Supplemental information

Diverse mixing state and ice nucleation properties of aerosol particles over the Western Pacific and the Southern Ocean

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Table S1. Information for the particle samples collected aboard the R/V *Araon* including the collecting date, time, and location; collection time (sampling duration); the mean values (average) of air temperature, relative humidity (RH), air pressure, black carbon (BC) concentration and relative wind speed; and the range of relative wind direction. The number of particles examined by CCSEM/EDX were listed and the samples used for ice nucleation experiments were marked.

Region	Samples ID	Collecting start time (UTC)	Collecting end time (UTC)	Collection Time (mins)	Collecting start location (latitude, longitude)	Collecting end location (latitude, longitude)	Average air temperature (°C)	Average RH (%)	Average Pressure (hPa)	Average relative wind speed (m/s)	Relative wind direction range (°)	Average BC concentration (ng/m3)	Number of particles examined by CCSEM	Ice nucleation experiment
WP-I	S1	10/31 19:19	10/31 19:29	10	34.96° N, 125.61° E	34.93° N, 125.61° E	18.6±0.0	85.5±0.2	1017.6±0.0	07.1±0.3	68 - 75	1963.3±42.0	1270	√
	S2	11/01 20:57	11/01 21:07	10	30.29° N, 128.67° E	30.26° N, 128.69° E	22.4±0.1	56.4±1.6	1014.1±0.0	09.8±0.3	357 - 9	217.0±127.5	910	
	S3	11/02 21:52	11/02 22:02	10	25.59° N, 131.56° E	25.57° N, 131.57° E	26.5±0.0	80.3±0.5	1009.8±0.1	14.1±0.1	337 - 342	189.8±174.2	1002	
	S4	11/03 21:06	11/03 21:16	10	21.90° N, 134.75° E	21.87° N, 134.77° E	27.7±0.0	80.2±0.2	1007.9±0.0	12.4±0.2	355 - 358	9.2±9.8	914	√
	S5	11/03 21:33	11/04 01:33	240	21.83° N, 134.81° E	21.12° N, 135.28° E	28.0±0.2	77.0±2.6	1007.1±0.2	10.0±0.4	337 - 4	25.9±15.4	1191	
	S6	11/04 23:14	11/05 03:14	240	17.23° N, 137.58° E	16.40° N, 138.29° E	28.6±0.4	80.2±2.2	1001.3±0.8	11.2±1.6	305 - 325	35.9±46.4	928	
	S7	11/05 23:48	11/06 03:48	240	13.03° N, 141.09° E	12.36° N, 141.56° E	28.8±0.2	77.9±1.5	1000.4±0.8	05.9±0.5	346 - 22	22.2±30.3	908	
WP-II	S8	11/08 02:01	11/08 06:07	246	04.26° N, 147.22° E	03.45° N, 147.54° E	29.3±0.1	76.5±2.0	1003.4±0.5	13.4±1.5	67 - 80	107.4±70.6	1247	
	S9	11/08 08:00	11/08 12:00	240	03.25° N, 148.09° E	02.43° N, 148.42° E	29.2±0.1	80.4±1.9	1005.2±0.7	10.7±0.7	11 - 67	50.3±31.5	1064	
	S10	11/08 13:00	11/08 17:00	240	02.32° N, 148.50° E	01.51° N, 149.22° E	28.8±0.1	81.5±1.6	1004.9±0.6	07.2±1.7	347 - 20	17.6±13.3	986	√
	S11	11/10 05:31	11/10 10:04	273	04.45° S, 154.02° E	05.46° S, 154.12° E	28.1±0.3	79.4±2.9	1005.0±0.8	08.1±1.6	326 - 10	26.3±29.8	1075	√
	S12	11/11 05:04	11/11 09:04	240	09.12° S, 155.36° E	10.02° S, 156.02° E	28.2±0.5	72.8±5.6	1005.1±0.8	16.9±0.7	359 - 15	60.9±47.1	1409	√
	S13	11/12 05:01	11/12 09:01	240	13.30° S, 157.55° E	14.11° S, 158.17° E	27.3±0.1	68.1±1.5	1007.1±0.9	16.4±0.4	353 - 5	161.5±50.1	979	
	S14	11/13 03:47	11/13 07:49	242	17.19° S, 160.01° E	18.02° S, 160.25° E	25.7±0.1	64.3±1.4	1007.8±0.4	13.6±0.3	352 - 3	674.7±59.3	966	√
	S15	11/14 03:38	11/14 07:38	240	20.59° S, 162.04° E	21.47° S, 162.21° E	23.5±0.1	62.6±1.2	1009.5±0.4	12.2±0.4	1 - 22	356.8±122.9	1483	
	S16	11/14 20:39	11/15 00:39	240	24.29° S, 163.03° E	25.20° S, 163.12° E	22.1±0.1	74.1±0.8	1013.0±0.2	07.7±1.2	331 - 353	31.1±20.0	1027	
	S17	11/17 19:31	11/17 23:31	240	37.13° S, 169.01° E	37.52° S, 169.39° E	14.7±0.7	78.7±4.7	1008.2±0.2	09.9±2.9	69 - 112	30.4±31.5	730	
SO	S18	11/28 19:15	11/28 23:15	240	61.33° S, 174.10° W	62.17° S, 173.58° W	1.7±0.5	97.9±0.3	982.9±0.2	11.8±2.5	26 - 344	52.1±81.2	666	
	S19	11/29 07:28	11/29 11:28	240	63.52° S, 173.16° W	64.38° S, 172.58° W	-0.9±0.2	93.8±0.7	966.8±7.3	17.9±1.4	325 - 341	65.6±106.6	794	
	S20	11/29 19:43	11/29 23:43	240	66.14° S, 172.17° W	66.55° S, 171.59° W	-3.5±0.1	88.1±1.6	973.5±5.7	13.3±1.4	323 - 11	4.4±5.6	1011	√
	S21	12/01 04:26	12/01 10:30	364	71.18° S, 171.49° W	72.04° S, 172.07° W	-4.6±0.4	80.9±6.9	992.0±1.3	07.8±0.7	3 - 356	36.3±6.0	992	
	S22	12/02 05:46	12/02 11:47	361	73.26° S, 176.20° W	74.21° S, 178.58° E	-4.2±1.1	81.9±14.4	990.7±14.0	09.3±5.2	19 - 61	17.0±88.4	924	
	S23	12/02 16:00	12/02 22:02	362	74.07° S, 177.09° W	75.12° S, 173.34° E	-2.2±0.3	75.8±2.8	984.1±1.0	13.2±2.4	349 - 49	3.5±20.2	978	
	S24	12/03 01:55	12/03 07:53	358	75.08° S, 171.07° E	74.55° S, 166.26° E	-2.3±0.2	69.1±0.2	963.9±0.7	15.6±1.7	8 - 67	59.2±28.3	638	
	S25	12/08 21:18	12/09 03:21	363	74.43° S, 164.49° E	74.59° S, 169.33° E	-2.7±0.2	79.2±2.1	993.4±0.3	07.5±1.1	7 - 359	14.9±20.4	861	
	S26	12/09 08:26	12/09 14:26	360	74.57° S, 173.14° E	74.18° S, 177.20° E	-3.2±0.7	79.6±3.4	990.4±0.7	11.0±2.6	305 - 331	14.3±36.9	944	
	S27	12/09 19:48	12/10 01:49	361	73.42° S, 179.06° W	73.07° S, 175.26° W	-4.3±0.4	85.7±3.9	992.7±0.5	09.0±2.4	344 - 36	25.4±54.5	928	√
	S28	12/10 19:56	12/11 01:58	362	70.39° S, 171.00° W	69.34° S, 171.11° W	-4.1±0.1	68.6±2.9	993.9±1.0	12.3±1.8	19 - 108	38.7±21.5	922	
	S29	12/11 20:03	12/12 02:03	360	66.59° S, 178.24° W	66.24° S, 179.52° E	-0.3±0.4	98.6±6.0	980.2±9.4	11.8±1.9	0 - 359	36.3±62.4	814	

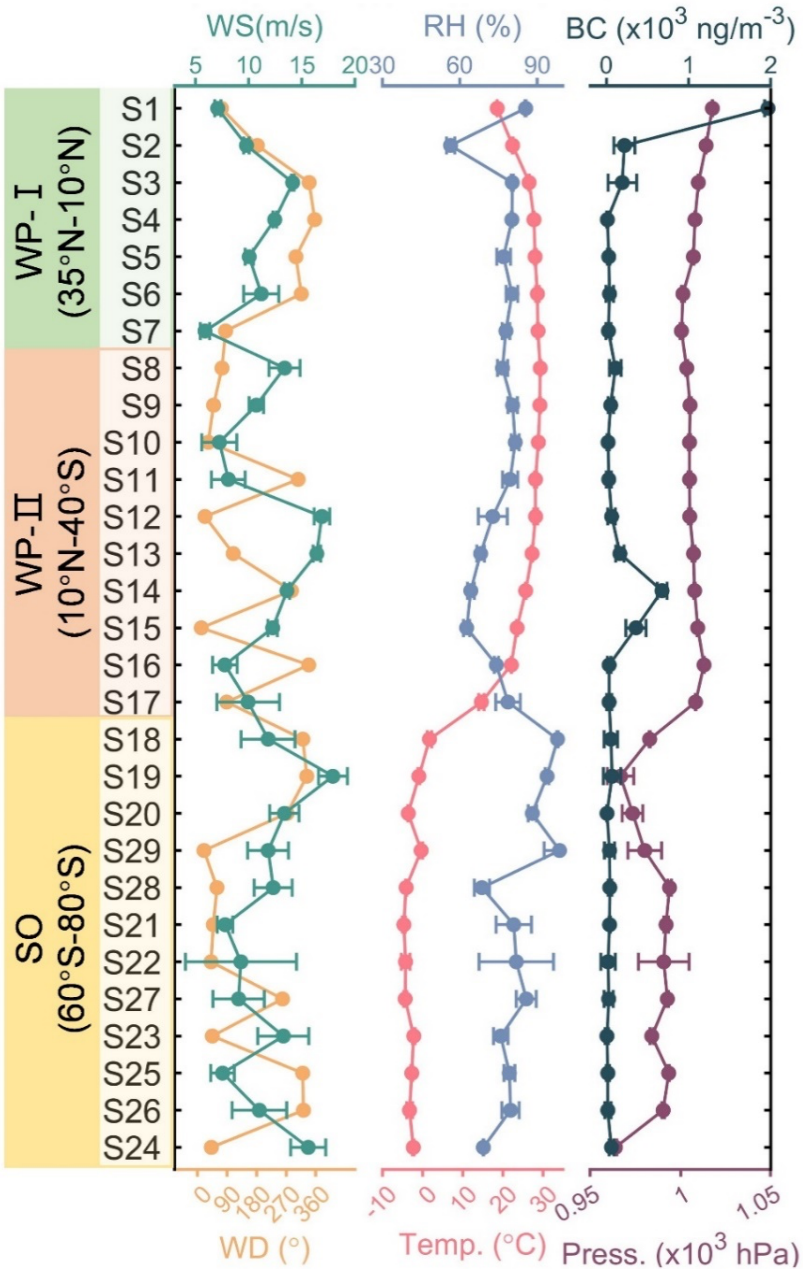

Figure S1. Meteorological data and BC concentrations for all samples. Mean values with one standard deviation for the relative wind speed (WS), relative wind direction (WD), air temperature (Temp.), RH, pressure (Press.), and BC concentration. Samples of S1 – S7, S8 – S17, and S18 – S24 were located in 35°N – 10°N, 10°N – 40°S, and 60°S – 80°S regions, respectively.

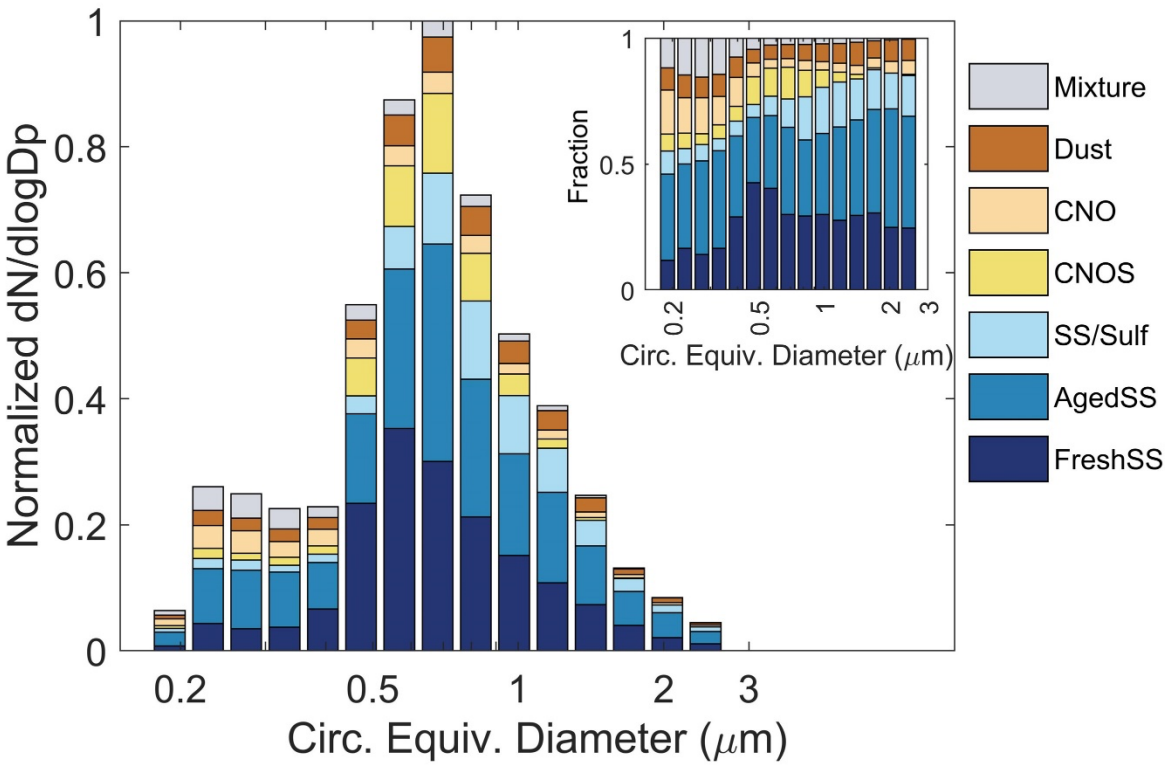

Figure S2. The size distribution of all types of particles for all samples. The inset plot shows the fractions of all types of particles.

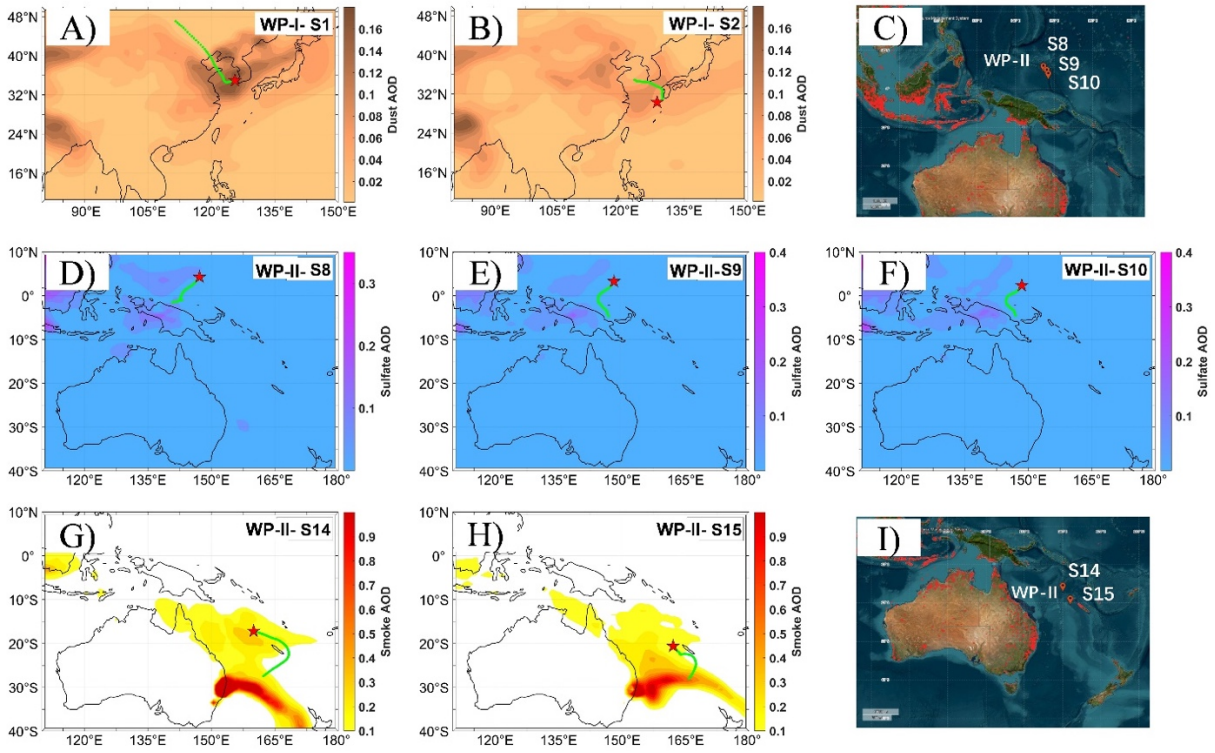

Figure S3. Aerosol optical depth (AOD) of dust (A and B), sulfates (D – F), and smoke (G and H) produced by Navy Aerosol Analysis and Prediction System reanalysis (NAAPS-RA, at 550nm) overlaid with HYSPLIT 72h backward trajectories (green line). (C) and (I) are the accumulated fire spots from Nov. 2nd to Nov. 8th, 2019 and from Nov. 8th to 14th, 2019, respectively, from Fire Information for Resource Management System (FIRMS). Sample labels are shown on the top right of each panel.

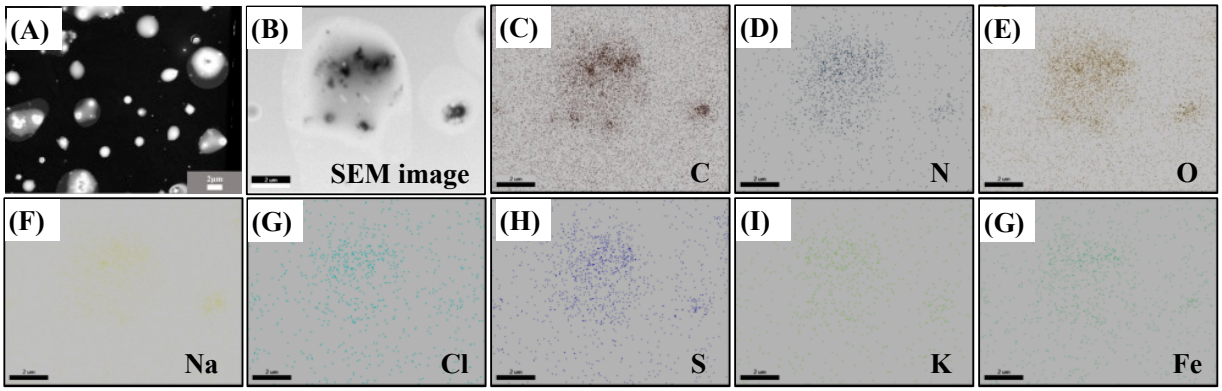

Figure S4. SEM images (A and B) and EDX elemental maps (C – G) indicate the characteristic of BBA particles from S14 sample. (A) SEM image of a larger field was captured on the darkfield mode using scanning transmission electron microscopy (STEM) detector. (B) SEM image of a smaller field for chemical mapping (C – G) by collecting EDX signals. Intense carbon signals within the particle (B) indicated high density of carbon inclusions which likely are aged soot. The scale bar for all images is 2 μm.

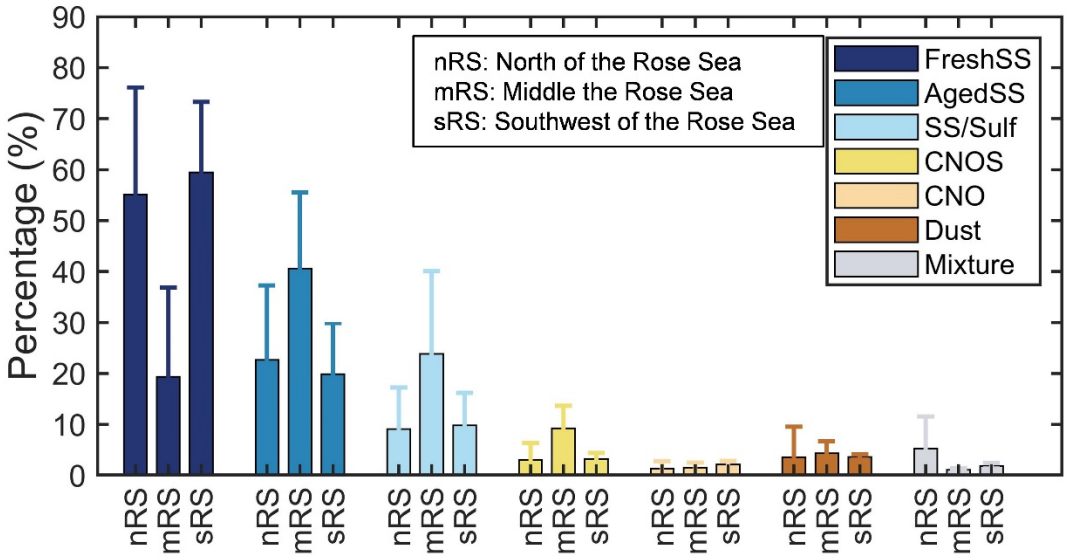

Figure S5. The average number percentages of all particle classes for samples that were collected in the middle of Rose Sea (S28, S21, S22, S27, and S23), the north part of Rose Sea (S18, S19, S20, and S29), and the southwest part of Rose Sea (S25, S26, and S24).

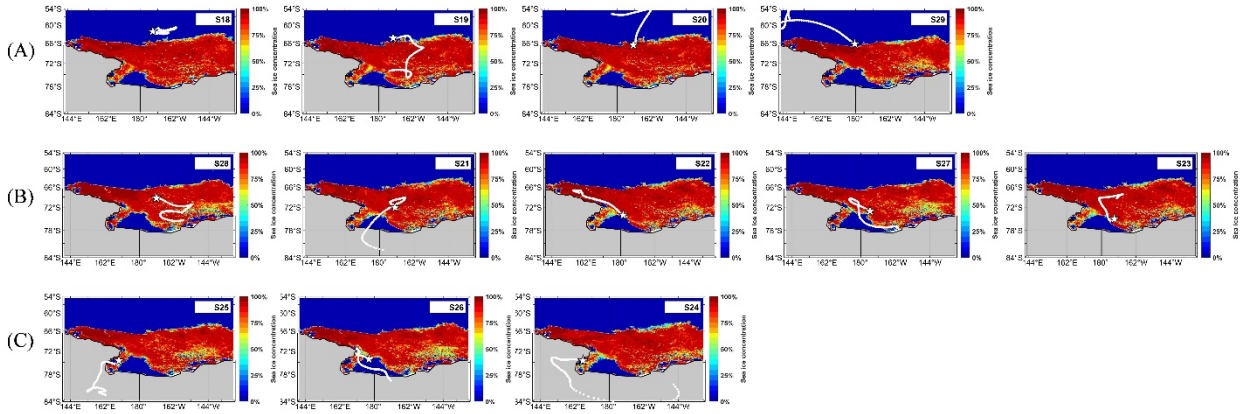

Figure S6 Sea ice concentration over the third sample region (SO) (data retrieved from https://seaice.uni-bremen.de/sea-ice-concentration/amsre-amsr2/) overlaid with HYSPLIT 120 h backward trajectories (white lines). The stars represent the sampling locations. The sample number is showed in the upper right corner of each panel. (A) north of the Rose Sea (S18, S19, S20, and S29), (B) middle of the Rose Sea (S28, S21, S22, S27, and S23), (C) southwest of the Rose Sea (S28, S21, S22, S27, and S23).

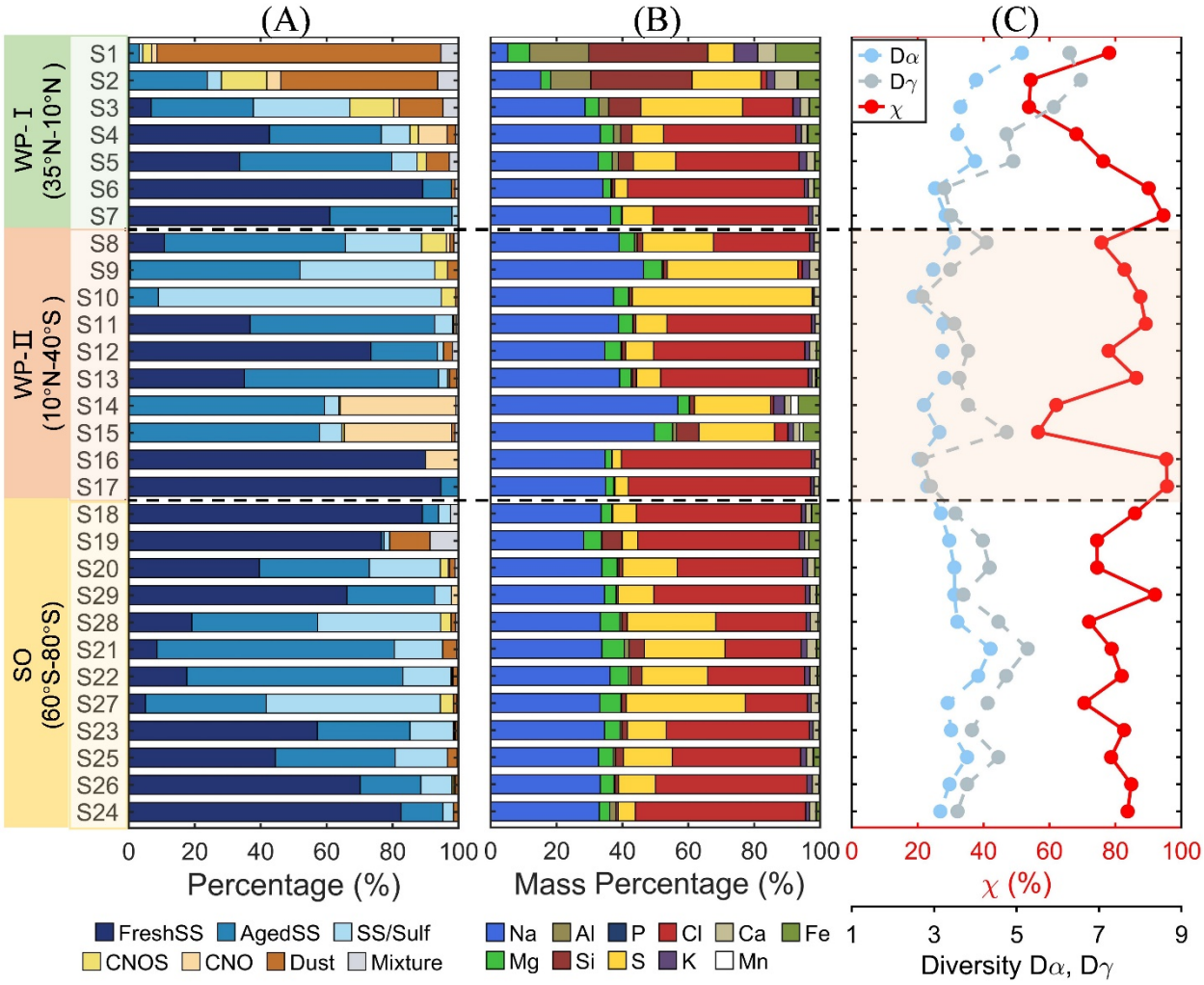

Figure S7. The number percentages of different particle classes (A), mass percentages of elements (B), and mixing state (C) of super-micron particles (diameter > 1 μm). Light blue, gray, and red points represent the average particle elemental diversity (D_α), bulk population elemental diversity (D_γ), and mixing state index (χ), respectively.

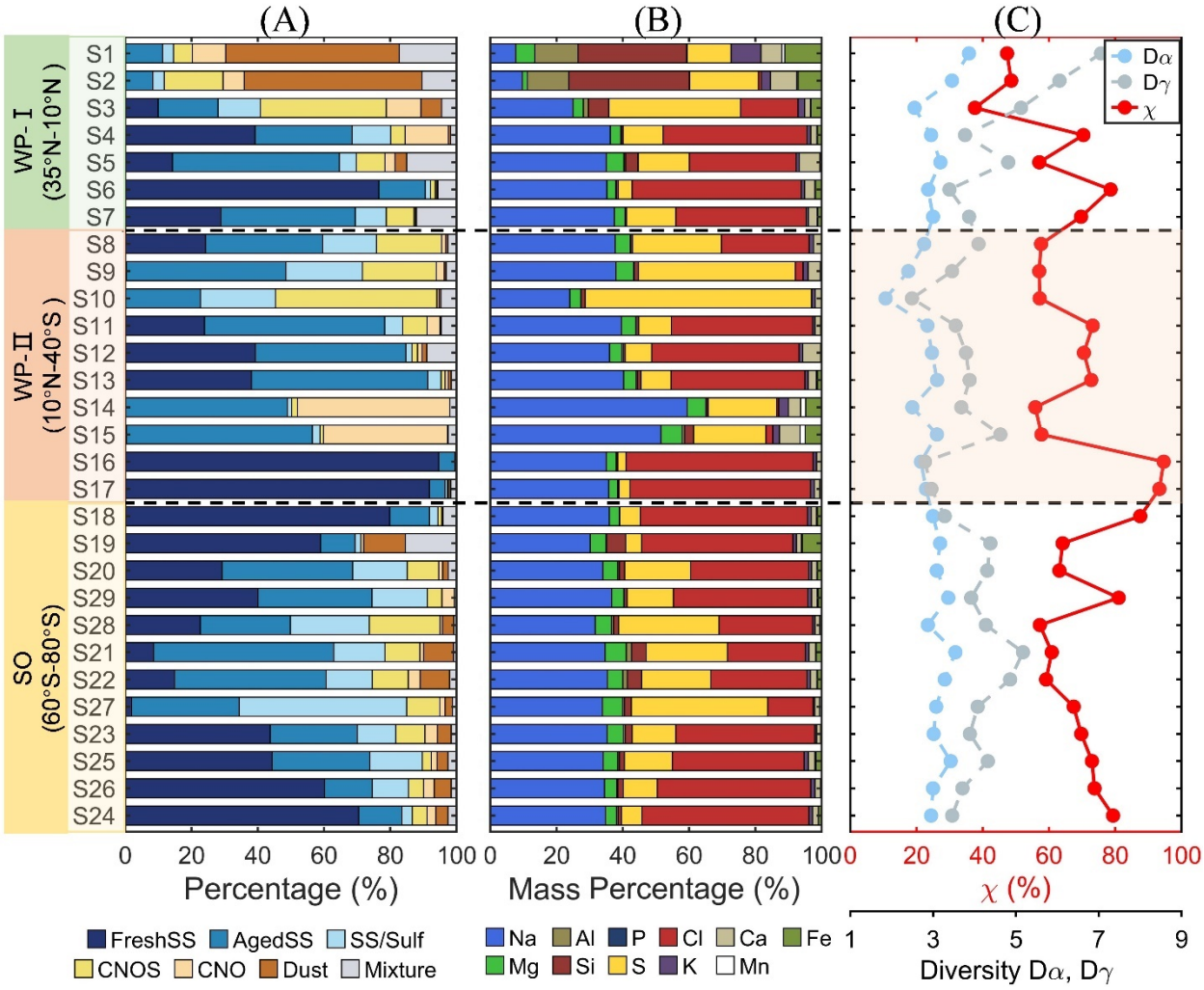

Figure S8. The number percentages of different particle classes (A), mass percentages of elements (B), and mixing state (C) of sub-micron particles (diameter between 0.2 and 1 μm). The light blue, gray, and red points represent D_{α}, D_{γ}, and χ, respectively.

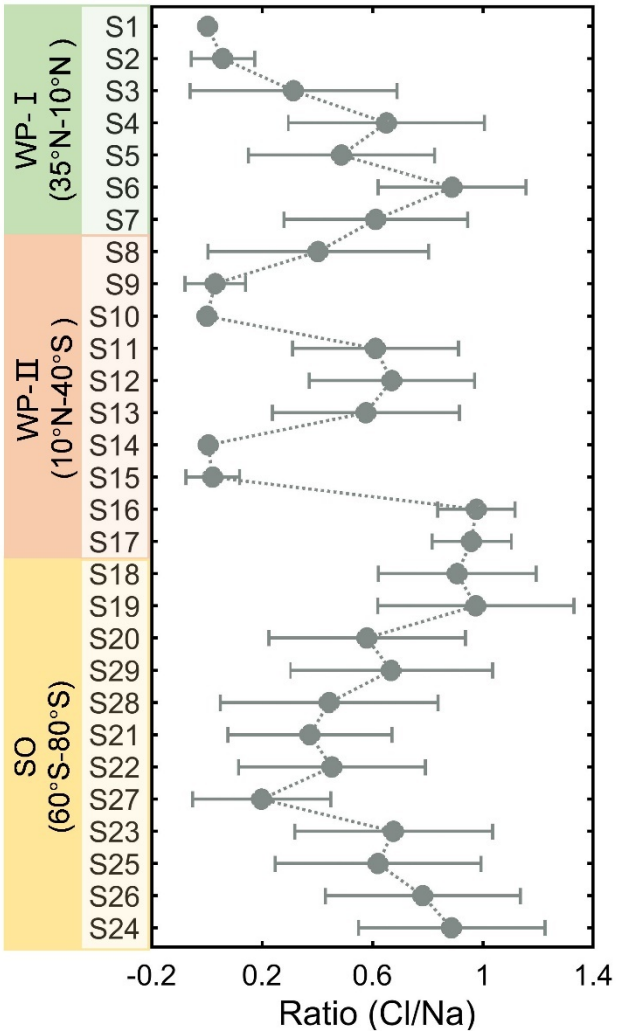

Figure S9. The ratio of chlorine to sodium in FreshSS, AgedSS and SS/Sulf particles. The data are mean value with one standard deviation.

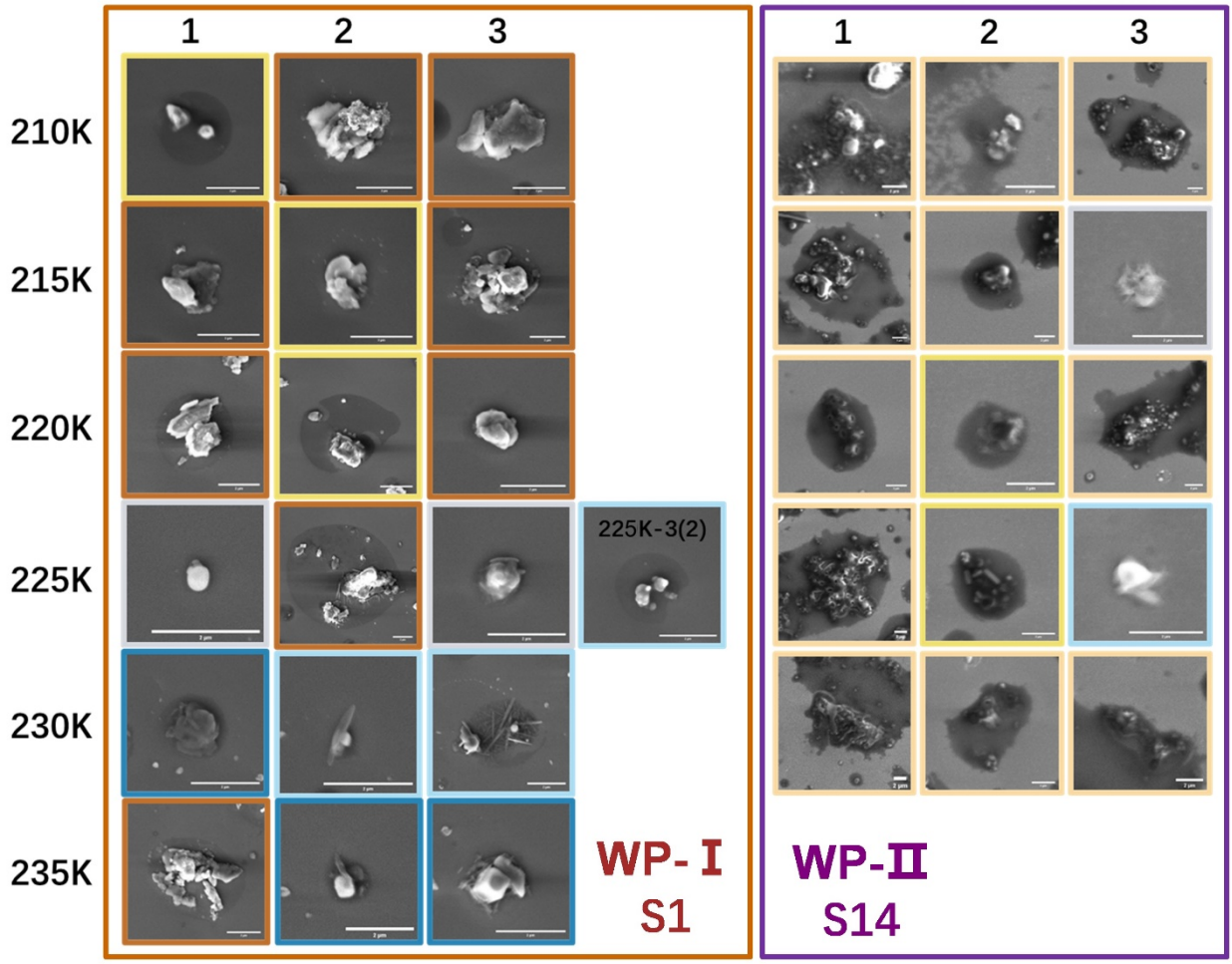

Figure S10. SEM images of INPs identified at different freezing temperatures for the Dust dominated samples (S1) and BBA influenced samples (S14). The frame color of images represents the types of INPs and use the same color codes in Figure 2. Blue, light blue, yellow, light orange, brown, and gray represent the FreshSS, AgedSS, SS/Sulf, CNOS, CNO, Dust, and Mixture particle class, respectively. The number on top represents the sequence of ice nucleation experiments. The number on the SEM image indicates the temperature and number of INPs at the onset conditions. For example, the label "225K-3(2)" means this is the second INPs nucleated ice at the same RH_{ice} for the 3rd ice nucleation experiment at 225 K. The scale bar for all images is 2μm.

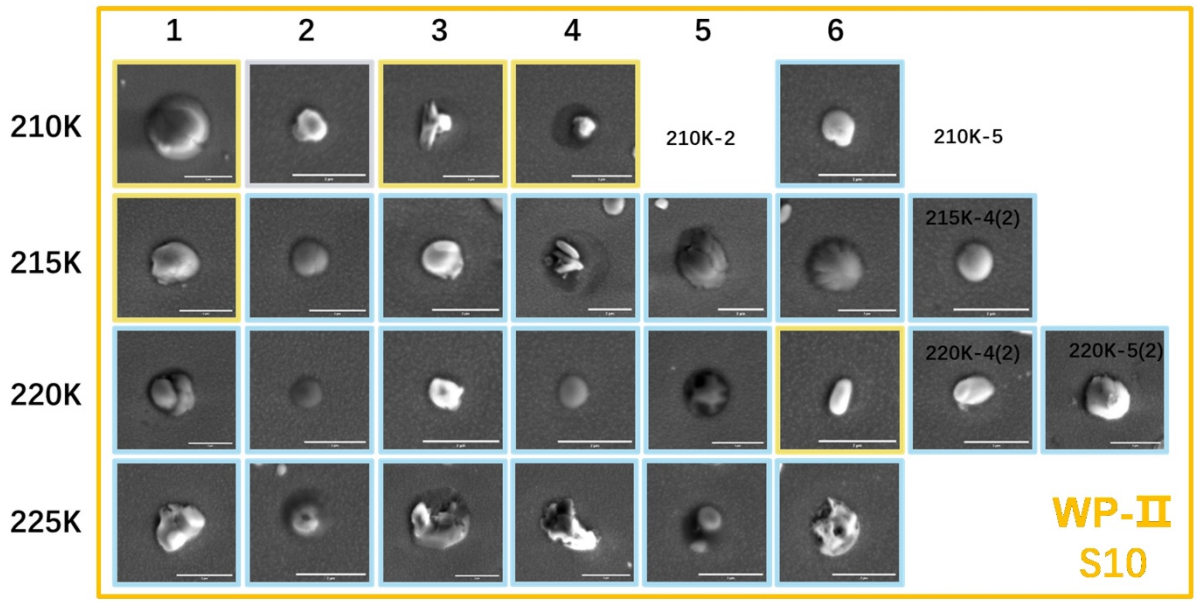

Figure S11. SEM images of INPs identified at different freezing temperatures for the CNOS and SS/Sulf dominated samples (S10). The label without SEM image indicates the same INP nucleated ice at a different ice nucleation experiment at the same temperature. For example, the lable "210K-2" means at the fifth ice nucleation experiment the INP is the same as the second run at 210 K. Other descriptions of the labels are the same as in Figure S9. The scale bar for all images is 2 μm.

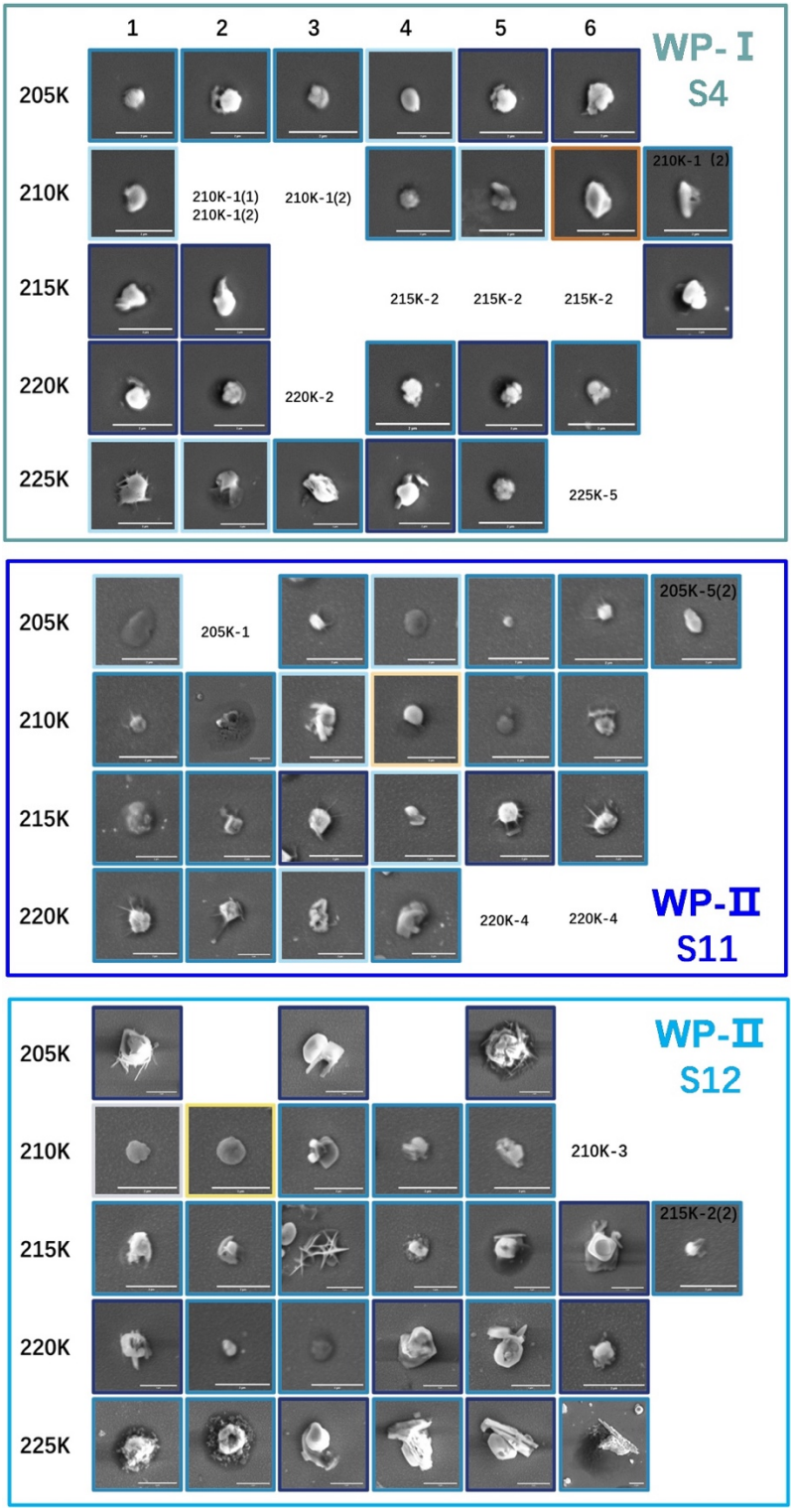

Figure S12. SEM images of INPs identified at different freezing temperatures for the FreshSS and AgedSS dominated samples (S4, S11, and S12). Other descriptions of the labels are the same as in Figure S9 and Figure S10. The scale bar for all images is 2 μm.

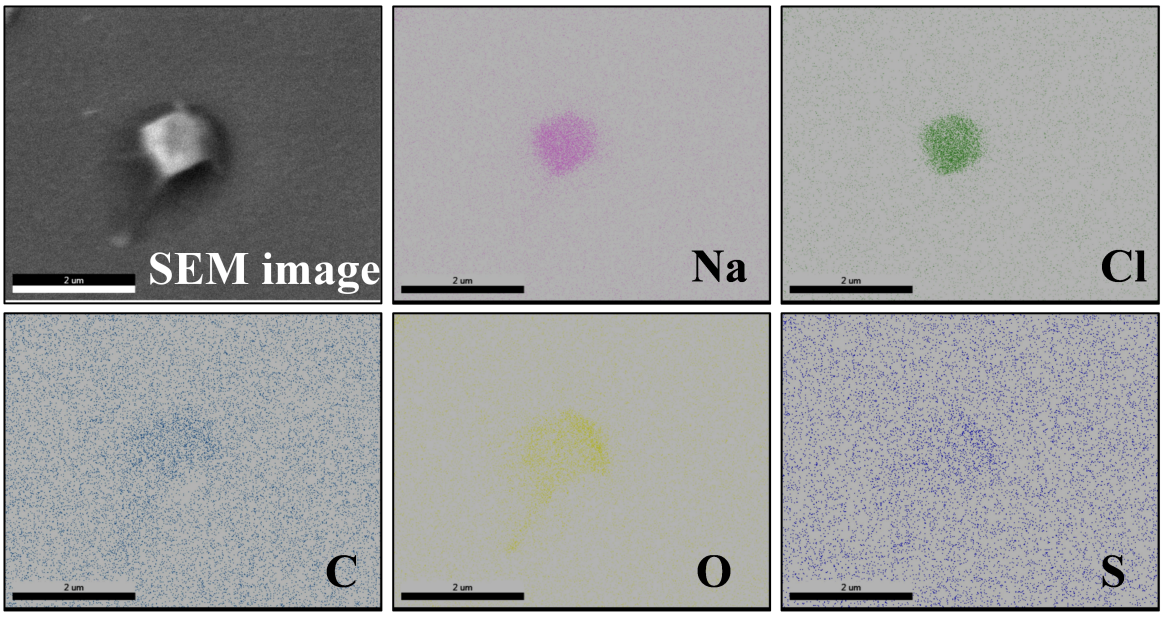

Figure S13. SEM image and EDX elemental maps for a typical AgedSS particle from S11 samples. The scale bar is 2 µm for all images.

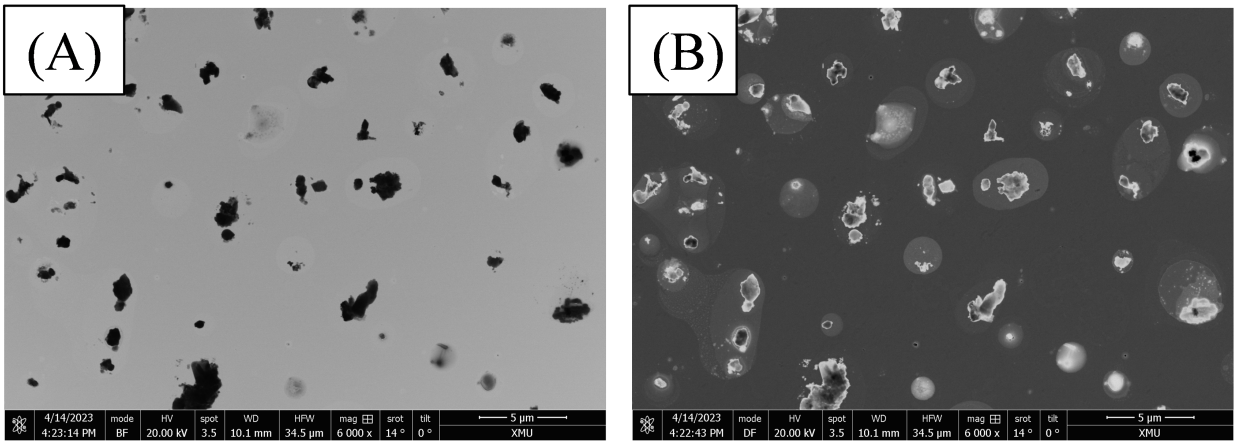

Figure S14. The SEM images of particles from S1 sample using scanning transmission electron microscopy (STEM) detector. (A) on the bright field mode, (B) on the dark field mode. The scale bar is 5 µm.

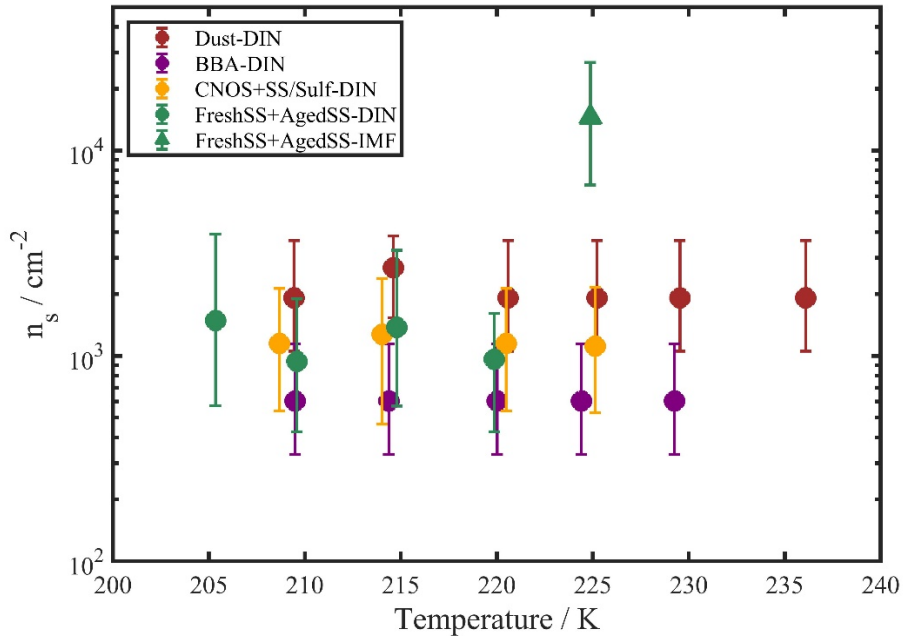

Figure S15 Median n_s with 25th and 75th percentiles for all types of samples. Circles represent DIN and triangles represent IMF. Brown, purple, orange, and green symbols represent Dust, BBA, CNOS and SS/Sulf, FreshSS and AgedSS dominated samples, respectively.

Table S2. Enrichment factors for different types of INPs. The lower and upper limits of EFs were calculated by Poisson distribution at 95% confidence level.

EF	FrehsSS	AgedSS	SS/Sulf	CNOS	CNO	Dust	Mixture	Aging process
WP-I-S1	\	$0.89^{2.29}_{0.24}$	$2.53^{6.53}_{0.69}$	$1.09^{2.83}_{0.30}$	$0.00^{1.64}_{0.00}$	$1.48^{2.68}_{0.74}$	$0.45^{1.41}_{0.08}$	$1.31^{2.59}_{0.57}$
WP-II-S14	\	$0.00^{1.03}_{0.00}$	$2.75^{13.05}_{0.14}$	$1.57^{4.95}_{0.28}$	$1.34^{2.23}_{0.75}$	$0.00^{16.48}_{0.00}$	$0.48^{2.27}_{0.02}$	$0.31^{1.45}_{0.02}$
WP-II-S10	\	$0.00^{1.59}_{0.00}$	$1.52^{2.21}_{1.01}$	$1.03^{2.17}_{0.41}$	\	$0.00^{8.35}_{0.00}$	$0.17^{0.83}_{0.01}$	$1.33^{1.93}_{0.88}$
WP-I-S4	$0.95^{1.66}_{0.50}$	$0.91^{1.60}_{0.48}$	$2.72^{5.72}_{1.07}$	$0.00^{8.15}_{0.00}$	$0.00^{32.58}_{0.00}$	$2.18^{10.32}_{0.11}$	$0.00^{1.55}_{0.00}$	$1.20^{1.87}_{0.72}$
WP-II-S11	$0.52^{1.63}_{0.09}$	$1.06^{1.66}_{0.64}$	$2.17^{4.57}_{0.86}$	$0.00^{8.14}_{0.00}$	$2.72^{12.88}_{0.14}$	$0.00^{32.55}_{0.00}$	$0.00^{1.63}_{0.00}$	$1.23^{1.80}_{0.80}$
WP-II-S12	$0.94^{1.65}_{0.49}$	$1.14^{1.75}_{0.70}$	$0.00^{2.75}_{0.00}$	$5.50^{26.09}_{0.28}$	$0.00^{32.96}_{0.00}$	$0.00^{16.48}_{0.00}$	$0.58^{2.75}_{0.03}$	$1.05^{1.62}_{0.65}$
All INPs	$0.88^{5.25}_{0.57}$	$0.93^{4.95}_{0.70}$	$1.86^{9.22}_{1.24}$	$0.90^{20.12}_{0.61}$	$1.09^{7.42}_{0.66}$	$1.21^{9.92}_{0.75}$	$0.28^{2.54}_{0.11}$	$1.26^{5.84}_{0.95}$

Note: '\' indicates that there was no such class in all particles.

Table S3 The a and b values in the parameterizations of n_s for the DIN and IMF, $n_s = 10^{(a \times T + b)}$.

Temperature	Type	a	b
205K-DIN	FreshSS+AgedSS	– 0.46	98.38
210K-DIN	Dust	– 1.08	229.50
	BBA	– 0.93	197.79
	CNOS+SS/Sulf	– 0.33	72.72
	FreshSS+AgedSS	– 0.45	96.37
215K-DIN	Dust	– 1.89	410.07
	BBA	– 1.05	228.38
	CNOS+SS/Sulf	– 2.35	505.43
	FreshSS+AgedSS	– 0.55	121.04
220K-DIN	Dust	– 0.73	163.69
	BBA	– 3.34	738.29
	CNOS+SS/Sulf	– 2.65	587.31
	FreshSS+AgedSS	– 0.32	72.87
225K-DIN	Dust	– 1.62	368.58
	BBA	– 1.29	293.44
	CNOS+SS/Sulf	– 1.40	317.75
225K-IMF	FreshSS+AgedSS	– 0.89	204.49
230K-DIN	Dust	– 1.42	328.27
	BBA	– 1.47	340.32
235K-DIN	Dust	– 0.54	130.47