

Development and characterization of an ice-selecting pumped counterflow virtual impactor (IS-PCVI) to study ice crystal residuals

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Supplementary information

15 Aerosol types and associated particle generators used in this work are summarized in Table S1. Detailed characteristics of IS-PCVI properties are summarized in Table S2. During INUIT05, the output flow was fixed at 2 lpm. Contrarily, the output flow was varied from 2.5 to 6 lpm in the FIN01 measurements listed in the table, depending on the flow requirements of the instruments deployed downstream of IS-PCVI. Note that varying the output flow only influences a concentration enhancement factor (Sect. 3.3) and does not affect the cut-size (Sect. 4.4). Other information regarding particle properties
20 (i.e., concentration and size distribution of aerosol, droplet and/or ice) before and during individual AIDA expansion experiments are available upon request (contact: Naruki Hiranuma, seong.moon@kit.edu). Temporal profiles of the AIDA cloud simulation experiments (as illustrated in Fig. 6) can also be provided.

Table S1. List of aerosol types and particle generation techniques.

Aerosol type	Generator	Exp. ID	Reference
Ammonium sulfate	Custom-made atomizer	INUIT05_29-37	-
Sodium chloride	Custom-made atomizer	INUIT05_51, 55-58, 60-64	-
Snomax	Custom-made atomizer	INUIT05_22	<i>Wex et al.</i> , 2015
PF CGina bacteria	Custom-made atomizer	FIN01_38	-
H ₂ O	Home-built nozzle spray	INUIT05_59	-
Ethiopia volcanic soil (VSE01)*	Rotating brush (Palas, RGB1000)	FIN01_18	-
Illite NX	Rotating brush (Palas, RGB1000)	FIN01_4-8, 28-29	<i>Hiranuma et al.</i> , 2015
Argentinian soil dust	Rotating brush (Palas, RGB1000)	FIN01_49	<i>Steinke</i> . 2013
K-rich Feldspar (FS01: microcline 76%, albite 24%)	Rotating brush (Palas, RGB1000)	FIN01_11-15, 29, 31, 33	<i>Peckhouse et al.</i> , 2016
K-rich Feldspar (FS04: microcline 80%, albite 18%, quartz 2%)	Rotating brush (Palas, RGB1000)	FIN01_46, 50-51, 53, 55	-
H ₂ SO ₄	In situ [†]	FIN01_27, 31, 33	-
SOA	In situ [‡]	FIN01_46, 51, 53, 55	<i>Saathoff et al.</i> 2009
Hematite	Small-Scale Powder Disperser (SSPD; TSI, Model 3433)	INUIT05_1-13	<i>Hiranuma et al.</i> , 2014
Soot (organic carbon content ~10%)	Graphite Spark Generator (GfG-1000)	FIN01_21, 38, 41; INUIT05_37, 63, 64	<i>Möhler et al.</i> , 2005b; <i>Helsper et al.</i> , 1993
Soot (organic carbon content between 16 and 40%)	CAST (Combustion Aerosol Standard) burner	FIN01_27	<i>Möhler et al.</i> , 2005a

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*63 µm sieved. XRD data is available upon request.

[†]H₂SO₄ was produced through in situ formation from SO₂ + OH reaction (nucleation and condensation growth). Ozone concentration was typically 900 ppb. Continuous addition of trimethylolethane created OH radical concentrations in the range 10⁶-10⁷ cm⁻³, which then oxidized SO₂ to H₂SO₄.

[‡]α-pinene SOA was produced via in situ formation from ozonolysis of α-pinene (nucleation and condensation growth). Ozone concentration was in the range of 150-250 ppb.

Table S2. Characterization of IS-PCVI properties during the AIDA expansion experiments. The critical cut-size [\pm standard deviation (Std. dev.)] of droplets and ice crystals are reflected in Fig. 8 and Fig. 9, respectively.

Experiment ID	Reference time (CET)	Aerosol type	Activation type	PCVI properties				
				IF (lpm)	CF (lpm)	CF-to-IF ratio	Critical cut-size, D_c (μm)	Std. dev.
INUIT05_22_c	5/21/2013 15:02:07	Snomax	Droplet	100.0	9.0	0.09	10.86	0.80
INUIT05_22_d	5/21/2013 15:03:13	Snomax	Droplet	100.0	12.0	0.12	12.60	0.65
INUIT05_22_f	5/21/2013 15:05:45	Snomax	Droplet	100.0	12.0	0.12	13.38	1.13
INUIT05_29	5/22/2013 15:13:00	(NH ₄) ₂ SO ₄	Droplet	70.0	7.0	0.10	15.32	1.85
INUIT05_30	5/22/2013 16:06:00	(NH ₄) ₂ SO ₄	Droplet	70.0	9.0	0.13	17.40	2.81
INUIT05_31	5/22/2013 16:56:01	(NH ₄) ₂ SO ₄	Droplet	70.0	6.0	0.09	13.08	1.68
INUIT05_32	5/23/2013 10:12:00	(NH ₄) ₂ SO ₄	Droplet	70.0	9.0	0.13	19.86	3.15
INUIT05_33	5/23/2013 11:19:00	(NH ₄) ₂ SO ₄	Droplet	70.0	9.0	0.13	20.13	3.30
INUIT05_34	5/23/2013 12:28:00	(NH ₄) ₂ SO ₄	Droplet	69.0	9.0	0.13	21.50	3.91
INUIT05_35	5/23/2013 13:28:00	(NH ₄) ₂ SO ₄	Droplet	70.0	9.0	0.13	17.57	2.66
INUIT05_36	5/23/2013 14:20:00	(NH ₄) ₂ SO ₄	Droplet	70.0	9.0	0.13	17.46	2.65
INUIT05_37	5/23/2013 15:51:00	(NH ₄) ₂ SO ₄ + GSG soot	Droplet	70.0	9.0	0.13	16.84	2.17
INUIT05_60_b	6/10/2013 9:45:29	NaCl	Droplet	100.0	9.0	0.09	10.78	0.77
INUIT05_61_b	6/10/2013 11:13:00	NaCl	Droplet	50.0	9.0	0.18	24.72	2.16
INUIT05_61_c	6/10/2013 11:15:00	NaCl	Droplet	50.0	7.0	0.14	21.99	1.20
INUIT05_61_d	6/10/2013 11:17:01	NaCl	Droplet	50.0	6.0	0.12	22.10	0.80
INUIT05_61_g	6/10/2013 11:23:02	NaCl	Droplet	50.0	7.0	0.14	20.81	1.82
INUIT05_62_b	6/10/2013 12:30:30	NaCl	Droplet	70.0	10.5	0.15	24.28	1.31
INUIT05_62_e	6/10/2013 12:36:00	NaCl	Droplet	70.0	12.0	0.17	27.29	3.76
INUIT05_63	6/10/2013 14:54:00	NaCl + GSG soot	Droplet	70.0	9.0	0.13	17.67	2.70
INUIT05_64_a	6/10/2013 16:17:00	NaCl + GSG soot	Droplet	100.0	10.0	0.10	12.59	N/A
INUIT05_64_c	6/10/2013 16:23:00	NaCl + GSG soot	Droplet	100.0	12.0	0.12	16.19	1.45
FIN01_4	11/6/2014 15:30:00	Illite NX	Immersion	70.0	9.5	0.14	23.77	2.99
FIN01_5	11/6/2014 17:44:00	Illite NX	Immersion	70.0	11.0	0.16	22.96	6.56
FIN01_6	11/6/2014 18:54:00	Illite NX	Immersion	70.0	11.5	0.16	27.02	3.74
FIN01_7	11/7/2014 12:23:00	Illite NX	Immersion	70.0	11.5	0.16	29.80	6.82
FIN01_8	11/7/2014 16:05:00	Illite NX	Immersion	71.2	12.7	0.18	30.38	5.22
FIN01_10	11/8/2014 12:08:00	K-rich feldspar (FS01)	Immersion	70.0	11.0	0.16	29.72	2.60
FIN01_11	11/8/2014 13:50:00	FS01	Immersion	70.0	9.0	0.13	23.21	1.66
FIN01_12_a	11/8/2014 15:58:00	FS01	Immersion	85.0	9.0	0.11	22.22	2.61
FIN01_12_d	11/8/2014 16:03:00	FS01	Immersion	85.0	8.0	0.09	20.10	0.91
FIN01_12_e	11/8/2014 16:04:00	FS01	Immersion	85.0	9.0	0.11	22.27	1.71
FIN01_13_a	11/8/2014 17:49:00	FS01	Immersion	50.0	6.5	0.13	24.93	1.84
FIN01_13_b	11/8/2014 17:53:00	FS01	Immersion	50.0	5.5	0.11	21.95	1.63
FIN01_13_c	11/8/2014 17:54:00	FS01	Immersion	50.0	5.0	0.10	21.58	3.24

FIN01_14_a	11/10/2014 10:24:00	FS01	Immersion	85.0	12.0	0.14	23.10	3.58
FIN01_14_b	11/10/2014 10:26:20	FS01	Immersion	85.0	8.0	0.09	17.67	0.78
FIN01_14_c	11/10/2014 10:29:25	FS01	Immersion	85.0	12.0	0.14	25.33	1.66
FIN01_15_a	11/10/2014 11:57:00	FS01	Immersion	85.0	12.0	0.14	25.95	2.73
FIN01_15_b	11/10/2014 12:01:00	FS01	Immersion	85.0	8.0	0.09	19.56	4.30
FIN01_15_c	11/10/2014 12:08:10	FS01	Immersion	85.0	12.0	0.14	24.36	10.54
FIN01_18_a	11/11/2014 11:11:11	Ethiopia volcanic soil (VSE01)	Immersion	75.0	11.5	0.15	27.22	3.96
FIN01_18_b	11/11/2014 11:16:10	VSE01	Immersion	75.0	9.0	0.12	26.26	4.03
FIN01_19_b	11/11/2014 12:38:15	VSE01	Immersion	75.0	9.0	0.12	26.72	3.80
FIN01_21_a	11/12/2014 10:31:00	GSG soot	Deposition	75.0	11.5	0.15	27.70	5.98
FIN01_21_b	11/12/2014 10:36:00	GSG soot	Deposition	75.0	9.0	0.12	21.21	3.71
FIN01_25_b	11/13/2014 12:31:45	H ₂ SO ₄	Deposition	75.0	9.0	0.12	24.61	3.97
FIN01_27_b	11/13/2014 18:22:10	H ₂ SO ₄ + CAST soot	Deposition	85.0	8.5	0.10	23.12	4.45
FIN01_28_b	11/14/2014 11:04:20	Illite NX	Deposition	85.0	8.0	0.09	16.13	1.11
FIN01_28_d	11/14/2014 11:16:30	Illite NX	Deposition	85.0	8.0	0.09	19.17	4.93
FIN01_29_a	11/14/2014 16:19:00	Illite NX + FS01	Deposition	85.0	9.0	0.11	20.74	6.81
FIN01_29_b	11/14/2014 16:21:30	Illite NX + FS01	Deposition	85.0	7.5	0.09	17.42	2.78
FIN01_29_e	11/14/2014 16:27:10	Illite NX + FS01	Deposition	105.0	9.0	0.09	14.14	1.46
FIN01_31_b	11/15/2014 17:30:50	FS01 + H ₂ SO ₄	Deposition	80.0	8.0	0.10	15.63	2.51
FIN01_31_c	11/15/2014 17:31:30	FS01 + H ₂ SO ₄	Deposition	90.0	8.0	0.09	14.59	7.44
FIN01_31_d	11/15/2014 17:33:20	FS01 + H ₂ SO ₄	Deposition	85.0	8.0	0.09	22.37	1.28
FIN01_31_e	11/15/2014 17:34:20	FS01 + H ₂ SO ₄	Deposition	85.0	7.5	0.09	16.35	4.81
FIN01_33_b	11/17/2014 17:27:30	FS01 + H ₂ SO ₄	Immersion	90.0	16.0	0.18	25.03	2.77
FIN01_33_c	11/17/2014 17:28:45	FS01 + H ₂ SO ₄	Immersion	90.0	12.0	0.13	20.10	2.03
FIN01_33_d	11/17/2014 17:29:37	FS01 + H ₂ SO ₄	Immersion	90.0	10.0	0.11	15.64	4.94
FIN01_33_f	11/17/2014 17:31:45	FS01 + H ₂ SO ₄	Immersion	90.0	10.0	0.11	15.64	7.54
FIN01_46	11/20/2014 17:13:00	K-rich Feldspar (FS04) + α - pinene SOA	Immersion	90.0	15.0	0.17	24.96	4.77
FIN01_49	11/22/2014 13:54:00	Argentinian soil dust	Immersion	90.0	14.0	0.16	23.11	6.55
FIN01_50_c	11/24/2014 11:07:30	FS04	Deposition	80.0	8.0	0.10	16.03	2.05
FIN01_50_d	11/24/2014 11:08:30	FS04	Deposition	80.0	10.0	0.13	22.37	3.87
FIN01_51_b	11/24/2014 15:52:00	FS04 + α - pinene SOA	Deposition	80.0	8.0	0.10	21.34	5.98
FIN01_53	11/25/2014 12:30:00	FS04 + α - pinene SOA	Deposition	80.0	10.0	0.13	27.05	2.78
FIN01_55_b	11/25/2014 16:58:00	FS04 + α - pinene SOA	Deposition	75.0	10.0	0.13	26.84	3.52

References

- Helsper, C., Molter, W., Löffler, F., Wadenpohl, C., and Kaufmann, S.: Investigation of a new aerosol generator for the production of carbon aggregate particles, *Atmos. Environ.*, 27A, 1271–1275, doi:10.1016/0960-1686(93)90254-V, 1993.
- 5 Hiranuma, N., Hoffmann, N., Kiselev, A., Dreyer, A., Zhang, K., Kulkarni, G., Koop, T., and Möhler, O.: Influence of surface morphology on the immersion mode ice nucleation efficiency of hematite particles, *Atmos. Chem. Phys.*, 14, 2315–2324, doi:10.5194/acp-14-2315-2014, 2014.
- Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., Diehl, K., Dreischmeier, K., Ebert, M., Frank, F., Hoffmann, N., Kandler, K., Kiselev, A., Koop, T., Leisner, T., Möhler, O., Nillius, B., Peckhaus, A., Rose, D., Weinbruch, S., Wex, H., Boose, Y., DeMott, P. J., Hader, J. D., Hill, T. C. J., Kanji, Z. A., Kulkarni, G., Levin, E. J. T., McCluskey, C. S., Murakami, M., Murray, B. J., Niedermeier, D., Petters, M. D., O'Sullivan, D., Saito, A., Schill, G. P., Tajiri, T., Tolbert, M. A., Welti, A., Whale, T. F., Wright, T. P., and Yamashita, K.: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, *Atmos. Chem. Phys.*, 15, 2489–2518, doi:10.5194/acp-15-2489-2015, 2015.
- 10 Möhler, O., Linke, C., Saathoff, H., Schnaiter, M., Wagner, R., Mangold, A., Kramer, M., and Schurath, U.: Ice nucleation on flame soot aerosol of different organic carbon content, *Meteorol. Z.*, 14, 477–484, doi: 10.1127/0941-2948/2005/0055, 2005a.
- 20 Möhler, O., Büttner, S., Linke, C., Schnaiter, M., Saathoff, H., Stetzer, O., Wagner, R., Kramer, M., Mangold, A., Ebert, V., and Schurath, U.: Effect of sulphuric acid coating on heterogeneous ice nucleation by soot aerosol particles, *J. Geophys. Res.*, 110, D11 210, doi:10.1029/2004JD005 169, 2005b.
- 25 Peckhaus, A., Kiselev, A., Hiron, T., Ebert, M., and Leisner, T.: A comparative study of K-rich and Na/Ca-rich feldspar ice nucleating particles in a nanoliter droplet freezing assay, *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-72, 2016.
- Saathoff, H., Naumann, K.-H., Möhler, O., Jonsson, Å. M., Hallquist, M., Kiendler-Scharr, A., Mentel, Th. F., Tillmann, R., and Schurath, U.: Temperature dependence of yields of secondary organic aerosols from the ozonolysis of α -pinene and limonene, *Atmos. Chem. Phys.*, 9, 1551–1577, doi:10.5194/acp-9-1551-2009, 2009.
- 30 Steinke, I.: Ice nucleation properties of mineral dusts. PhD thesis, Karlsruhe Institute of Technology, Karlsruhe, Germany, 160 pp., 2013.
- 35 Wex, H., Augustin-Bauditz, S., Boose, Y., Budke, C., Curtius, J., Diehl, K., Dreyer, A., Frank, F., Hartmann, S., Hiranuma, N., Jantsch, E., Kanji, Z. A., Kiselev, A., Koop, T., Möhler, O., Niedermeier, D., Nillius, B., Rösch, M., Rose, D., Schmidt, C., Steinke, I., and Stratmann, F.: Intercomparing different devices for the investigation of ice nucleating particles using Snomax® as test substance, *Atmos. Chem. Phys.*, 15, 1463–1485, doi:10.5194/acp-15-1463-2015, 2015.