S1 Methodology

S1.1 Particle classification thresholds

Table S1: Initial SPIN OPC parameters used to perform supervised SVM classification where x is the 1 second averaged particle property with respect to each class.

Class	δ_{SPIN}	$\log_{10}(\mathrm{I_{size}})$	$\mathrm{S_{liq}}$
Aerosol	$x < 0.16$	$x \leq 0.125$	-
Droplet	$0.16 \leq x \leq 0.35$	$0.35 \leq x$	$1 \leq x$
Ice	$0.4 \leq x$	$0.4 \leq x$	-
Water Uptake	$x < 0.35$	$0.125 \leq x \leq 0.4$	-

S1 Results

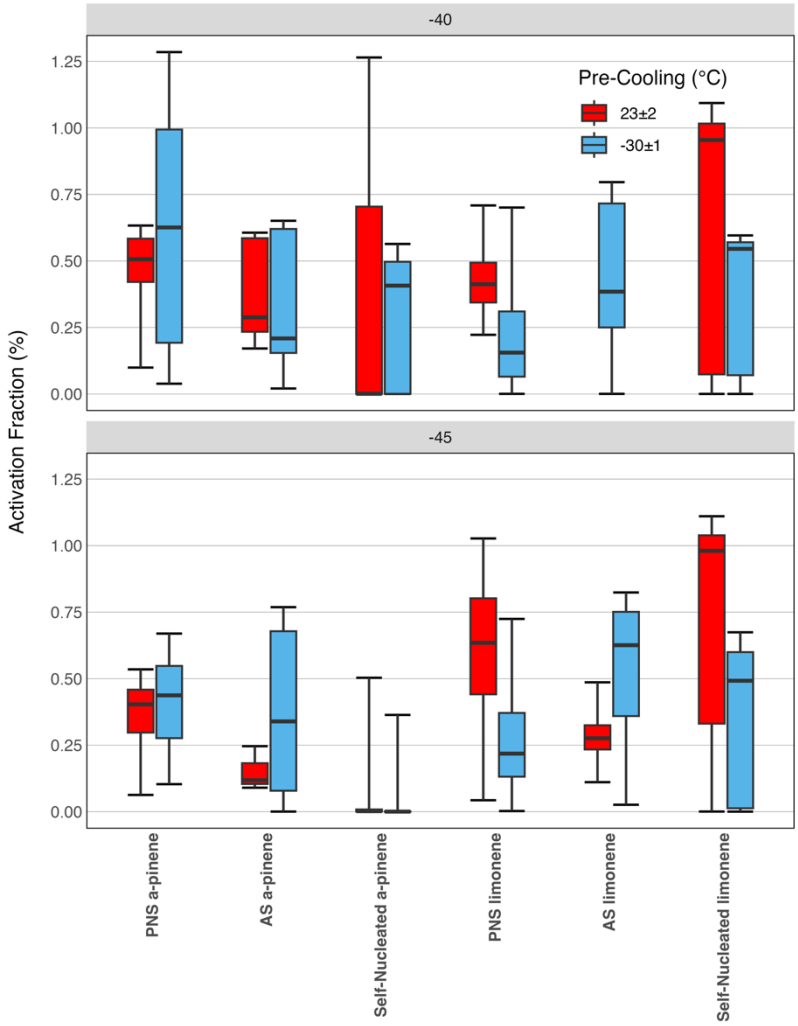

Figure S1 – Boxplot of heterogeneous AFs for each experiment without any smoothing or filtering as described in Sect. 2.6.3. Colors indicate the average pre-cooling temperature of the experiment. Panels are labeled by the average lamina temperature for the experiment. Whiskers are the min and maximum AF for each experiment without any outlier removal to accommodate the bias of AF values of 0 for much of each experiment until critical S_{ice} was reached.

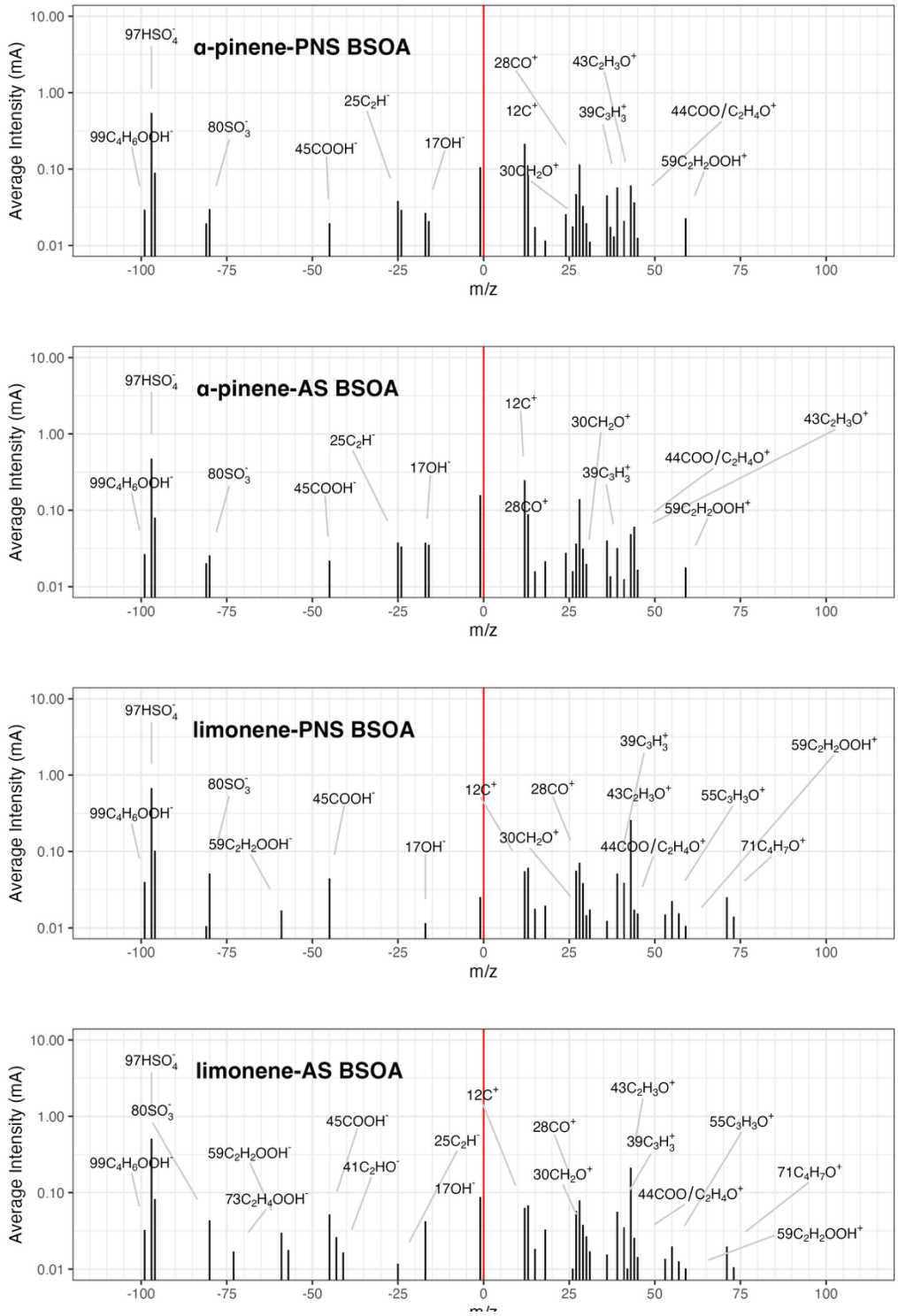

Figure S2 – Averaged mass spectra for each precursor-seed combination as labeled. Aerosol generation conditions correspond to Table 1. Spectra are plotted in log scale to accommodate the six orders of magnitude in dynamic range for PALMS-NG. Both positive

and negative ions are shown and separated by the red line centered at *m/z* = 0. Known PALMS specific ion fragments are labeled as cataloged or previously identified in upper tropospheric measurements (Froyd et al., 2009; Murphy et al., 2006). The following ion fragments are unique to limonene BSOA: -59 *m/z* ($C_2H_2OOH^-$), +55 *m/z* ($C_3H_3O^+$), and +71 *m/z* ($C_4H_7O^+$). For α-pinene BSOA, +12 *m/z* (C^+) was the strongest positive ion fragment followed by +28 *m/z* (CO^+), with comparable intensities for +39 ($C_3H_3^+$) and +43 ($C_2H_3O^+$) *m/z*. No unique ion fragments were identified for α-pinene. A primary difference between α-pinene and limonene BSOA was the positive ion fragment ratio of 39:43, with seed specific ratios of (AS 0.66, PNS 0.94) and (AS 0.27, PNS 0.20) respectively.

Table S2 – A summary of all thermodynamic onsets and corresponding experimental parameters. From left to right: seed particle composition, gas-phase precursor for BSOA generation, SPIN-OPC classification for particle type, number of observations for a given onset, 0.5% AF onset S_{ice}, onset temperature, geometric mean diameter of the primary particle mode, geometric standard deviation of the primary particle mode, CPC particle concentration larger than 100 nm, BSOA coating thickness when applicable, PCU temperature, PCU RH, PCU inlet RH, percentage of particles that were self-nucleated, and percentage of singlet to doublet particles.

Seed	Precursor	Class	Obs.	Onset S_{ice}	Onset temperature (°C)	D_{pg} (nm)	σ_g	Coating thickness (nm)	CPC (n cm^{-3})	PCU temperature (°C)	PCU RH (%)	PCU inlet RH (%)	Self-nucleated organic (%)	Singlet to doublet (%)
AS	Seed Only	Ice	3132	1.34 ± 0.08	-44.8 ± 0.4	288 ± 0.8	1.08	-	7356	24.3 ± 1.0	0 ± 5	0 ± 5	0	91 ± 0.02
AS	Seed Only	Ice	1846	1.36 ± 0.08	-39.8 ± 0.4	286 ± 0.6	1.08	-	7485	24.7 ± 1.0	0 ± 5	0 ± 5	0	90 ± 0.02
AS	Seed Only	Ice	455	1.53 ± 0.10	-34.7 ± 0.5	283 ± 0.5	1.09	-	8091	24.8 ± 1.0	0 ± 5	0 ± 5	0	91 ± 0.02
AS	Seed Only	Droplet	308	1.54 ± 0.10	-34.7 ± 0.5	283 ± 0.5	1.09	-	8091	24.8 ± 1.0	0 ± 5	0 ± 5	0	91 ± 0.02
AS	α-pinene	Ice	1080	1.49 ± 0.10	-45.2 ± 0.4	310 ± 5.7	1.1	10	12782	23.3 ± 1.0	0 ± 5	0 ± 5	0 ± 1	85 ± 0.03
AS	α-pinene	Ice	1290	1.44 ± 0.08	-44.9 ± 0.4	316 ± 0.5	1.1	19	19086	-29.8 ± 1.1	-	0 ± 5	4 ± 4	86 ± 0.03
AS	α-pinene	Ice	658	1.56 ± 0.10	-40.3 ± 0.4	303 ± 0.0	1.09	6	9533	22.1 ± 1.0	0 ± 5	0 ± 5	0 ± 0	89 ± 0.02
AS	α-pinene	Ice	1106	1.46 ± 0.09	-40.2 ± 0.4	317 ± 4.3	1.11	20	13241	22.0 ± 1.0	0 ± 5	0 ± 5	1 ± 1	84 ± 0.03
AS	α-pinene	Ice	1056	1.41 ± 0.09	-39.9 ± 0.4	318 ± 0.7	1.11	21	20885	-29.8 ± 1.1	-	0 ± 5	12 ± 6	85 ± 0.05
AS	α-pinene	Droplet	108	1.55 ± 0.10	-40.4 ± 0.5	303 ± 0.0	1.09	6	9533	22.1 ± 1.0	0 ± 5	0 ± 5	0 ± 0	89 ± 0.02
AS	α-pinene	Droplet	575	1.60 ± 0.11	-35.2 ± 0.5	313 ± 9.2	1.11	18	11518	21.5 ± 1.0	0 ± 5	0 ± 5	0 ± 2	84 ± 0.02
AS	α-pinene	Water Uptake	282	1.48 ± 0.09	-45.2 ± 0.4	310 ± 5.7	1.1	10	12782	23.3 ± 1.0	0 ± 5	0 ± 5	0 ± 1	85 ± 0.03
AS	α-pinene	Water Uptake	170	1.48 ± 0.09	-45.1 ± 0.4	316 ± 0.5	1.1	19	19086	-29.8 ± 1.1	-	0 ± 5	4 ± 4	86 ± 0.03
AS	α-pinene	Water Uptake	184	1.48 ± 0.09	-40.4 ± 0.4	303 ± 0.0	1.09	6	9533	22.1 ± 1.0	0 ± 5	0 ± 5	0 ± 0	89 ± 0.02
AS	α-pinene	Water Uptake	105	1.41 ± 0.08	-40.1 ± 0.4	317 ± 4.3	1.11	20	13241	22.0 ± 1.0	0 ± 5	0 ± 5	1 ± 1	84 ± 0.03

AS	α-pinene	Water Uptake	326	1.32 ± 0.07	-39.9 ± 0.4	318 ± 0.7	1.11	21	20885	-29.8 ± 1.1	-	0 ± 5	12 ± 6	85 ± 0.05
AS	limonene	Ice	1549	1.46 ± 0.09	-44.8 ± 0.4	314 ± 2.8	1.1	18	18532	24.3 ± 1.0	0 ± 5	0 ± 5	0 ± 1	89 ± 0.06
AS	limonene	Ice	2179	1.36 ± 0.08	-44.7 ± 0.4	318 ± 2.0	1.11	21	17217	-30.8 ± 1.1	-	0 ± 5	1 ± 2	87 ± 0.06
AS	limonene	Ice	1204	1.34 ± 0.07	-39.6 ± 0.4	325 ± 1.2	1.11	27	18356	-30.5 ± 1.1	-	0 ± 5	2 ± 2	87 ± 0.09
PNS	Seed Only	Ice	1528	1.47 ± 0.10	-44.4 ± 0.5	297 ± 0.4	1.08	-	5879	24.3 ± 1.0	0 ± 5	0 ± 5	0	89 ± 0.01
PNS	Seed Only	Ice	909	1.48 ± 0.10	-39.4 ± 0.5	298 ± 0.8	1.08	-	5466	23.6 ± 1.0	0 ± 5	0 ± 5	0	90 ± 0.01
PNS	Seed Only	Droplet	593	1.54 ± 0.11	-34.4 ± 0.5	299 ± 1.1	1.08	-	5364	23.6 ± 1.0	0 ± 5	1 ± 5	0	90 ± 0.01
PNS	α-pinene	Ice	1307	1.47 ± 0.09	-44.8 ± 0.4	328 ± 2.0	1.13	30	20808	20.5 ± 1.0	0 ± 5	0 ± 5	15 ± 7	86 ± 0.03
PNS	α-pinene	Ice	1313	1.47 ± 0.09	-45.0 ± 0.4	318 ± 0.7	1.11	21	17169	-29.9 ± 1.1	-	0 ± 5	13 ± 3	87 ± 0.02
PNS	α-pinene	Ice	1194	1.41 ± 0.08	-39.9 ± 0.4	331 ± 1.3	1.12	34	19689	23.3 ± 1.0	0 ± 5	0 ± 5	27 ± 2	86 ± 0.03
PNS	α-pinene	Ice	1259	1.39 ± 0.08	-39.7 ± 0.4	321 ± 6.7	1.11	22	24062	-29.7 ± 1.1	-	0 ± 5	44 ± 24	86 ± 0.03
PNS	α-pinene	Water Uptake	416	1.45 ± 0.09	-45.0 ± 0.4	318 ± 0.7	1.11	21	17169	-29.9 ± 1.1	-	0 ± 5	13 ± 3	87 ± 0.02
PNS	α-pinene	Water Uptake	261	1.37 ± 0.08	-40.1 ± 0.4	331 ± 1.3	1.12	34	19689	23.3 ± 1.0	0 ± 5	0 ± 5	27 ± 2	86 ± 0.03
PNS	α-pinene	Water Uptake	287	1.31 ± 0.07	-40.0 ± 0.4	321 ± 6.7	1.11	22	24062	-29.7 ± 1.1	-	0 ± 5	44 ± 24	86 ± 0.03
PNS	α-pinene	Water Uptake	322	1.26 ± 0.07	-34.8 ± 0.4	316 ± 7.7	1.11	18	28927	-29.4 ± 1.1	-	0 ± 5	27 ± 12	78 ± 0.03
PNS	limonene	Ice	1890	1.37 ± 0.08	-44.7 ± 0.4	311 ± 2.6	1.09	12	11687	-	-	-	0 ± 0	91 ± 0.05
PNS	limonene	Ice	1983	1.43 ± 0.08	-44.7 ± 0.4	325 ± 2.2	1.12	28	16098	-31.6 ± 1.1	-	0 ± 5	0 ± 2	89 ± 0.11
PNS	limonene	Ice	1207	1.40 ± 0.08	-39.7 ± 0.4	327 ± 2.3	1.12	30	15095	24.8 ± 1.0	0 ± 5	0 ± 5	1 ± 5	87 ± 0.05
PNS	limonene	Ice	1091	1.38 ± 0.08	-39.8 ± 0.4	322 ± 1.7	1.11	24	15616	-30.9 ± 1.1	-	0 ± 5	0 ± 1	87 ± 0.06
PNS	limonene	Droplet	223	1.61 ± 0.11	-34.8 ± 0.5	326 ± 5.4	1.12	29	16352	-30.8 ± 1.1	-	0 ± 5	0 ± 2	88 ± 0.07
-	α-pinene	Ice	1164	1.48 ± 0.09	-44.8 ± 0.4	85 ± 6.9	1.39	-	14817	24.0 ± 1.1	0 ± 5	0 ± 5	100	-
-	α-pinene	Ice	524	1.44 ± 0.09	-44.7 ± 0.4	89 ± 3.1	1.58	-	27444	-30.8 ± 1.1	-	0 ± 5	100	-
-	α-pinene	Ice	1013	1.40 ± 0.08	-40.0 ± 0.4	88 ± 7.7	1.25	-	12620	22.4 ± 1.1	0 ± 5	0 ± 5	100	-

-	α-pinene	Ice	1020	1.39 ± 0.09	-40.1 ± 0.4	88 ± 6.1	1.54	-	25480	-30.3 ± 1.1	-	0 ± 5	100	-
-	α-pinene	Droplet	650	1.48 ± 0.10	-34.8 ± 0.5	90 ± 8.4	1.27	-	15336	22.0 ± 1.1	0 ± 5	0 ± 5	100	-
-	α-pinene	Water Uptake	495	1.40 ± 0.08	-44.8 ± 0.4	85 ± 6.9	1.39	-	14817	24.0 ± 1.1	0 ± 5	0 ± 5	100	-
-	α-pinene	Water Uptake	790	1.26 ± 0.07	-39.8 ± 0.4	88 ± 7.7	1.25	-	12620	22.4 ± 1.1	0 ± 5	0 ± 5	100	-
-	α-pinene	Water Uptake	128	1.41 ± 0.08	-39.9 ± 0.4	88 ± 6.1	1.54	-	25480	-30.3 ± 1.1	-	0 ± 5	100	-
-	α-pinene	Water Uptake	517	1.26 ± 0.07	-34.8 ± 0.4	90 ± 8.4	1.27	-	15336	22.0 ± 1.1	0 ± 5	0 ± 5	100	-
-	limonene	Ice	2564	1.28 ± 0.07	-44.8 ± 0.3	72 ± 4.3	1.27	-	14800	24.0 ± 1.0	0 ± 5	0 ± 5	100	-
-	limonene	Ice	2196	1.31 ± 0.07	-44.7 ± 0.4	92 ± 4.4	1.26	-	24242	-30.2 ± 1.1	-	0 ± 5	100	-
-	limonene	Ice	1593	1.29 ± 0.07	-39.7 ± 0.4	77 ± 9.5	1.24	-	16995	23.8 ± 1.0	0 ± 5	0 ± 5	100	-
-	limonene	Ice	1500	1.27 ± 0.07	-39.8 ± 0.3	99 ± 0.0	1.17	-	26227	-30.3 ± 1.1	-	0 ± 5	100	-

Table S3 – Chemical formulas of organosulfate compounds detected in coating experiments using VIA-Vocus-CIMS

(amu)	α-Pinene–AS BSOA	α-Pinene–PNS BSOA	Limonene–AS BSOA	Limonene–PNS BSOA
388	–	–	–	$C_{17}H_{24}O_6S_2$
374	–	–	–	$C_{12}H_{22}O_9S_2$
332	–	–	–	$C_{15}H_{24}O_4S_2$
320	–	–	–	$C_{11}H_{12}O_9S$
312	–	–	$C_{15}H_{20}O_5S$	–
298	$C_{13}H_{14}O_6S$	–	–	–
265	–	$C_8H_9O_8S$	–	–
246	–	–	–	$C_9H_{10}O_6S$
244	–	–	$C_8H_{20}O_6S$	–
240	$C_8H_{16}O_6S$	$C_8H_{16}O_6S$	$C_8H_{16}O_6S$	–
238	–	–	–	$C_9H_{18}O_5S$
234	–	–	$C_{10}H_{18}O_4S$; $C_{11}H_{22}O_3S$	–
226	–	–	–	$C_8H_{18}O_5S$
196	–	–	$C_{10}H_{12}O_2S$	$C_6H_{12}O_5S$
166	–	–	–	$C_6H_{14}O_3S$
162	–	–	–	$C_7H_{14}O_2S$
148	–	–	–	$C_6H_{12}O_2S$

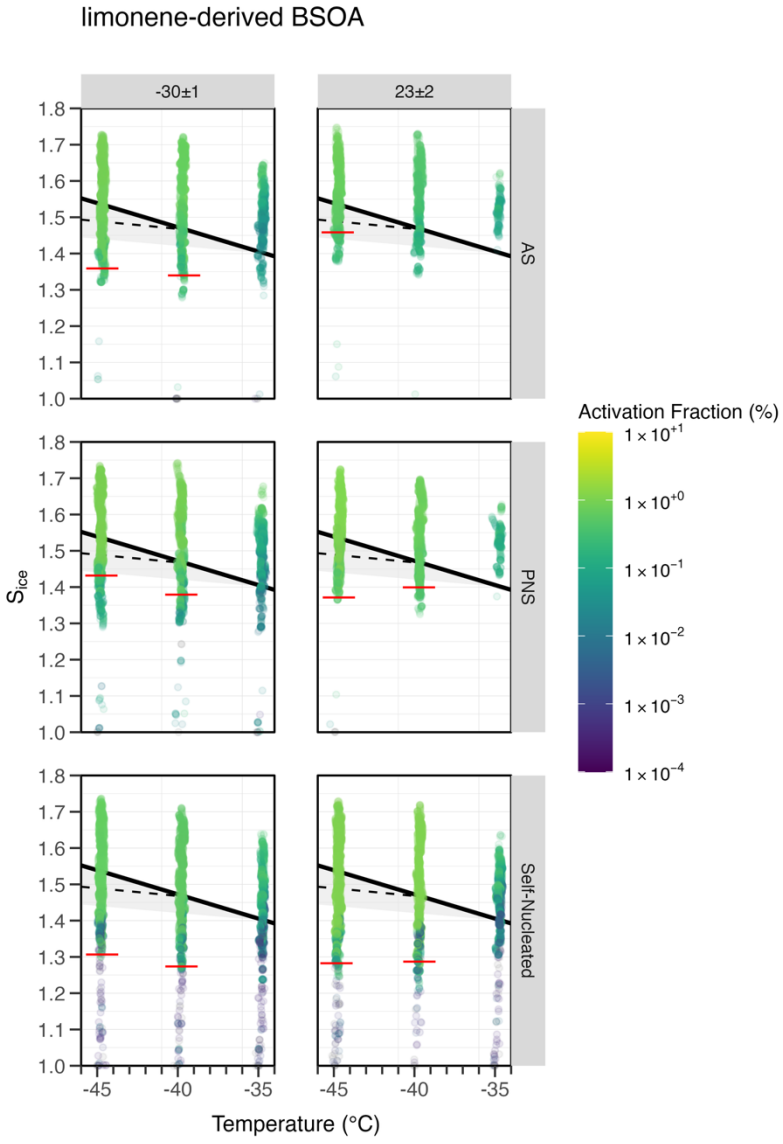

Figure S3 – Activated fractions of limonene-derived BSOA as a function of temperature and S_{ice}. These include all OPC classes. Panel labels indicate pre-cooling temperature (top) or whether a seed was present (right). In each panel, solid black line represents water saturation, dashed black line is the homogeneous freezing threshold of 100 nm diameter solution droplets as defined by (Koop et al., 2000), with uncertainty in water activity (a_w) of ±2.5% represented by the gray shaded region (Koop, 2004). Red horizontal lines are the 0.5% ice onsets reported.

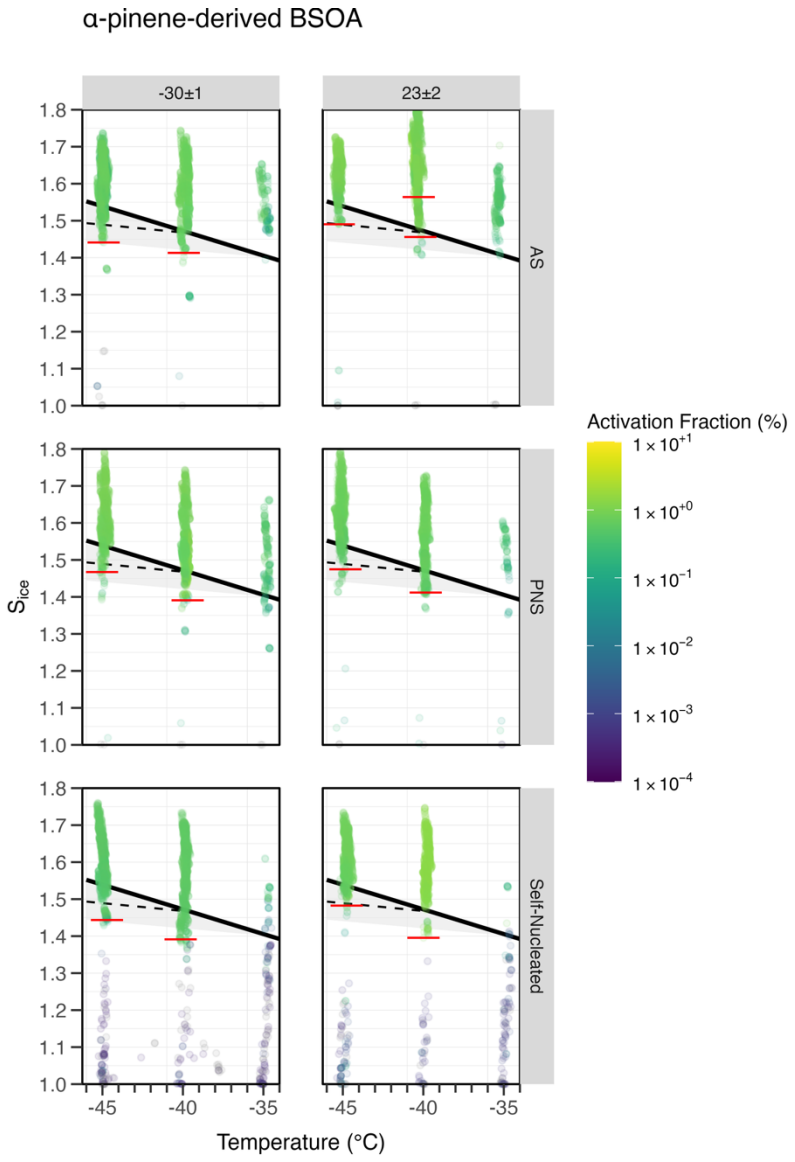

Figure S4 – Activated fractions of α-pinene-derived BSOA as a function of temperature and S_{ice}. Panel structure is identical to Figure S3.

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S1 References

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