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Supplement of

Marine carbohydrates and other sea spray aerosol constituents across altitudes in the lower troposphere of Ny-Ålesund, Svalbard

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A1: Collection efficiency for TSP sampling using HALFBAC

The HALFBAC sampler (Grawe et al., 2023) was secured directly below the balloon using ropes. The inlet (20 cm, ½" conductive tubing) was positioned facing into the wind but allowed some degree of rotational motion between -90° and 90°.

To assess particle losses, calculations were performed using the Particle Loss Calculator (von der Weiden et al., 2012), considering aspiration efficiency and losses through the inlet (**Figure S1**). Four scenarios were evaluated, varying the inlet orientation (0–90°) and wind speed (1 and 5 m s^{-1}). The resulting D_{50} values, which means the aerosol diameter where still 50% of the particles at this size range is collected, ranged from 4.6 µm to 35.1 µm, indicating that the system is generally suitable for collecting supermicron particles.

For particles smaller than the filter pore size (> 0.8 µm), transmission through the inlet is possible, though collection on the polycarbonate filters may be less efficient. Soo et al. (2016) suggested a minimum collection efficiency of >90% for particles around 100 nm, confirming the system's suitability for submicron particle collection as well.

Overall, this TSP sampling setup effectively collects aerosol particles across a size range that may closely align with PM_{10}.

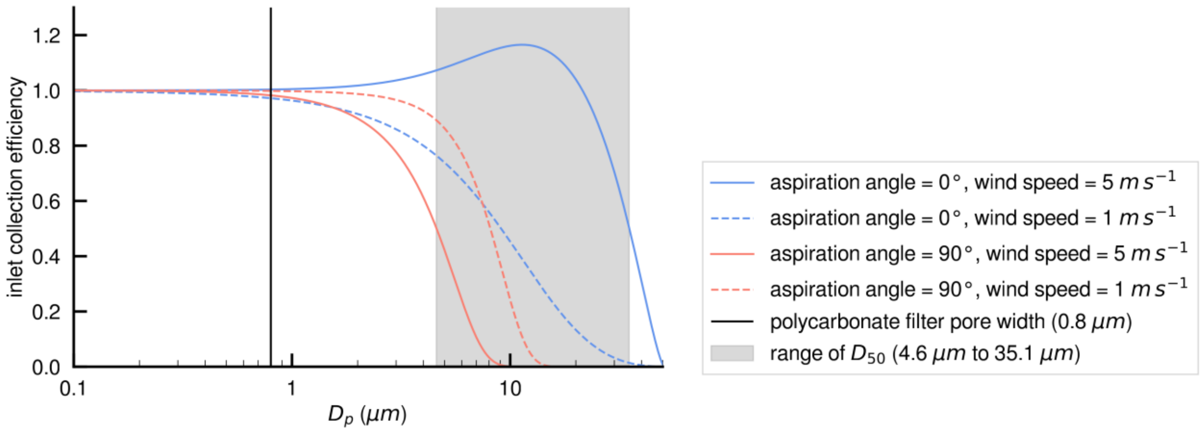

Figure S1. Diameter-dependent aerosol particle collection efficiency of total suspended particles using HALFBAC, calculated with the Particle Loss Calculator (von der Widen et al., 2012). The analysis considers two aspiration angles and wind speeds within the operational range for balloon flights. The grey area represents the D_{50} range, indicating the particle diameters where less than 50% of aerosols are collected, depending on sampling conditions (sampling orientation and wind speed).

A2: Good agreement between HALFBAC aerosol sampling and routine Zeppelin Observatory measurements

At the Zeppelin Observatory, Filter_3pack is routinely operated by NILU for aerosol particle monitoring with a time resolution of 24 hours. The major inorganic ions are among the key chemical constituents typically measured from these filters. Since these chemical parameters were also part of our study, we conducted a test to assess comparability.

On 10 May 2022, HALFBAC was used for TSP sampling over a two-hour period at the Zeppelin Observatory, near Filter_3pack (**Figure S2a**). Despite differences in time resolution, sampling methods and chemical analysis protocols, four analyzed chemical parameters, sodium (0.85 & 0.85 µg m^{-3}), potassium (0.03 & 0.07 µg m^{-3}), chloride (1.28 & 1.71 µg m^{-3}), and sulfate (1.04 & 0.89 µg m^{-3}) (first value: Filter_3pack, second: HALFBAC), showed good agreement (**Figure S2b)**. Additional parameters could not be included due to detection limit constraints.

Although this was a single, not entirely ideal comparability test, the results suggest that both approaches produce comparable outcomes.

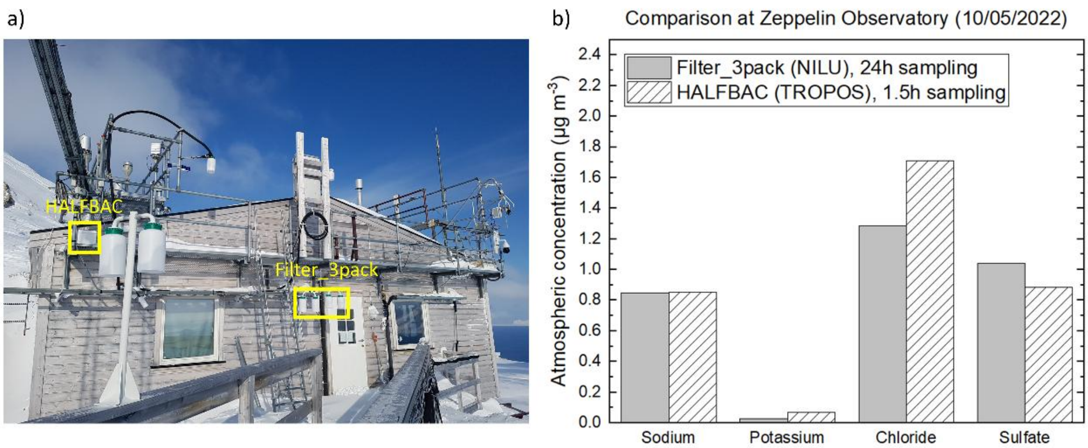

Figure S2. a) Photo of the Zeppelin Observatory showing the standard TSP sampling with the Filter_3pack and a one-time TSP aerosol sampling using the HALFBAC sampler on 10 May 2022; b) Comparison of major inorganic ions in ambient total suspended aerosol particles (TSP) collected with the HALFBAC and Filter_3pack sampling instruments. Both instruments were operated at the Zeppelin Observatory in Ny-Ålesund. HALFBAC (Sample 62, 10/05/2022, 07:00–08:32 UTC, TSP, 0.8 µm polycarbonate filter) vs. Filter_3pack (10/05/2021, 07:00 – 11/05/2022, 07:00 UTC).

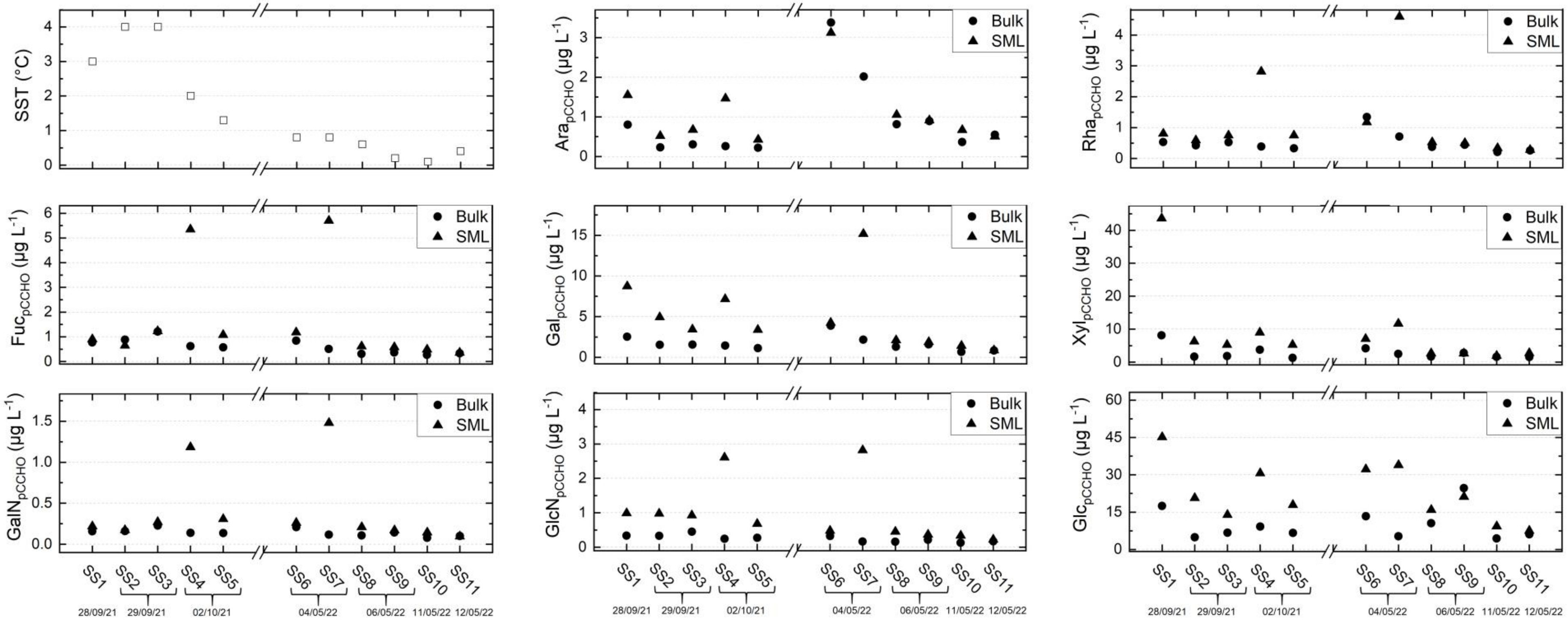

Figure S3. Concentration of measured monosaccharide units in pCCHO from bulk and SML samples collected in Kongsfjorden during autumn 2021 and spring 2022, together with SST measured in bulk samples at the time of collection.

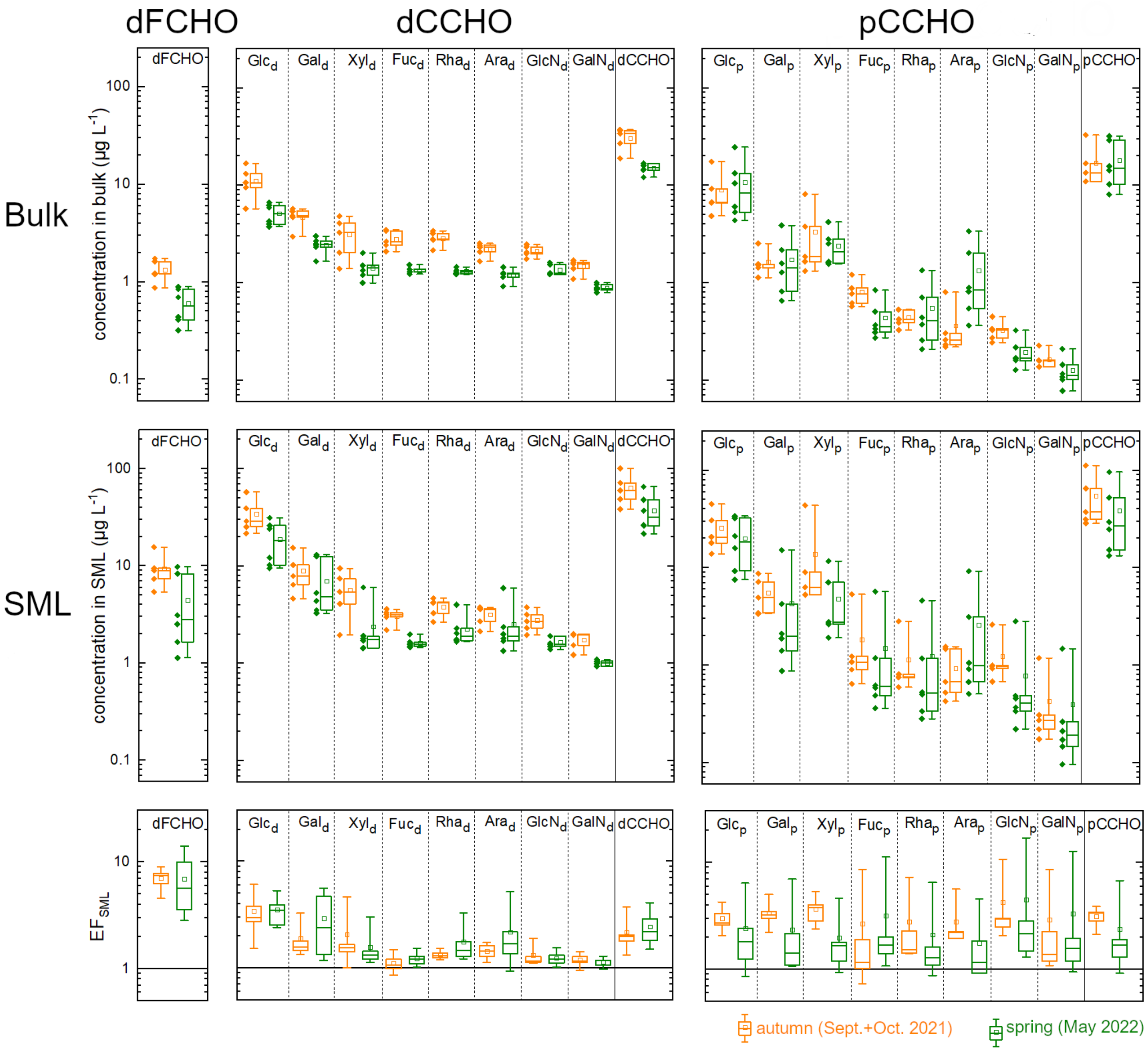

Figure S4. Box-whisker plots illustrating the inter-seasonal and intra-seasonal variability of individual monosaccharide units in dissolved free carbohydrates (dFCHO), dCCHO, and pCCHO from bulk and SML samples in Kongsfjorden, separated into autumn 2021 and spring 2022, along with enrichment factors in the SML (EF_{SML}).

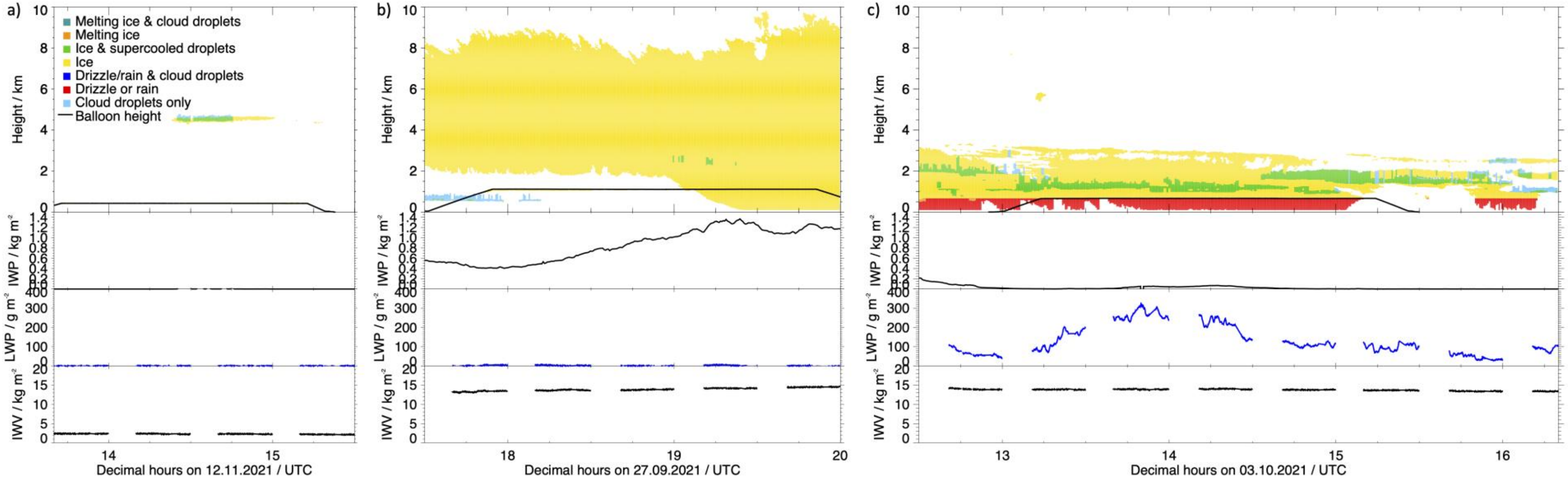

Figure S5. Clouds and hydrometer types as a Cloudnet classification product, ice water path (IWP), cloud liquid water path (LWP) and integrated water vapor (IWV) from three selected cases: a) 12 November 2021 (Case I), b) 27 September 2021 (Case II), c) 03 October 2021 (Case III).

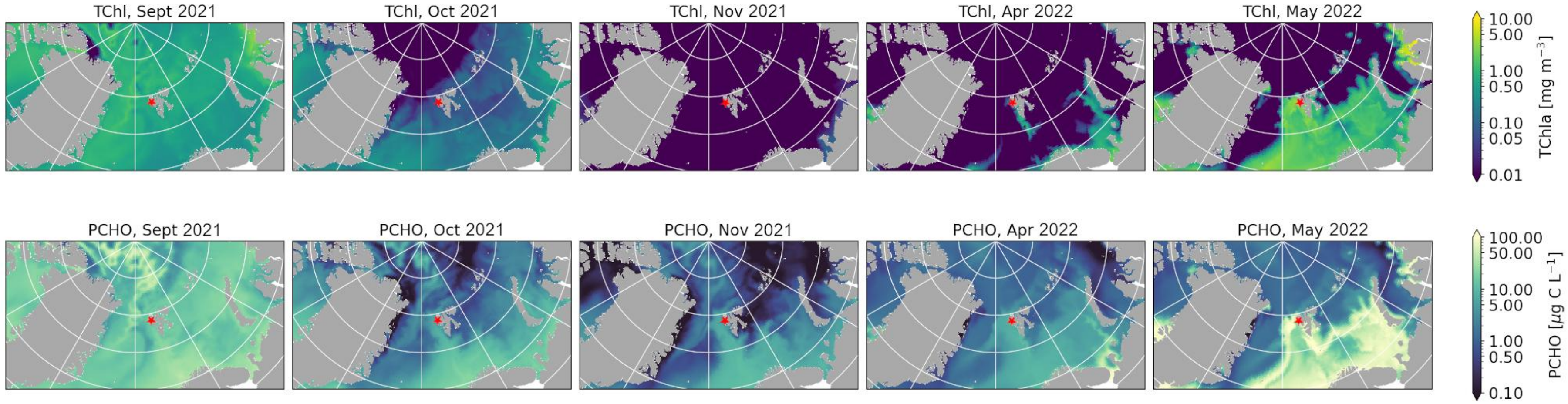

Figure S6. Modeled total chlorophyll a (TChl-a) and dissolved acidic polysaccharides (PCHO) in the Northern Atlantic, Fram Strait, Barents Sea, and Arctic Ocean from FESOM2.1-REcoM3 (Gürses et al.,2023) for September–November 2021 and April–May 2022. The red star marks the measurement location (Ny-Ålesund).

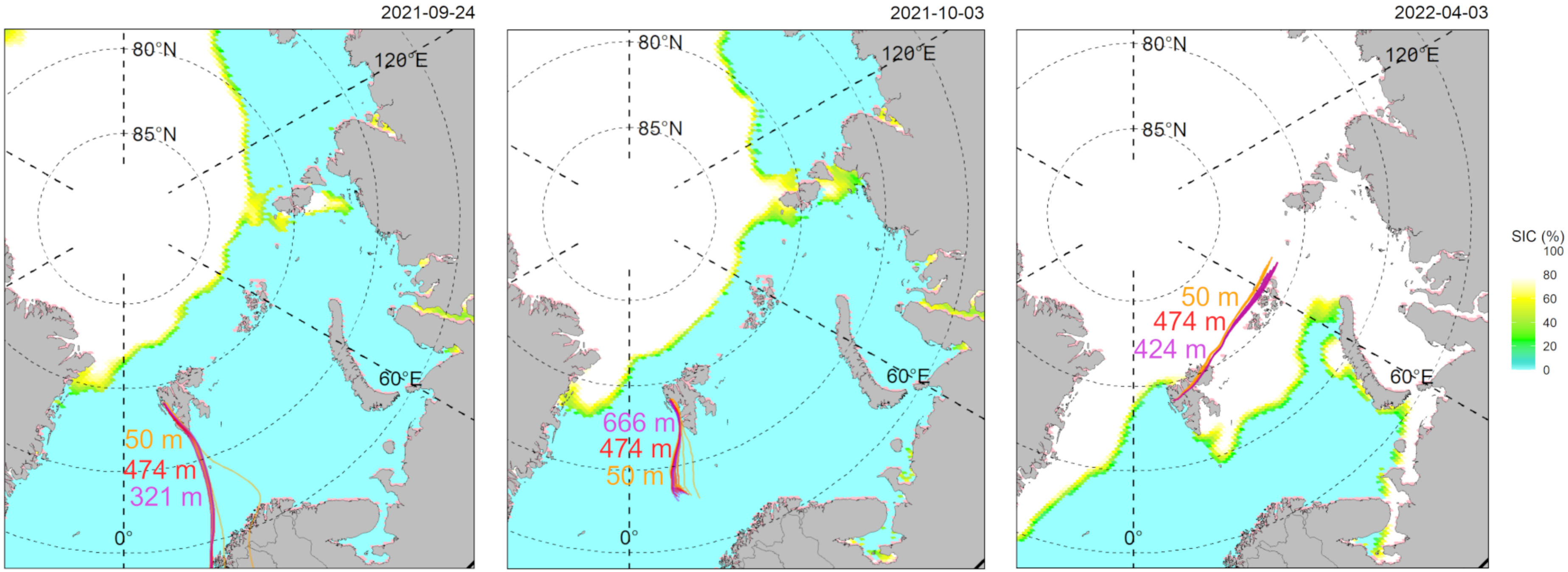

Figure S7. Hourly 48-hour back-trajectories for three arrival heights: orange (50 m, ground-level air masses), red (474 m, height of the Zeppelin Observatory), and purple (variable arrival height, high-altitude air masses sampled at tethered balloon). These trajectories are presented alongside daily sea ice concentration (SIC) maps for three selected aerosol sampling cases, where $CCHO_{aer}$ concentrations at the balloon were significantly higher than at the ground**.** However, in these cases, air masses did not pass through the biologically active marginal ice zone (SIC: 15–80%) prior to sampling in Ny-Ålesund**.**

Table S1. Bulk and SML samples collected from Kongsfjorden. SST=Sea surface temperature.

Sample ID	Sampling time (UTC)	Latitude (°N)	Longitude (°E)	Weather Conditions	SST (°C)
Bulk 1 SML 1	28/09/2021 10:00	78.9375	11.9990	Sunny, very little waves	3.0
Bulk 2 SML 2	29/09/2021 09:20	78.9273	12.0087	Windy, cloudy, waves up to 1 m	4.0
Bulk 3 SML 3	29/09/2021 09:40	78.9342	11.9293	Windy, cloudy, waves up to 1 m	4.0
Bulk 4 SML 4	02/10/2021 11:30	78.9485	12.0798	Sunny, very little waves	2.0
Bulk 5 SML 5	02/10/2021 12:30	78.9639	12.2023	Sunny, very little waves	1.3
Bulk 6 SML 6	04/05/2022 09:36	78.9328	11.9684	Mostly sunny, few clouds, no wind waves, but swells	0.8
Bulk 7 SML 7	04/05/2022 09:59	78.9530	11.9180	Mostly sunny, few clouds, no wind waves, but swells	0.8
Bulk 8 SML 8	06/05/2022 12:04	78.9222	12.1163	Cloudy, little wind	0.6
Bulk 9 SML 9	06/05/2022 12:36	78.9348	12.1556	Cloudy, little wind	0.2
Bulk 10 SML 10	11/05/2022 11:45	78.9800	12.3410	Sunny, no wind, very calm sea	0.1
Bulk 11 SML 11	12/05/2022 13:46	78.9001	12.3413	Sunny, no wind, very calm sea	0.4

Table S2. Aerosol sampling at the Old Pier. Average air temperatures were calculated from data measured 2 meters above ground (13 meters above sea level) at the AWIPEV Atmospheric Observatory (Maturilli, 2020).

Start (UTC)	Stop (UTC)	Sample ID	Air temperature (°C)
25/09/2021 11:30	30/09/2021 14:30	Old Pier 1	2.2
30/09/2021 14:30	06/10/2021 08:50	Old Pier 2	2.8
06/10/2021 08:50	11/10/2021 08:30	Old Pier 3	-3.8
11/10/2021 08:30	18/10/2021 11:56	Old Pier 4	-3.5
24/10/2021 12:07	30/10/2021 14:08	Old Pier 6	-5.4
30/04/2022 14:07	05/05/2022 08:11	Old Pier 8	-9.7
05/05/2022 08:11	09/05/2022 12:00	Old Pier 9	-8.1
09/05/2022 12:00	14/05/2022 10:20	Old Pier 10	-6.8

Table S3. Aerosol sampling at ground (winch), balloon and Zeppelin Observatory.

Date	Ground (at winch)			High altitude (at balloon)			
	Sample ID	Start (UTC)	Stop (UTC)	Sample ID	Start (UTC)	Stop (UTC)	Height (m) *median (min-max)*
24/09/2021	5	15:57	17:54	4	15:50	17:02	321 (151-324)[1]
25/09/2021	7	12:31	14:31	-	-	-	-
27/09/2021	12	17:31	19:52	11	17:50	19:55	1112 (913-1118)[1]
30/09/2021	14	18:03	20:03	13	17:57	19:57	760 (648-762)[1]
01/10/2021	16	11:49	13:53	15	11:50	13:50	359 (340-517)[1]
02/10/2021	18	12:00	14:00	17	11:57	13:57	1003 (629-1009)[1]
03/10/2021	20	12:35	16:15	19	13:10	15:19	666 (373-671)[1]
09/10/2021	24	13:15	15:10	23	13:15	15:05	731 (412-735)[1]
13/10/2021	25	08:33	13:50	-	-	-	-
14/10/2021	27	12:15	16:15	26	12:55	15:00	618 (429-629)[1]
05/11/2021	46	14:40	16:55	-	-	-	-
07/11/2021	49	09:57	14:05	-	-	-	-
08/11/2021	51	18:10	20:15	50	17:55	20:05	449 (51-450)[1]
11/11/2021	-	-	-	52	16:45	18:45	410 (16-415)[2]
12/11/2021	53	13:47	15:26	54	13:44	15:13	428 (402-430)[1]
03/04/2022	58	16:30	18:30	59	16:30	18:30	424 (110-430)[1]
05/04/2022	60	10:06	12:06	61	10:06	12:06	588 (583-590)[1]
10/05/2022	-	-	-	62	07:00	08:32	474 (Zeppelin Observ.)
11/05/2022	64	13:30	15:30	65	10:30	12:30	900 (566-908)[1]

[1]Heights measured at standard meteorology package, [2]heights measured at HALFBAC

Table S4. Meteorological data from the standard meteorology package during the aerosol particle sampling at the balloon (excluding the ascents and descents), presented as mean (min – max). The green-highlighted rows indicate cases discussed in detail in section 3.2 of the main article.

Sample ID	Date	Air pressure (hPa)	Temperature (°C)	Relative humidity (%)	Wind speed (m s^{-1})	Wind direction (%)							
						000°-045°	045°-090°	090°-135°	135°-180°	180°-225°	225°-270°	270°-315°	315°-360°
4	24/09/2021	957 (956 – 976)	4.6 (3.4 – 5.7)	79 (73 – 84)	7.5 (4.5 – 9.2)	0	81	19	0	0	0	0	0
11	27/09/2021	886 (885 – 907)	-1.9 (-4.8 – 0.9)	87 (83 – 89)	5.5 (3.8 – 6.9)	0	0	0	3	96	1	0	0
13	30/09/2021	935 (934 – 944)	3.7(3.4 – 4.1)	71 (70 – 73)	5.9 (3.3– 11.0)	0	0	100	0	0	0	0	0
15	01/10/2021	973 (955 – 975)	2.1 (1.3 – 2.7)	83 (80 – 89)	6.6 (4.2 – 8.9)	0	81	19	0	0	0	0	0
17	02/10/2021	890 (888 – 931)	-2.3 (-2.6 – 0.7)	88 (80 – 92)	6.7 (5.8 – 7.8)	0	99	1	0	0	0	0	0
19	03/10/2021	929 (928 – 957)	-1.3 (-1.7 – 0.4)	96 (86 – 99)	6.8 (3.5 – 10.4)	0	62	38	0	0	0	0	0
23	09/10/2021	932 (930 – 969)	-8.9 (-9.3 – -6.8)	75 (66 – 82)	4.2 (3.2 – 5.2)	52	4	0	0	0	0	0	44
26	14/10/2021	927 (925 – 949)	-5.0 (-5.2 – -3.5)	68 (61 – 70)	8.5 (7.3 – 9.4)	0	100	0	0	0	0	0	0
50	08/11/2021	960 (956 – 1009)	-12.6 (-13.1 – -10.7)	79 (75 – 87)	3.7 (2.2 – 5.5)	19	29	32	14	2	0	1	3
52	11/11/2021	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
54	12/11/2021	958 (958 – 961)	-17.5 (-17.9 – - 17.2)	72 (65 – 76)	4.3 (2.9 – 6.3)	8	61	31	0	0	0	0	0
59	03/04/2022	961 (959 – 993)	-16.2 (-16.6 – -15.3)	54 (51 – 66)	5.2 (4.6 – 6.2)	0	100	0	0	0	0	0	0
61	05/04/2022	941 (941 – 944)	-10.9 (-12.4 – -8.3)	36 (24 – 47)	3.4 (2.9 – 3.9)	57	2	0	0	0	2	2	37
65	11/05/2022	899 (896 – 936)	-11.8 (-12.3 – -7.7)	55 (41 – 61)	4.1 (3.3 – 4.7)	2	97	1	0	0	0	0	0

Table S5. Meteorological data from the AWIPEV Atmospheric Observatory (Maturilli, 2020), measured at 2 meters above ground (13 meters above sea level), represent the weather conditions during aerosol particle sampling at the winch. The data is presented as mean (min – max) values. Rows highlighted in green correspond to cases discussed in detail in section 3.2 of the main article.

Sample ID	Date	Air pressure (hPa)	Temperature (°C)	Relative humidity (%)	Wind speed (m s^{-1})	Wind direction (%)							
						000°-045°	045°-090°	090°-135°	135°-180°	180°-225°	225°-270°	270°-315°	315°-360°
5	24/09/2021	995 (995 – 995)	5.7 (4.8 – 6.6)	80 (74 – 84)	0.7 (0.1 – 1.5)	0	0	0	0	28	34	28	10
7	25/09/2021	1000 (1000 – 1000)	5.0 (4.6 – 5.6)	81 (76 – 86)	2.2 (0.1 – 4.3)	7	77	0	0	5	6	5	0
12	27/09/2021	1018 (1018 – 1018)	0.4 (0.2 – 0.6)	83 (80 – 86)	0.7 (0.2 – 1.2)	0	0	0	1	13	74	12	0
14	30/09/2021	1026 (1026 – 1027)	1.7 (0.6 – 2.3)	96 (93 – 100)	1.0 (0.4 – 1.9)	0	0	0	33	12	51	4	0
16	01/10/2021	1017 (1017 – 1018)	2.2 (1.8 – 2.8)	87 (81 – 91)	1.2 (0.4 – 2.2)	0	0	0	0	1	69	26	4
18	02/10/2021	1007 (1006 – 1007)	2.4 (1.5 – 3.0)	83 (80 – 89)	1.2 (0.7 – 1.8)	0	0	4	66	23	7	0	0
20	03/10/2021	1008 (1007 – 1008)	3.0 (2.1 – 4.3)	89 (80 – 96)	0.7 (0.1 – 1.6)	0	2	14	18	15	43	5	3
24	09/10/2021	1021 (1021 – 1022)	-6.1 (-7.8 – -5.5)	70 (65 – 79)	1.0 (0.1 – 1.6)	0	0	1	26	23	50	0	0
25	13/10/2021	1007 (1007 – 1008)	0.2 (-0.6 – 1.2)	71 (64 – 79)	4.2 (1.9 – 7.6)	0	0	78	22	0	0	0	0
27	14/10/2021	1002 (1001 – 1002)	-3.7 (-4.7 – -2.5)	63 (56 – 66)	3.0 (1.5 – 4.7)	0	0	61	39	0	0	0	0
46	05/11/2021	1008 (1008 – 1009)	-10.5 (-12.8 – -8.8)	60 (54 – 71)	1.6 (0.8 – 2.9)	0	0	0	16	18	66	0	0
49	07/11/2021	1011 (1010 – 1012)	-11.2 (-14.8 – -10.2)	58 (51 – 73)	2.0 (0.2 – 5.4)	16	5	5	19	10	9	0	36
51	08/11/2021	1013 (1013 – 1014)	-10.8 (-11.2 – -10.4)	72 (69 – 75)	1.8 (0.7 – 2.7)	0	0	2	77	14	7	0	0
53	12/11/2021	1015 (1014 – 1015)	-16.7 (-21.4 – -14.2)	69 (61 – 83)	1.5 (0.2 – 2.7)	0	0	0	3	34	62	1	0
58	03/04/2022	1015 (1015 – 1015)	-13.4 (-14.1 – -12.5)	53 (51 – 57)	3.2 (1.8 – 4.7)	0	0	19	81	0	0	0	0
60	05/04/2022	1015 (1015 – 1016)	-9.4 (-10.8 – -8.7)	50 (47 – 55)	1.2 (0.4 – 1.9)	0	0	0	32	64	4	0	0
64	11/05/2022	1007 (1007 – 1007)	-6.9 (-7.2 – -6.3)	62 (57 – 67)	1.5 (1.1 – 1.9)	0	0	0	0	100	0	0	0

Table S6. Comparison of atmospheric sodium measurements in TSP between Beluga balloon and Zeppelin Observatory. Sampling resolution differed—1–2 hours for the balloon vs. 24 hours at the Zeppelin Observatory—potentially reflecting different air masses. In cases of significant deviation, adjacent-day data were also considered.

Sampling time at balloon	$Na^{+}_{aer, balloon}$ (ng m^{-3})	Sampling time at Zeppelin Observatory	$Na^{+}_{aer, Zeppelin}$ (ng m^{-3})	$Na^{+}_{aer, balloon}$ / $Na^{+}_{aer, Zeppelin}$ (%)
24.09.2021 15:50 – 17:02	99	24.09.2021 07:00 – 25.09.2021 07:00	75	131
27.09.2021 17:50 – 19:55	23	*27.09.2021 07:00 – 28.09.2021 07:00*	*849*	*3*
		28.09.2021 07:00 – 29.09.2021 07:00	29	80
30.09.2021 17:57 – 19:57	207	30.09.2021 07:00 – 01.10.2021 07:00	97	213
01.10.2021 11:50 – 13:50	30	01.10.2021 07:00 – 02.10.2021 07:00	21	142
02.10.2021 11:57 – 13:57	36	*02.10.2021 07:00 – 03.10.2021 07:00*	*18.8*	*194*
		03.10.2021 07:00 – 04.10.2021 07:00	38	96
03.10.2021 13:10 – 15:19	35	03.10.2021 07:00 – 04.10.2021 07:00	38	92
09.10.2021 13:15 – 15:05	60	08.10.2021 07:00 – 09.10.2021 07:00	60	101
		09.10.2021 07:00 – 10.10.2021 07:00	*119*	*51*
14.10.2021 12:55 – 15:00	180	14.10.2021 07:00 – 15.10.2021 07:00	322	56
08.11.2021 17:55– 20:05	174	07.11.2021 07:00 – 08.11.2021 07:00	251	69
		08.11.2021 07:00 – 09.11.2021 07:00	*568*	*31*
11.11.2021 16:45 – 18:45	183	11.11.2021 07:00 – 12.11.2021 07:00	277	66
12.11.2021 13:44 – 15:13	223	12.11.2021 07:00 – 13.11.2021 07:00	209	107
03.04.2022 16:30 – 18:30	194	03.04.2022 07:00 – 04.04.2022 07:00	137	142
05.04.2022 10:06 – 12:06	54	04.04.2022 07:00 – 05.04.2022 07:00	66	82
		05.04.2022 07:00 – 06.04.2022 07:00	*124*	*44*
10.05.2022 07:00 – 08:32[a]	853	10.05.2022 07:00 – 11.05.2022 07:00	849	101
11.05.2022 10:30 – 12:30	125	11.05.2022 07:00 – 12.05.2022 07:00	89	140

a) sample was taken at the Zeppelin Observatory, not at the balloon.

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