6542
citric acid, anhydrous	140 (°C), 0.1 L min-1	-13 ± 10	-30.1 ± 1.1	Water Uptake	1 ± 0.01	-35.03 ± 0.09	-	0 ± 5	0.315	2.09	5724
citric acid, anhydrous	145 (°C), 0.1 L min-1	-13 ± 10	-70 ± 1.1	Water Uptake	1 ± 0.03	-35.05 ± 0.21	-	0 ± 5	0.167	1.74	11088
dodecyl-OS	Atomizer	74 ± 13	20.7 ± 1	Droplet	1.48 ± 0.11	-39.95 ± 0.46	0 ± 5	0 ± 5	0.222	1.32	15318

dodecyl-OS	Atomizer	74 ± 13	20.6 ± 1	Droplet	1.4 ± 0.12	-35.07 ± 0.5	0 ± 5	0 ± 5	0.224	1.31	15751
dodecyl-OS	Atomizer	74 ± 13	-	Water Uptake	1.32 ± 0.09	-44.95 ± 0.41	-	-	0.223	1.31	15675
dodecyl-OS	Atomizer	74 ± 13	20.7 ± 1	Water Uptake	1.32 ± 0.09	-39.99 ± 0.43	0 ± 5	0 ± 5	0.223	1.31	15532
dodecyl-OS	Atomizer	74 ± 13	20.6 ± 1	Water Uptake	1.24 ± 0.09	-34.97 ± 0.43	0 ± 5	0 ± 5	0.224	1.31	15611
ethyl-OS	Atomizer	-83 ± 34	22.6 ± 1	Droplet	1.5 ± 0.12	-40.15 ± 0.49	0 ± 5	1 ± 5	0.21	1.34	21038
ethyl-OS	Atomizer	-83 ± 34	22.3 ± 1	Droplet	1.39 ± 0.13	-34.86 ± 0.51	1 ± 5	3 ± 5	0.212	1.33	23591
ethyl-OS	Atomizer	-83 ± 34	-68.4 ± 1.1	Droplet	1.56 ± 0.13	-39.87 ± 0.5	-	1 ± 5	0.216	1.38	20572
ethyl-OS	Atomizer	-83 ± 34	-68.4 ± 1.1	Droplet	1.49 ± 0.13	-34.91 ± 0.52	-	1 ± 5	0.216	1.38	21284
ethyl-OS	Atomizer	-83 ± 34	22.9 ± 1	Water Uptake	1.38 ± 0.09	-45.22 ± 0.41	0 ± 5	0 ± 5	0.21	1.33	20332
ethyl-OS	Atomizer	-83 ± 34	22.6 ± 1	Water Uptake	1.32 ± 0.09	-40.14 ± 0.43	0 ± 5	1 ± 5	0.21	1.34	21327
ethyl-OS	Atomizer	-83 ± 34	22.3 ± 1	Water Uptake	1.25 ± 0.09	-34.97 ± 0.43	0 ± 5	3 ± 5	0.212	1.34	23201
ethyl-OS	Atomizer	-83 ± 34	-68.3 ± 1.1	Water Uptake	1.36 ± 0.09	-44.77 ± 0.41	-	1 ± 5	0.214	1.37	19318
ethyl-OS	Atomizer	-83 ± 34	-68.3 ± 1.1	Water Uptake	1.36 ± 0.1	-40.1 ± 0.44	-	1 ± 5	0.216	1.38	20387
ethyl-OS	Atomizer	-83 ± 34	-68.5 ± 1.1	Water Uptake	1.35 ± 0.11	-34.85 ± 0.5	-	1 ± 5	0.217	1.38	21048
methyl-OS	Atomizer	-83 ± 38	23 ± 1	Droplet	1.48 ± 0.11	-40.04 ± 0.47	6 ± 5	6 ± 5	0.223	1.28	26211
methyl-OS	Atomizer	-83 ± 38	-70.1 ± 1.1	Droplet	1.54 ± 0.15	-34.99 ± 0.56	-	1 ± 5	0.219	1.31	19084
methyl-OS	Atomizer	-83 ± 38	23.5 ± 1	Water Uptake	1.28 ± 0.08	-44.83 ± 0.38	3 ± 5	5 ± 5	0.22	1.28	21564
methyl-OS	Atomizer	-83 ± 38	23.1 ± 1	Water Uptake	1.2 ± 0.07	-39.9 ± 0.37	6 ± 5	7 ± 5	0.222	1.28	26355
methyl-OS	Atomizer	-83 ± 38	23.5 ± 1	Water Uptake	1.22 ± 0.08	-35.04 ± 0.41	7 ± 5	8 ± 5	0.221	1.3	32730
methyl-OS	Atomizer	-83 ± 38	-69.5 ± 1.1	Water Uptake	1.04 ± 0.05	-44.84 ± 0.28	-	0 ± 5	0.215	1.3	12461
methyl-OS	Atomizer	-83 ± 38	-69.9 ± 1.1	Water Uptake	1.44 ± 0.11	-39.97 ± 0.47	-	1 ± 5	0.218	1.31	19720
methyl-OS	Atomizer	-83 ± 38	-70.2 ± 1.1	Water Uptake	1.41 ± 0.11	-34.95 ± 0.46	-	1 ± 5	0.219	1.31	19216