



Supplement of

Atmospheric ice-nucleating particles in the eastern Mediterranean and the contribution of mineral and biological aerosol

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S1 Aerosol sampling details

Table S1: Details of the aerosol samples collected during the field campaign in Rehovot, Israel, in October-November 2018. Samples were either collected using a filter-based platform (BGI PQ100 ambient air samplers with a PM₁₀ inlet, Mesa Laboratories), or using an impinger (Coriolis® Micro, Bertin Technologies), then the particles suspended in purified water of a known volume (i.e. the wash volume, V_{wash}). All times are local, and dates are provided in the DD/MM/YY format.

Date (DD/MM/YY)	Start time	End time	Sampling time (min)	Sampling rate (L min ⁻¹)	Volume of sampled air, V_{air} (L)	Wash volume, V_{wash} (mL)	Sampling method
<u>3 h filter (PQ100) samples</u>							
25/10/18	10:10	11:10	60	16.66	1,000	4	Filter-based
25/10/18	15:07	18:10	183	16.66	3,049	4	Filter-based
26/10/18	12:09	15:17	188	16.66	3,132	4	Filter-based
26/10/18	15:40	18:40	180	16.66	2,999	4	Filter-based
27/10/18	10:20	13:20	180	16.66	2,999	4	Filter-based
27/10/18	15:18	18:18	180	16.66	2,999	4	Filter-based
28/10/18	09:28	12:28	180	16.66	2,999	4	Filter-based
28/10/18	14:49	17:49	180	16.66	2,999	4	Filter-based
29/10/18	09:42	12:42	180	16.66	2,999	4	Filter-based
29/10/18 (PM ₁₀ and PM _i)	13:58	16:58	180	16.66	2,999	4	Filter-based
30/10/18	06:00	09:00	180	16.66	2,999	4	Filter-based
30/10/18	09:37	12:37	180	16.66	2,999	4	Filter-based
30/10/18	13:13	16:13	180	16.66	2,999	4	Filter-based
31/10/18	09:38	12:38	180	16.66	2,999	4	Filter-based
31/10/18	14:28	17:28	180	16.66	2,999	4	Filter-based
01/11/18	09:47	12:47	180	16.66	2,999	4	Filter-based

01/11/18	14:55	17:55	180	16.66	2,999	4	Filter-based
02/11/18	10:24	13:24	180	16.66	2,999	4	Filter-based
02/11/18	14:36	17:36	180	16.66	2,999	4	Filter-based
03/11/18	09:34	12:34	180	16.66	2,999	4	Filter-based
03/11/18	14:08	17:08	180	16.66	2,999	4	Filter-based
<u>24 h filter (PQ100) samples</u>							
25/10/18 to 26/10/18	11:45 (25/10/18)	11:45 (26/10/18)	1,440	16.66	23,990	4	Filter-based
26/10/18 to 27/10/18	11:58 (26/10/18)	11:58 (27/10/18)	1,440	16.66	23,990	4	Filter-based
27/10/18 to 28/10/18	12:20 (27/10/18)	12:20 (28/10/18)	1,440	16.66	23,990	4	Filter-based
28/10/18 to 29/10/18	11:58 (28/10/18)	05:29 (29/10/18)	1,051	16.66	17,510	4	Filter-based
<u>Impinger (Coriolis) samples</u>							
26/10/18	(1) 11:21 (2) 11:37 (3) 11:50 (4) 12:08 (5) 12:19 (6) 12:30	(1) 11:31 (2) 11:47 (3) 12:00 (4) 12:18 (5) 12:29 (6) 12:40	60	100	6,000	6.64	Impinger-based
27/10/18	(1) 15:36 (2) 15:53	(1) 15:46 (2) 16:03	20	300	6,000	9.61	Impinger-based
28/10/18	(1) 15:00 (2) 15:19	(1) 15:10 (2) 15:29	20	300	6,000	4.73	Impinger-based
29/10/18	(1) 15:32 (2) 15:48	(1) 15:42 (2) 15:58	20	300	6,000	4.87	Impinger-based
30/10/18	(1) 16:11 (2) 16:25	(1) 16:21 (2) 16:35	20	300	6,000	4.80	Impinger-based
02/11/18	(1) 15:16 (2) 15:38	(1) 15:26 (2) 15:48	20	300	6,000	5.91	Impinger-based
03/11/18	(1) 15:09 (2) 15:23	(1) 15:19 (2) 15:33	20	300	6,000	4.99	Impinger-based

Table S2: Details of particle concentrations of the aerosol samples, including particle number concentration (dN), mass concentration of particulate matter less than 10 µm in diameter (PM₁₀), and particle surface area (dS). Data for the filter-based samples (see Table S1) are for PM₁₀ since the PQ100 filter sampler used a PM₁₀ inlet head. The dN and dS concentrations were calculated from a combination of the GRIMM Model 1.109 optical particle counter (OPC; 0.325-10 µm particle diameter) and the TSI Model 3938 scanning mobility particle sizer (SMPS) spectrometer (0.0128–0.315 µm) measurements, with the SMPS volume-equivalent particle diameters calculated assuming a dynamic shape factor (χ) of 1.1 ± 0.1 . PM₁₀ mass concentration data was obtained from an Israeli Ministry of Environment (IME) monitoring station located ~1 km from the sampling site. All times are local, and dates are in the DD/MM/YY format.

Date (DD/MM/YY)	Start time	End time	Sample designation	Average particle concentration, dN (cm ⁻³)	Average PM ₁₀ mass concentration, IME station (µg m ⁻³)	Average particle surface area concentration, dS (µm ² cm ⁻³)	Air mass category and ID
<u>3 h filter (PQ100) samples</u>							
25/10/18	10:10	11:10	181025 morning	9,014 ± 1,465	183 ± 18	745 ± 401	SW _D
25/10/18	15:07	18:10	181025 afternoon	12,960 ± 2,687	201 ± 43	815 ± 241	SW _D
26/10/18	12:09	15:17	181026 morning	4,345 ± 7,565	19 ± 12	94 ± 63	NW1
26/10/18	15:40	18:40	181026 afternoon	4,273 ± 700	1 ± 3	109 ± 21	NW1
27/10/18	10:20	13:20	181027 morning	9,175 ± 2,474	28 ± 10	84 ± 25	NW2
27/10/18	15:18	18:18	181027 afternoon	9,707 ± 883	10 ± 7	166 ± 51	NW2
28/10/18	09:28	12:28	181028 morning	12,623 ± 2,562	39 ± 8	213 ± 61	E _D 1
28/10/18	14:49	17:49	181028 afternoon	15,483 ± 583	54 ± 11	278 ± 69	E _D 1
29/10/18	09:42	12:42	181029 morning	5,865 ± 1,517	65 ± 10	267 ± 51	E _D 2
29/10/18 (PM ₁₀ and PM ₁)	13:58	16:58	181029 afternoon	8,744 ± 2,990	64 ± 12	311 ± 55	E _D 2

30/10/18	06:00	09:00	181030 early morning	136 ± 8	90 ± 10	348 ± 21	Ed3
30/10/18	09:37	12:37	181030 morning	4,173 ± 1,535	96 ± 9	349 ± 25	Ed3
30/10/18	13:13	16:13	181030 afternoon	5,459 ± 610	84 ± 9	303 ± 40	Ed3
31/10/18	09:38	12:38	181031 morning	4,590 ± 421	65 ± 11	255 ± 27	Ed4
31/10/18	14:28	17:28	181031 afternoon	5,883 ± 766	64 ± 16	309 ± 54	Ed4
01/11/18	09:47	12:47	181101 morning	6,429 ± 2,104	39 ± 10	217 ± 59	E1
01/11/18	14:55	17:55	181101 afternoon	8,017 ± 2125	40 ± 8	283 ± 88	E1
02/11/18	10:24	13:24	181102 morning	10,287 ± 909	33 ± 11	218 ± 138	E2
02/11/18	14:36	17:36	181102 afternoon	10,368 ± 964	25 ± 7	240 ± 49	E2
03/11/18	09:34	12:34	181103 morning	9,596 ± 4,437	48 ± 7	246 ± 62	E3
03/11/18	14:08	17:08	181103 afternoon	18,196 ± 1,984	31 ± 8	267 ± 55	E3
<u>24 h filter (PQ100) samples</u>							
25/10/18 to 26/10/18	11:45 (25/10/18)	11:45 (26/10/18)	181025 to 181026 (24 h)	-	-	-	SW _D /NW1
26/10/18 to 27/10/18	11:58 (26/10/18)	11:58 (27/10/18)	181026 to 181027 (24 h)	-	-	-	NW1/NW2
27/10/18 to 28/10/18	12:20 (27/10/18)	12:20 (28/10/18)	181027 to 181028 (24 h)	-	-	-	NW2/Ed1
28/10/18 to 29/10/18	11:58 (28/10/18)	05:29 (29/10/18)	181028 to 181029 (17.5 h)	-	-	-	Ed2/Ed3

<u>Impinger (Coriolis) samples</u>							
26/10/18	11:21	12:40	181026 morning (impinger)	2,281 ± 1013	27 ± 8	92 ± 24	NW1
27/10/18	15:36	16:03	181027 afternoon (impinger)	9,004 ± 714	15 ± 6	143 ± 36	NW2
28/10/18	15:00	15:29	181028 afternoon (impinger)	15,265 ± 392	48 ± 10	249 ± 32	Ed1
29/10/18	15:32	15:58	181028 afternoon (impinger)	7,740 ± 635	72 ± 2	331 ± 25	Ed2
30/10/18	16:11	16:35	181030 afternoon (impinger)	5,930 ± 800	93 ± 1	371 ± 61	Ed3
02/11/18	15:16	15:48	181102 afternoon (impinger)	10,684 ± 716	26 ± 5	238 ± 43	E2
03/11/18	15:09	15:33	181102 afternoon (impinger)	18,573 ± 543	29 ± 2	247 ± 45	E3

S2 Size-resolved biological analysis

Table S3: List of the quantitative Polymerase Chain Reaction (qPCR) primers used for the determination of total bacterial and total fungal airborne concentrations. 16S ribosomal RNA was used for bacterial analysis, while the nuclear ribosomal internal transcribed spacer (ITS) region was used for fungal analysis.

Target gene	Primers	Sequence	Reference
16S ribosomal RNA (bacteria)	331F	TCCTACGGGAGGCAGCAGT	Bräuer et al., 2011
	518R	ATTACCGCGGCTGCTGG	
ITS (fungi)	ITS1F	CTTGGTCATTTAGAGGAAGTAA	White et al., 1990
	ITS2R	GCTGCGTTCTTCATCGATGC	

S3 Fraction frozen curves for blank measurements

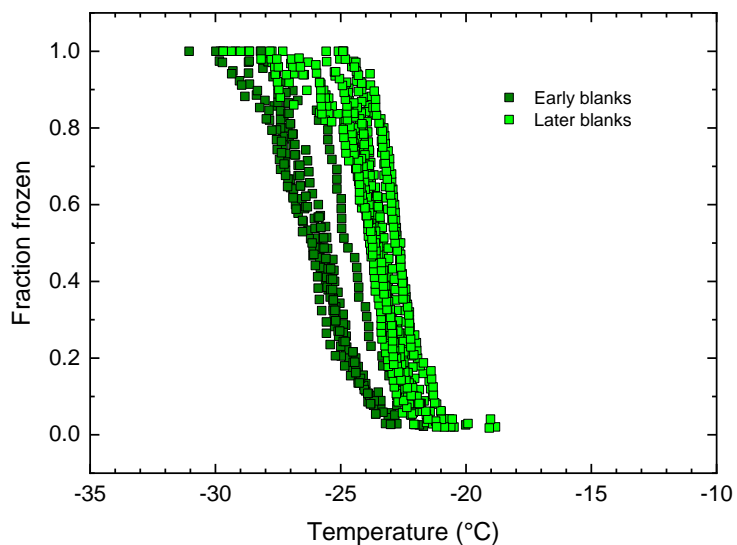


Figure S1: Plot showing the fraction frozen curves of all of the purified water blanks analysed during the field campaign in the Eastern Mediterranean using the Microlitre Nucleation by Immersed Particle Instrument ($\mu\text{L-NIPI}$) droplet freezing assay. The blanks are separated into two categories since the quality of the blanks changed suddenly during the campaign. “Early blanks” refers to those measured from 25/10/18 (DD/MM/YY) to 30/10/18. “Later blanks” refers to those measured between 31/10/18 and 04/11/18. The later blanks froze at slightly warmer temperatures than the early blanks.

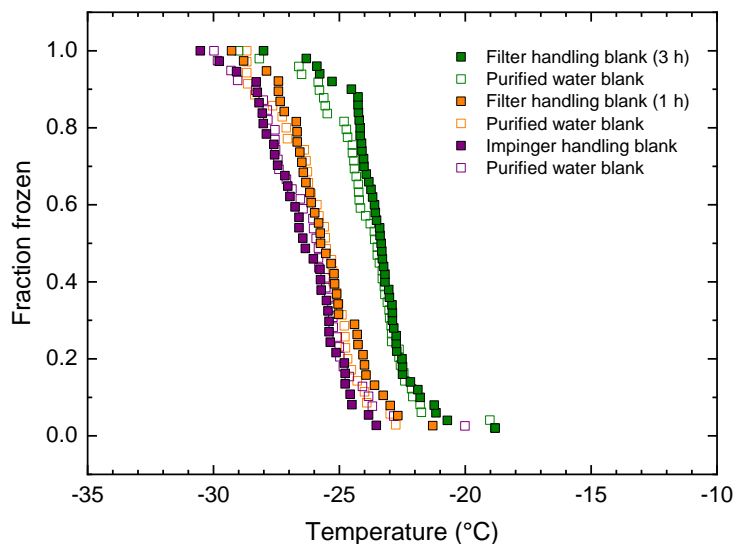


Figure S2: Fraction frozen curves showing the handling blanks from the BGI PQ100 filter system and the Coriolis® Micro impinger system versus purified water blanks that used the same water as in the handling blanks. Filter handling blanks were collected by sampling air through a HEPA (high-efficiency particulate air) filter and onto a polycarbonate track-etched membrane filter (1.0 μm pore size, 47 mm diameter, Whatman® Nuclepore™) using a Mesa Labs BGI PQ100 ambient air sampler. Two membrane filters were collected for handling blank tests: one for 1 hour of sampling through the HEPA filter (collected on 25/10/18, so compared to one of the “early” blanks), and one for 3 hours of sampling through the HEPA filter (collected on 03/11/18, so compared to one of the blanks in the “later” set). The filters were then washed with water, which was subsequently analysed using the μL -NIPI droplet freezing assay technique. The Coriolis impinger handling blank (collected on 27/10/18, so compared to one of the “early” blanks) was obtained by filling a sampling cone with water, attaching it to the impinger, then removing it and analysing the water via the μL -NIPI; no air was sampled into the cone due to the shape of the impinger inlet making it difficult to form a seal with a HEPA filter. The results show no notable differences between the handling blanks and the purified water blanks.

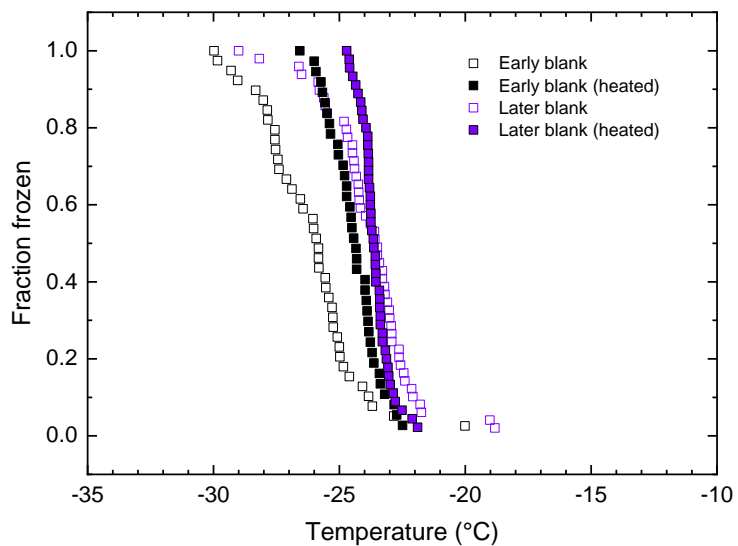


Figure S3: Plot showing the effect of heating on purified water blanks as a control for the sample heating tests. Examples are shown for an “early” blank (27/10/18) and a “later” blank (03/11/18) due to the change in characteristics of the blanks partway through the campaign. The heated “later” sample was similar to the “later” non-heated blank, while the heated “early” blank froze at warmer temperatures than the “early” non-heated blank, being more similar to the “later” blanks.

S4 Fraction frozen curves for aerosol samples

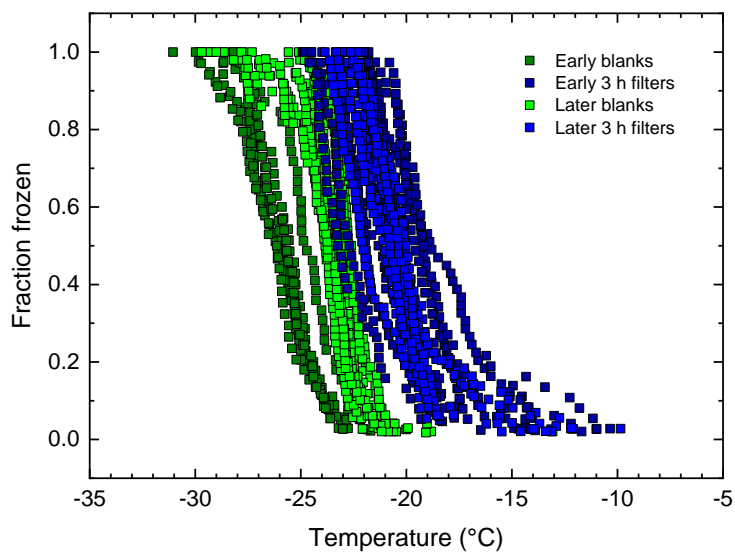


Figure S4: Fraction frozen curves for all of the aerosol samples collected onto filters for 3 h via the BGI PQ100 ambient air sampler and analysed using the $\mu\text{L-NIPI}$ droplet freezing assay. The blanks and samples are separated into categories of “early” (25/10/18 to 30/10/18) and “later” (31/10/18 to 04/11/18) due to the quality of the blanks changing on 31/10/18, hence the samples collected during one time period can be compared correctly with the blanks from that same period.

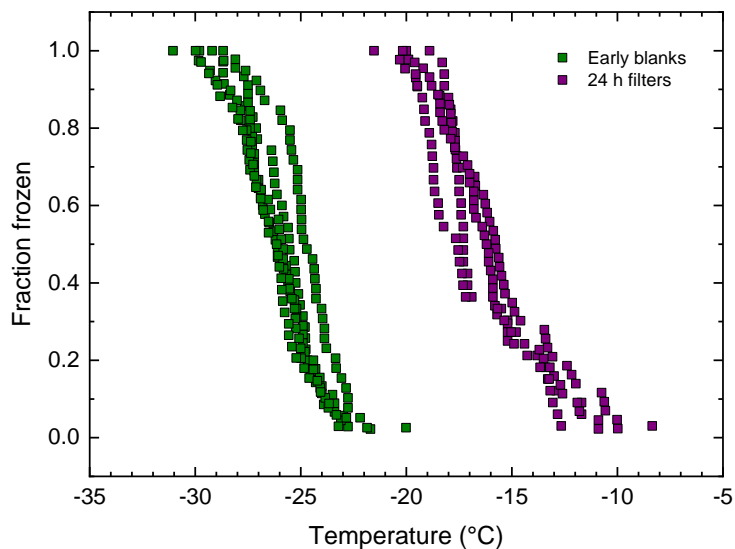


Figure S5: Fraction frozen curves for all 24 h filter-based samples collected using the BGI PQ100 ambient air sampler and analysed using the μL -NIPI droplet freezing assay. The 24 h samples were only collected prior to the change in quality of the purified water blanks partway through the campaign, hence only the “early” blanks are shown against the aerosol samples.

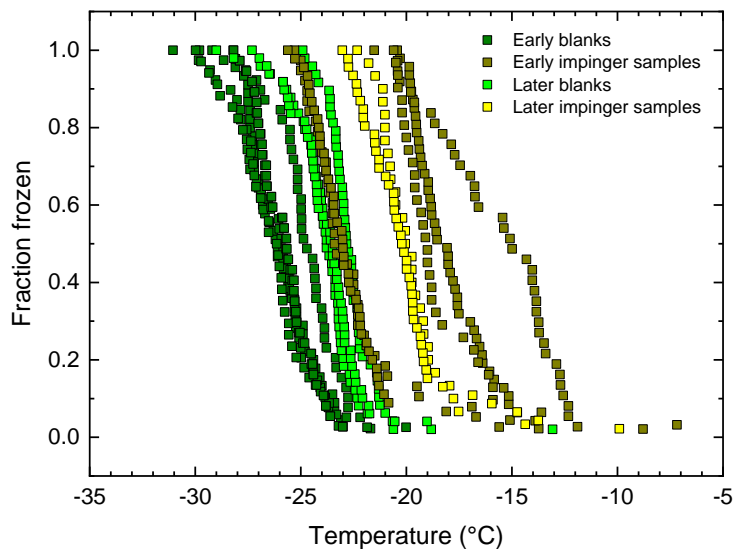


Figure S6: Fraction frozen curves for all aerosol samples collected using the Coriolis® Micro impinger system for 20-60 min and analysed using the μL -NIPI droplet freezing assay. The blanks and samples are separated into categories of “early” (25/10/18 to 30/10/18) and “later” (31/10/18 to 04/11/18) due to the quality of the blanks changing on 31/10/18, hence the samples collected during one time period can be compared correctly with the blanks from that same period.

S5 Fraction frozen curves for heat treated aerosol samples

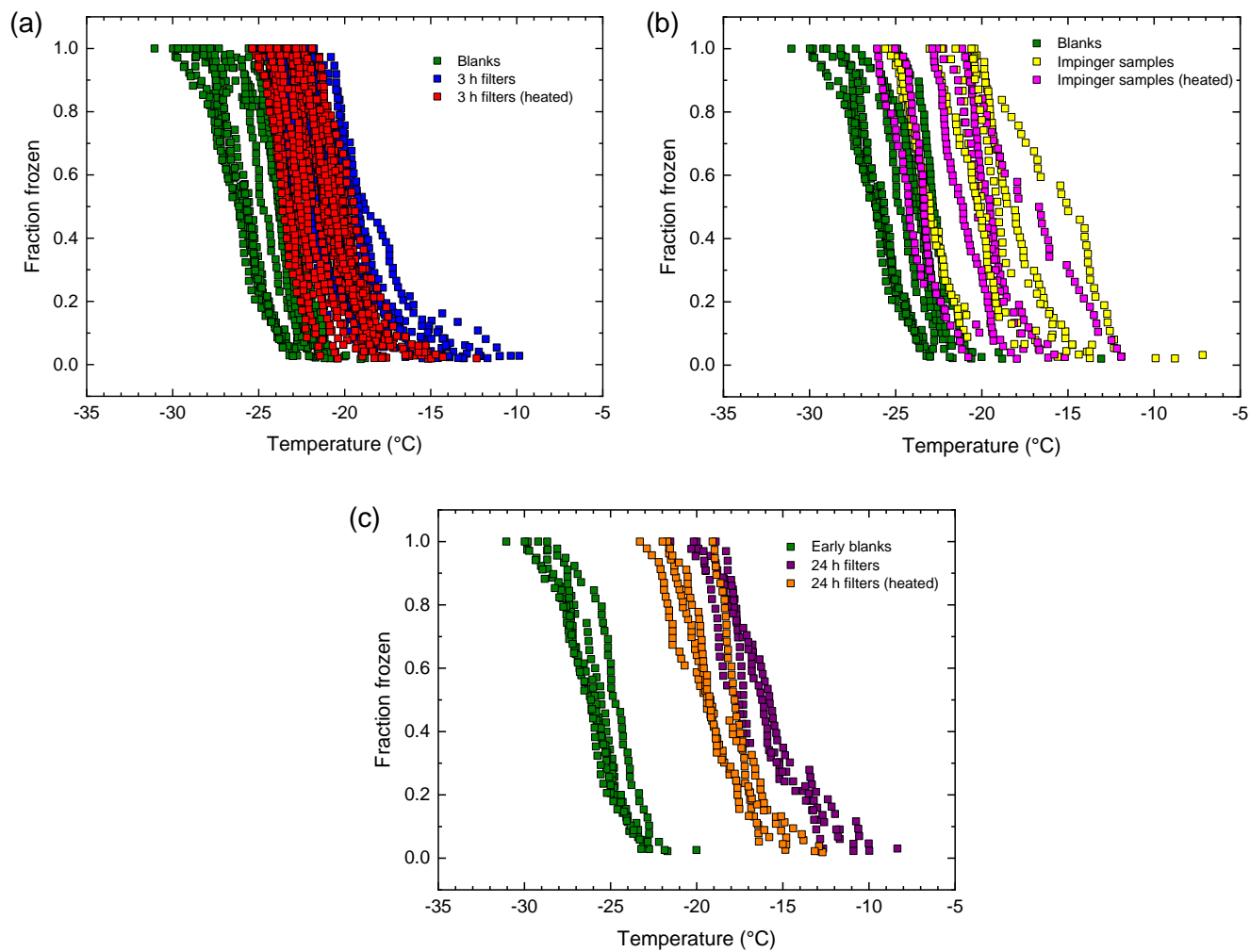


Figure S7: Fraction frozen curves for heat treated and untreated aerosol samples (a) 3 h filter samples, (b) Coriolis impinger samples, and (c) 24 h filter samples.

S6 Background subtractions of INP spectra

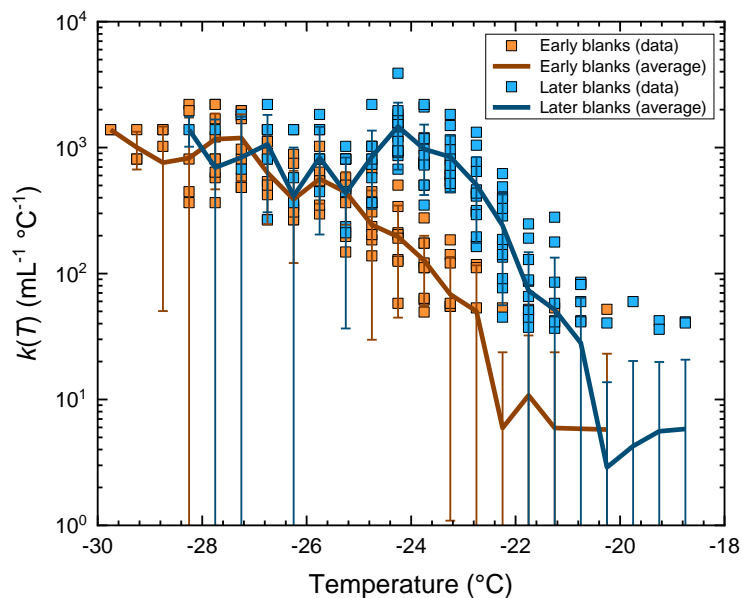


Figure S8: Blank data binned into 0.5 $^\circ\text{C}$ temperature intervals and represented in terms of the differential freezing nucleus spectrum, $k(T)$, as described by Vali (2019) and by Harrison et al. (2022). Data are shown separately for the “early” blanks (in orange) and the “later” blanks (in blue). The two sets of blanks were each averaged and these are shown represented by lines together with standard deviations. These average values were later subtracted from the ice nucleation data from the aerosol samples. During some blank runs, certain temperature bins did not have any droplets freeze within them, giving a $k(T)$ value of zero. Zeros are included in the averaging but are not shown on the logarithmic scale of the plot, hence the average being lower than some of the illustrated data and resulting in some of the error bars being large.

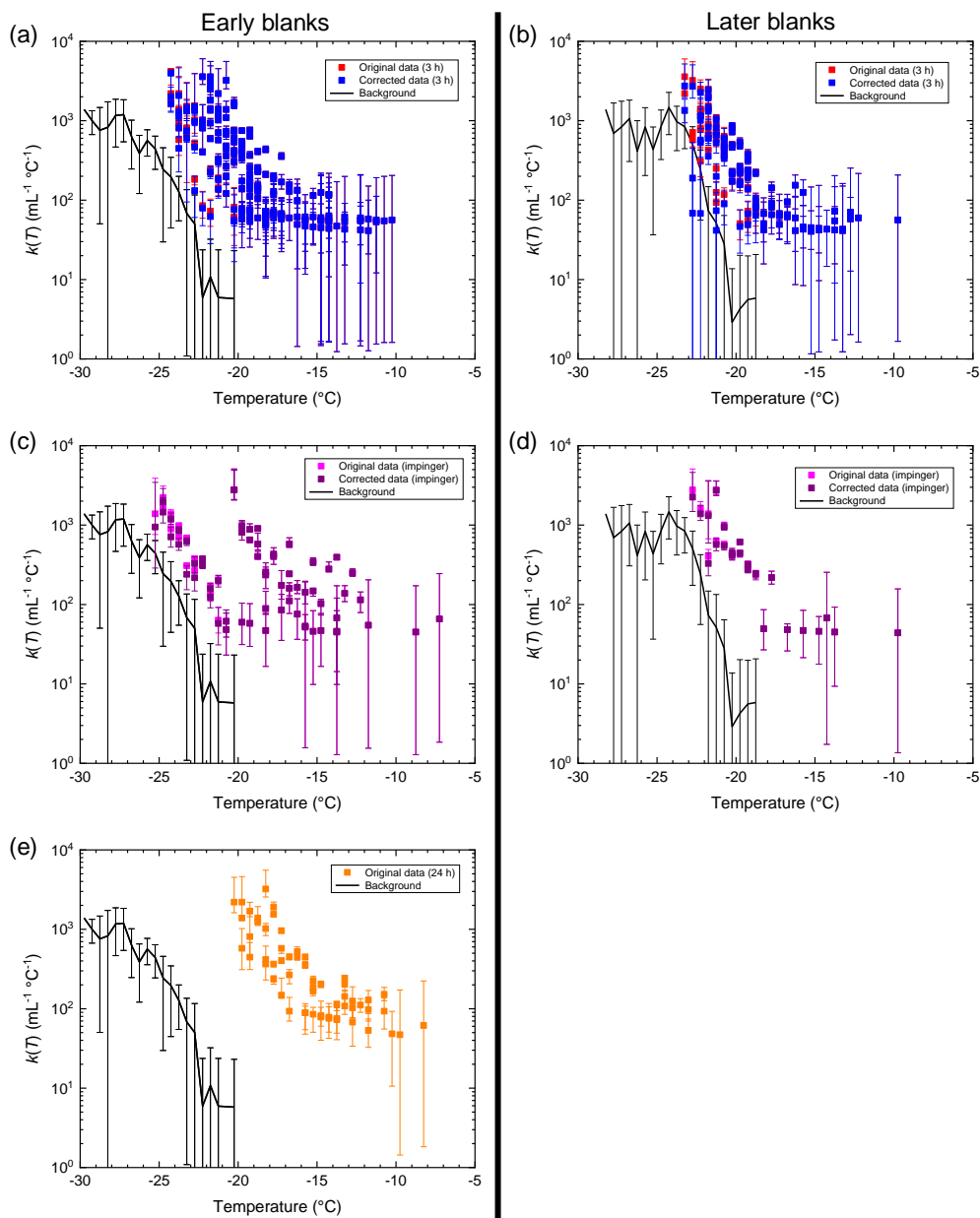


Figure S9: Sample data binned into 0.5 °C temperature intervals and represented in terms of the differential freezing nucleus spectrum, $k(T)$, as per Vali (2019) and Harrison et al. (2022). The plots show original and background-subtracted data for (a) 3 h filter samples during the “early blanks” period of time, (b) 3 h filter samples during the “later blanks” period of time, (c) impinger samples during the “early blanks” period of time, (d) impinger samples during the “later blanks” period of time, (e) 24 h filter samples during the “early blanks” period of time. Background-subtracted sample values would next be converted to the cumulative ice-active site volume density, $K(T)$, by summing the background-subtracted sample $k(T)$ values for temperatures warmer than T .

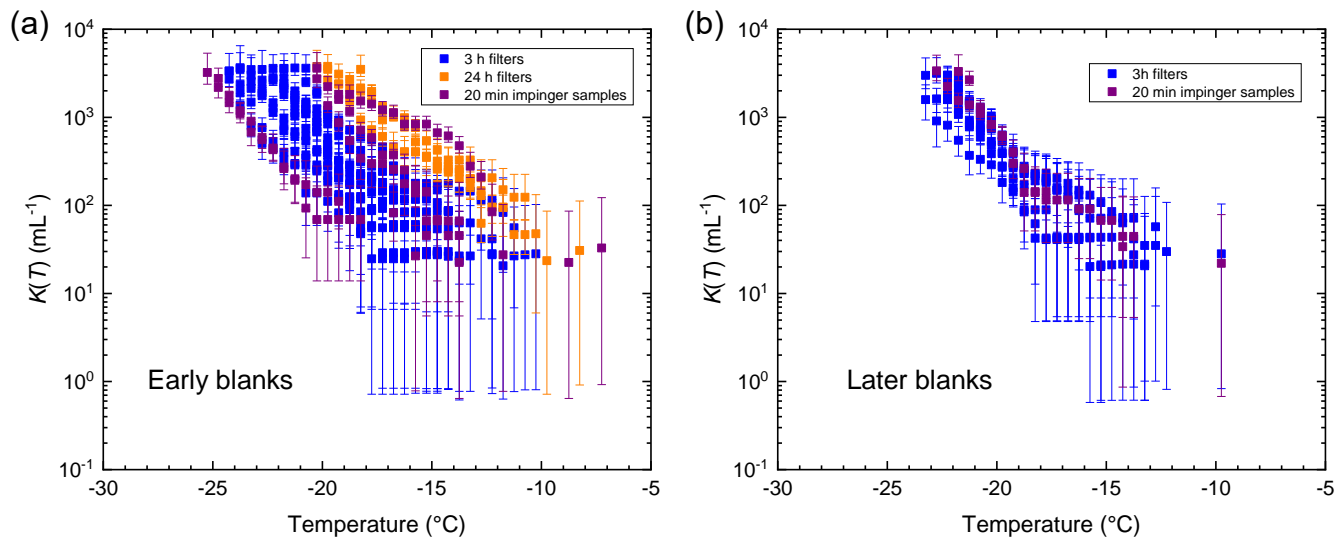


Figure S10: Plots showing background-corrected spectra for the samples in terms of the cumulative number of ice nucleation sites per unit volume of water, $K(T)$, versus freezing temperature during the time periods for (a) the “early” and (b) the “later” blanks.

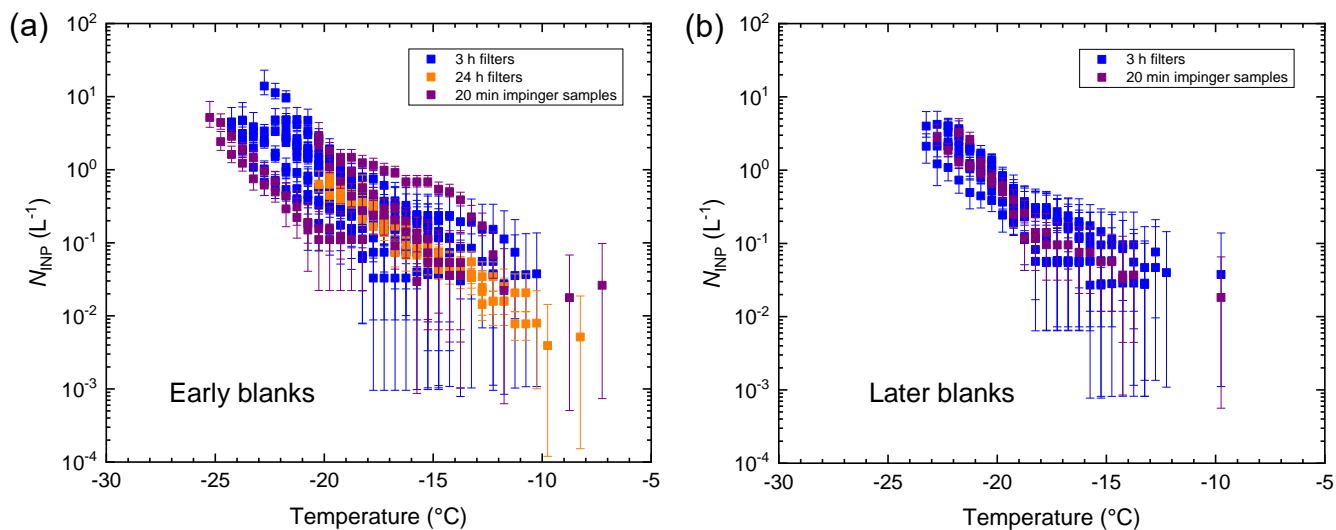


Figure S11: Plots showing the background-corrected ice-nucleating particle concentrations, N_{INP} , for all samples collected during the time periods for (a) the “early” blanks, and (b) the “later” blanks. Figure 1a in the main paper combines these plots together for the 3 h filter data, while the 24 h filter N_{INP} data and 20 min impinger N_{INP} data are shown individually below in Figure S12 and Figure S13, respectively.

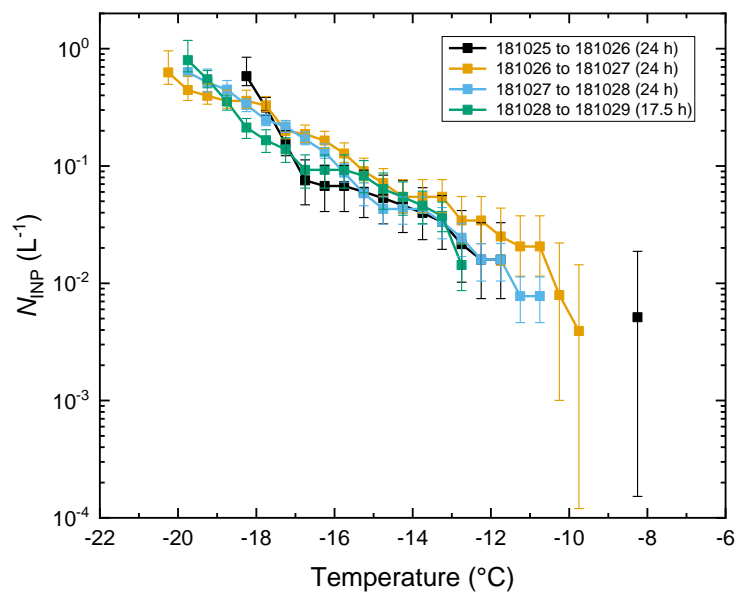


Figure S12: Plot of background-corrected ice-nucleating particle concentrations, N_{INP} , for samples collected onto filters for 24 h from 25 October to 29 October 2018, using a Mesa Labs BGI PQ100 filter sampler with a PM_{10} inlet head. Lines represent the bulk of the data, with single droplets that froze at much warmer temperature shown as individual, unconnected data points. Dates are given in the YYMMDD format.

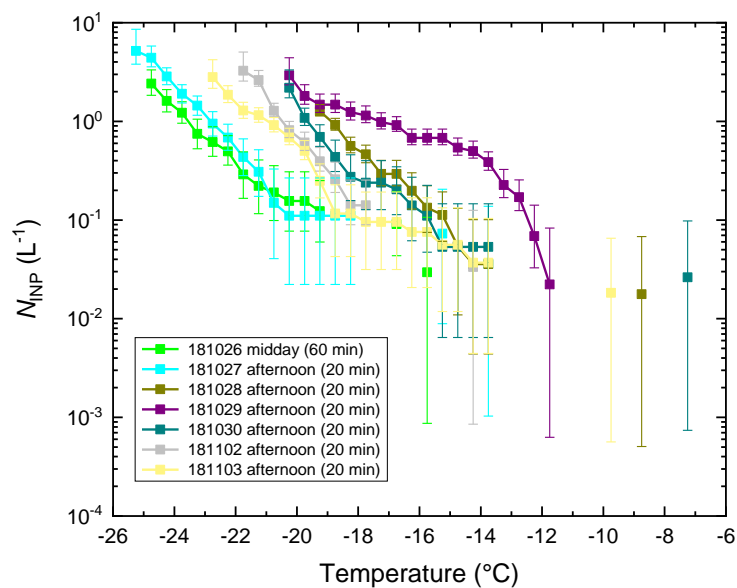


Figure S13: Plot of background-corrected ice-nucleating particle concentrations, N_{INP} , for samples collected using a Bertin Technologies Coriolis® Micro impinger for 20-40 min into <10 mL of purified water from 25 October to 3 November 2018. Lines and symbols are colour-coded to match those corresponding to the same timeframes (i.e. date and morning/afternoon) for the 3 h filter samples shown in Figure 1a in the main paper. Lines represent the bulk of the data, with single droplets that froze at much warmer temperature shown as individual, unconnected data points. Dates are given in the YYMMDD format.

S7 Particle size distributions

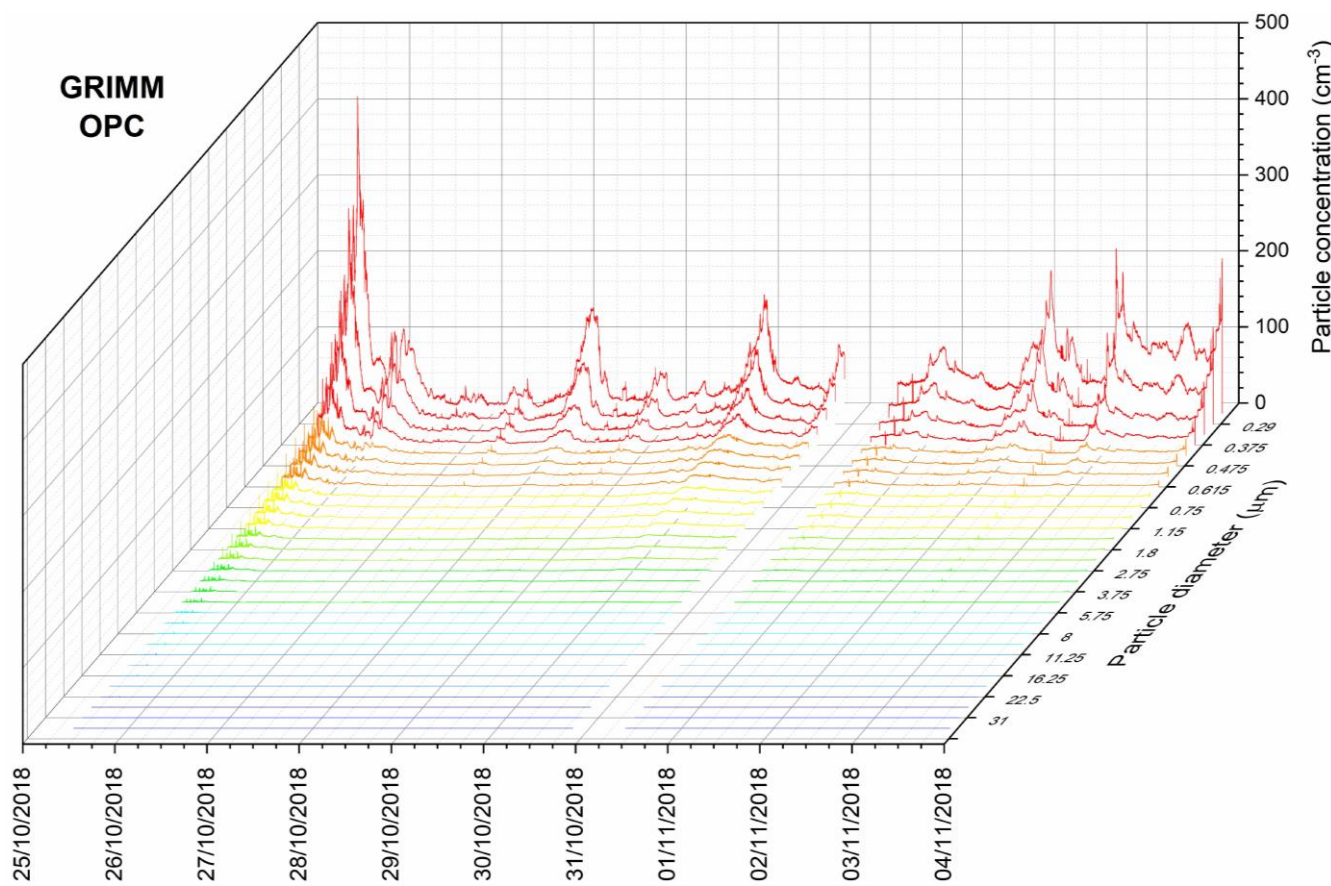


Figure S14: Time series showing the raw particle concentration (dN) data from throughout the campaign as measured by the GRIMM Model 1.109 optical particle counter (OPC; 0.25 – 32 μm particle diameter).

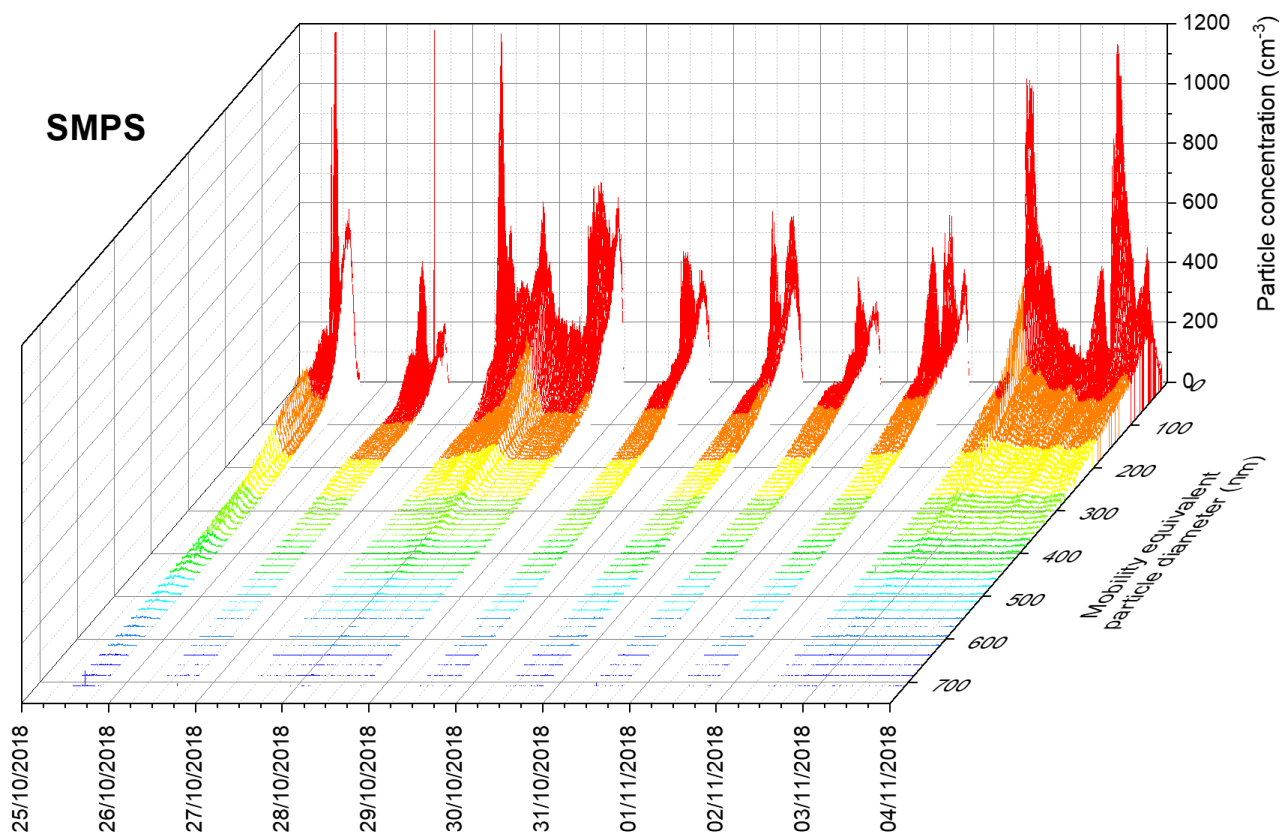


Figure S15: Time series showing the raw particle concentration (dN) data from throughout the campaign as measured using the TSI Model 3938 scanning mobility particle sizer (SMPS) spectrometer (14.1 – 710.5 nm mobility equivalent particle diameter, d_{me}).

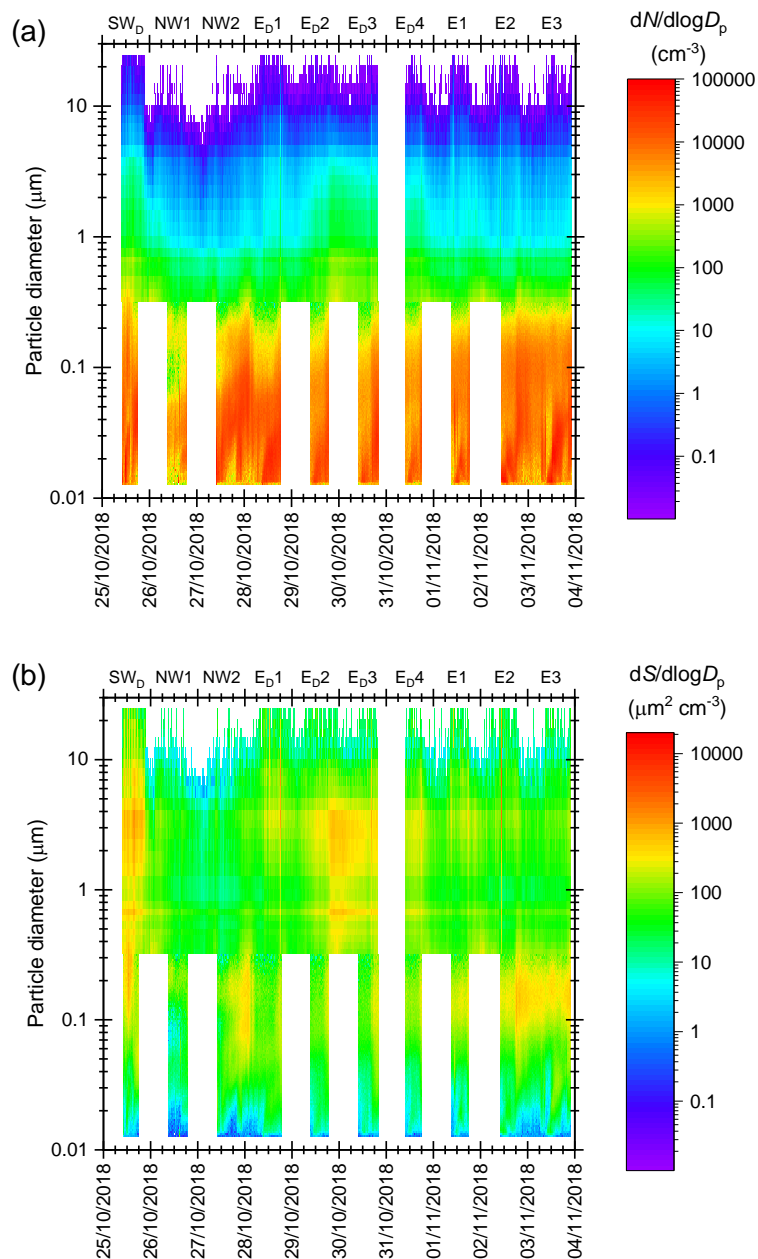
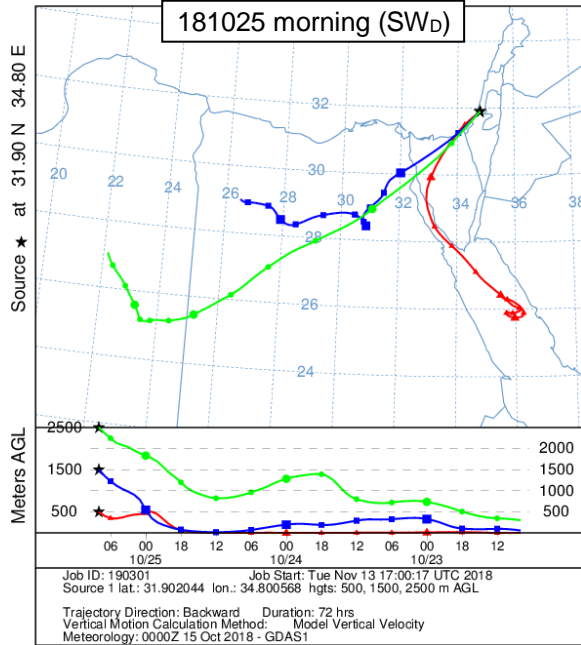


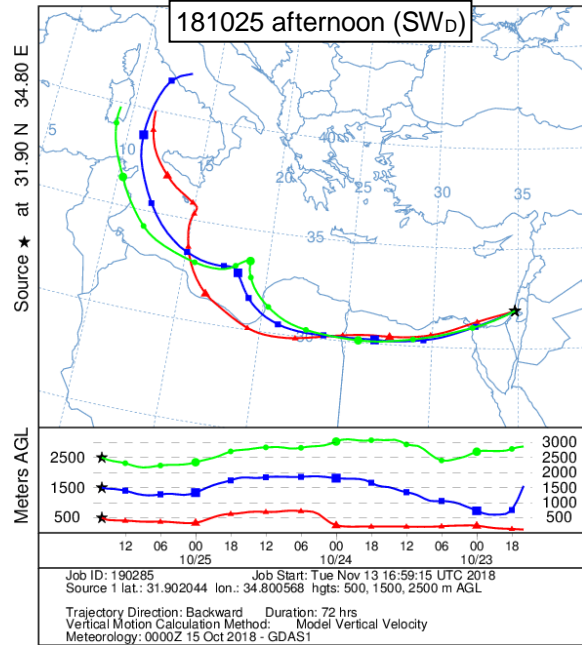
Figure S16: Time series showing particle size distributions throughout the campaign using the GRIMM OPC (0.325 – 31 μm) and the SMPS (0.0128 – 0.315 μm , $\chi = 1.1$). The gaps in the SMPS data are due to the instrument being turned off each night, with measurements only taken during the day. (a) Particle diameter (D_p) versus normalised particle concentration (dN) in terms of $dN/d\log D_p$. The OPC data shown here is the same as shown in Figure 2h in the main paper. (b) Particle diameter (D_p) versus normalised particle surface area (dS) in terms of $dS/d\log D_p$. These values were used to calculate the ice-active surface site density, $n_s(T)$, of the INP populations. Air categories and IDs are given at the top of the plots.

S8 Air mass back trajectories

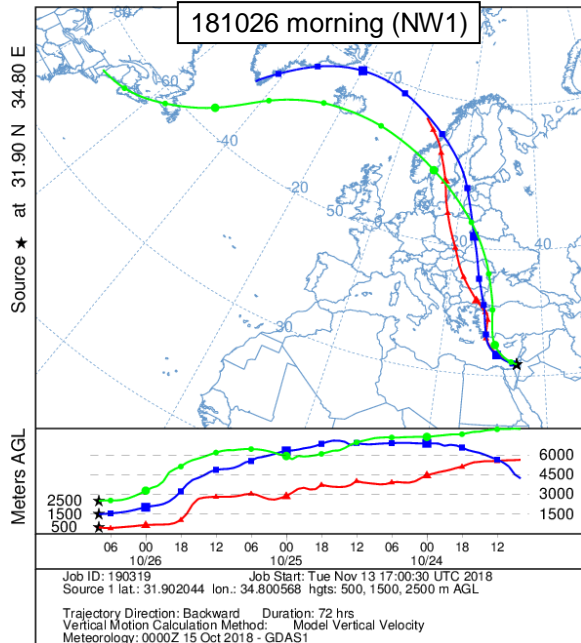
NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 25 Oct 18
GDAS Meteorological Data



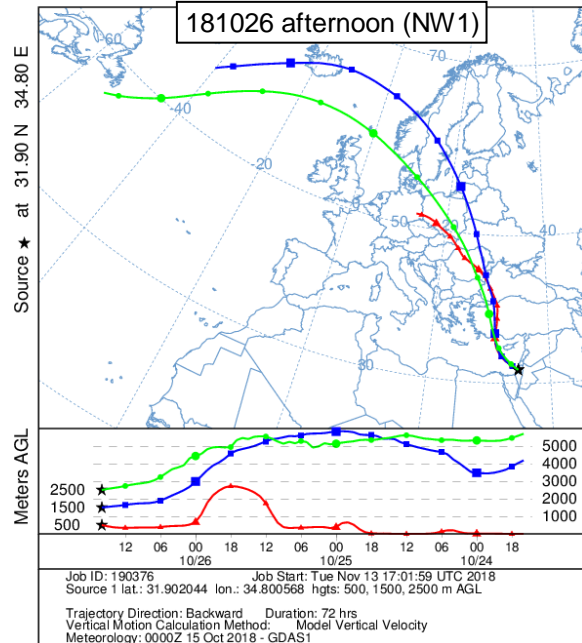
NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 25 Oct 18
GDAS Meteorological Data



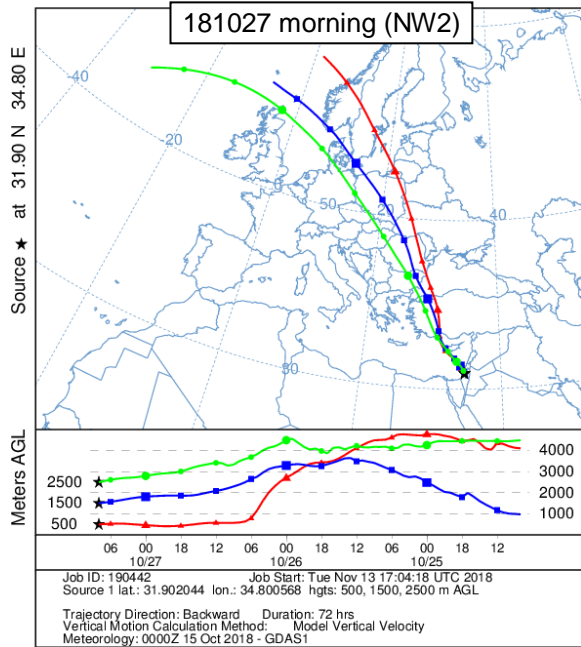
NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 26 Oct 18
GDAS Meteorological Data



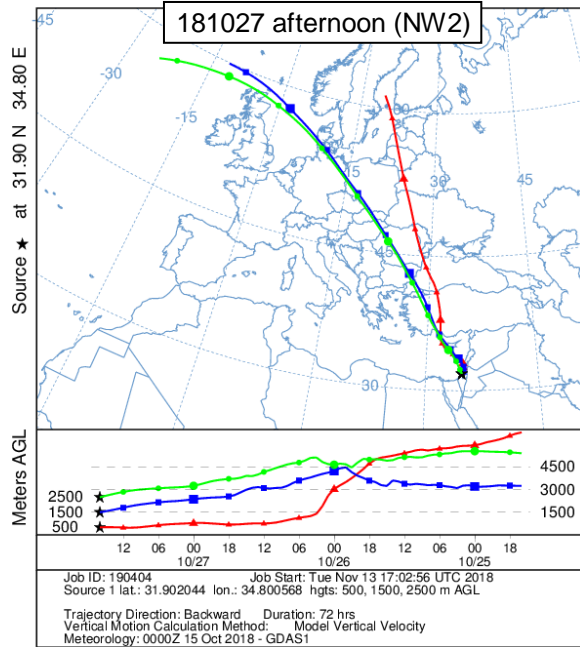
NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 26 Oct 18
GDAS Meteorological Data



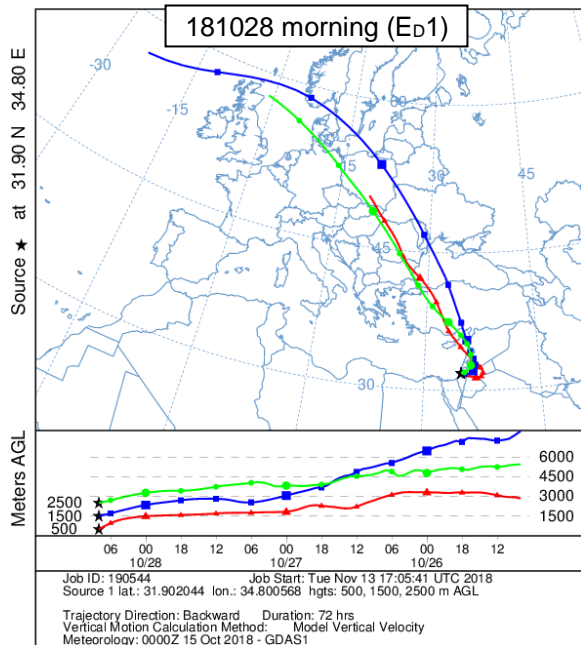
NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 27 Oct 18
GDAS Meteorological Data



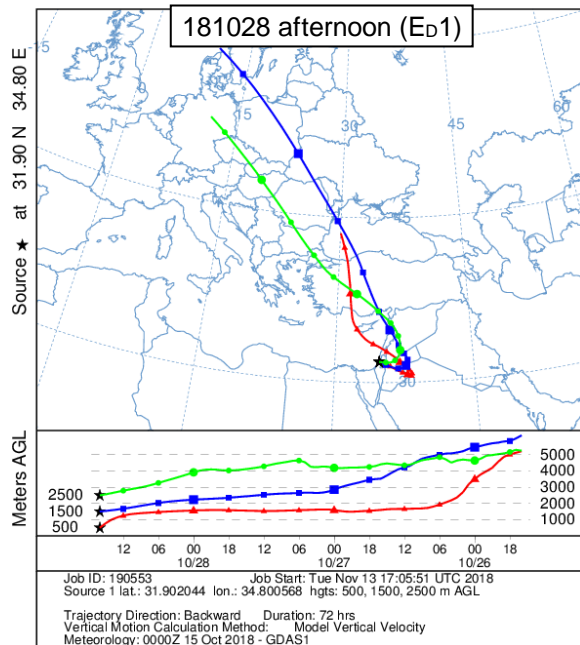
NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 27 Oct 18
GDAS Meteorological Data



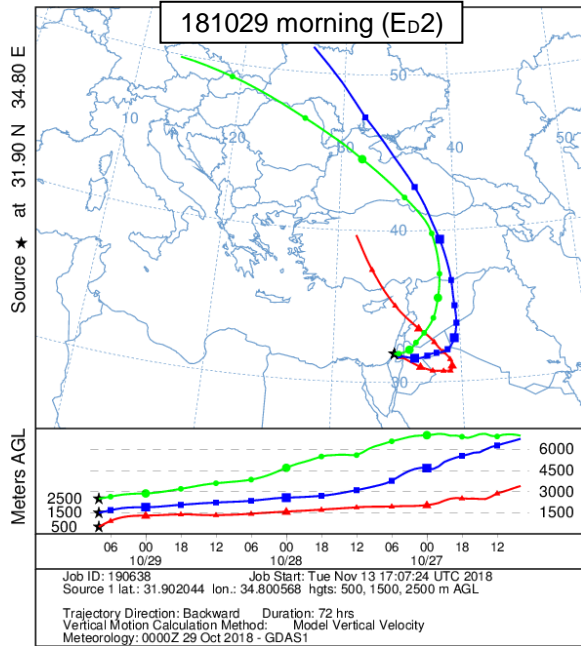
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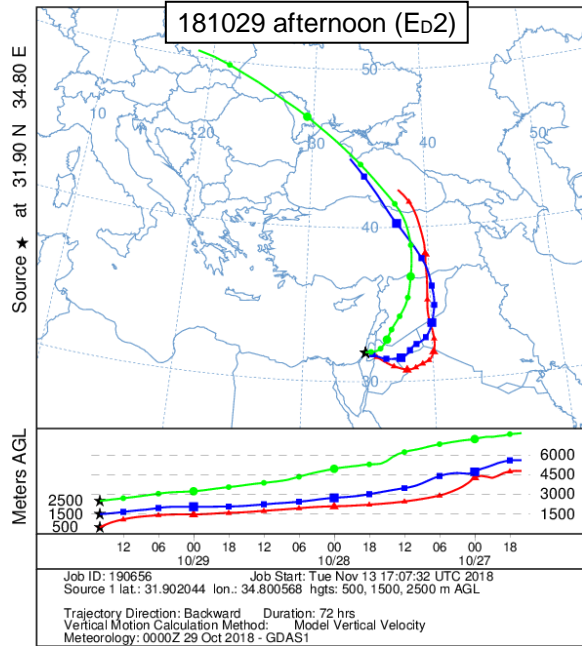
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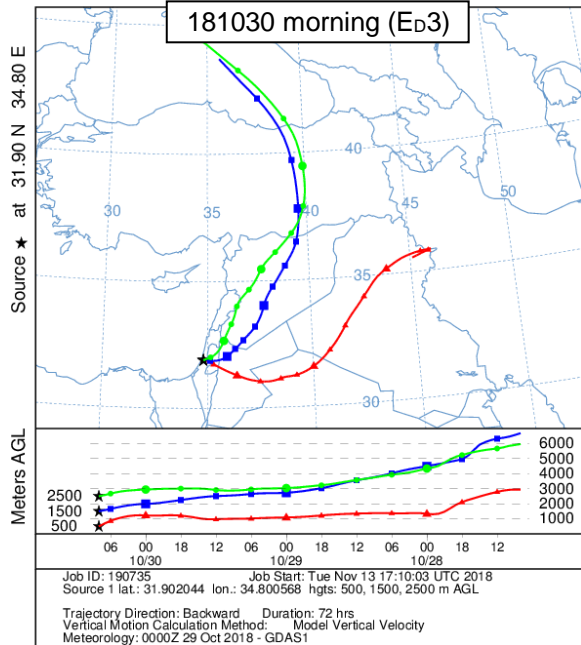
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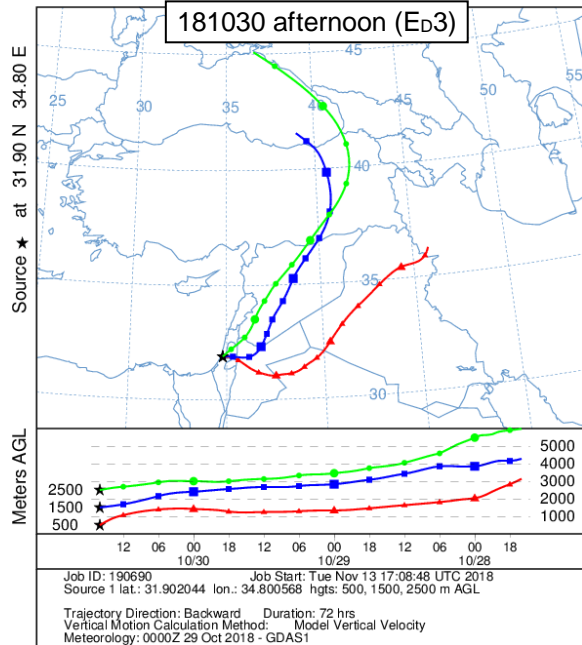
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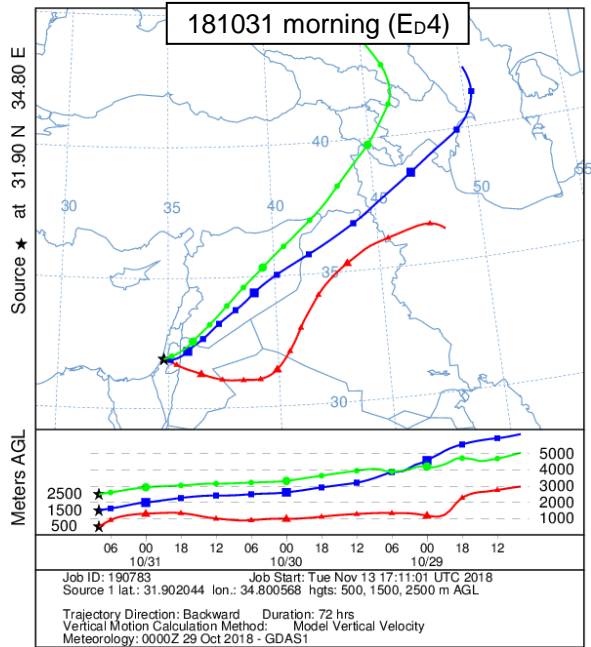
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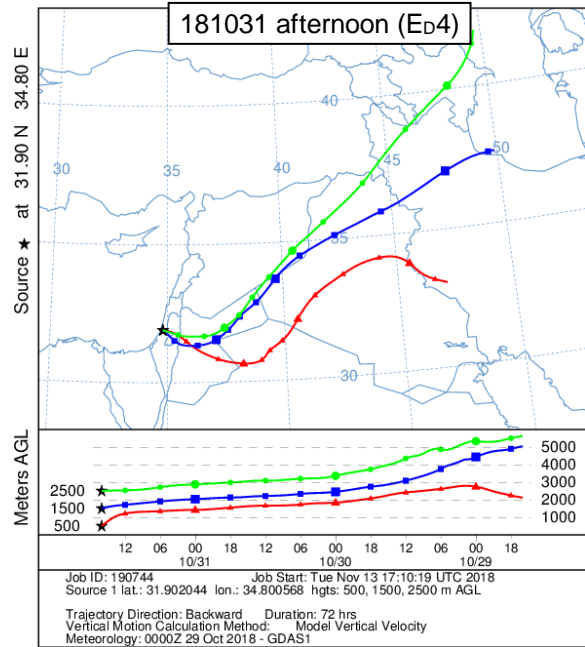
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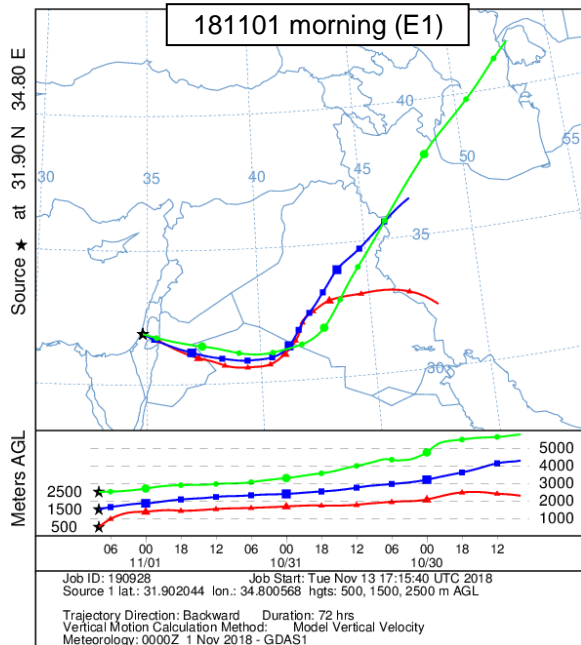
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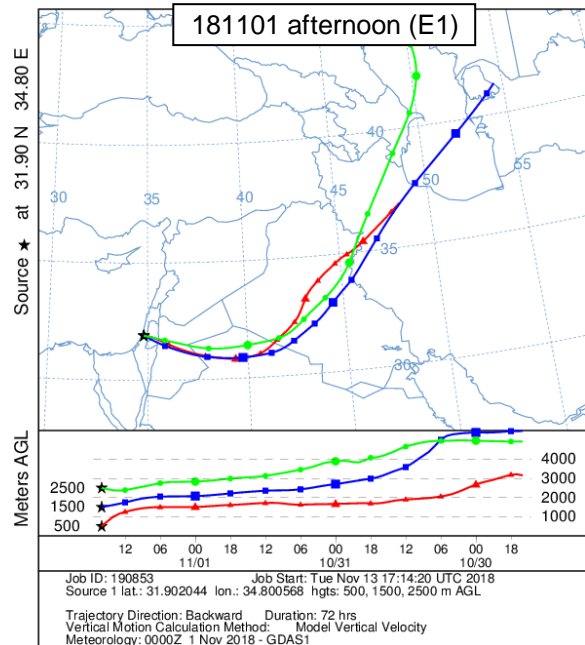
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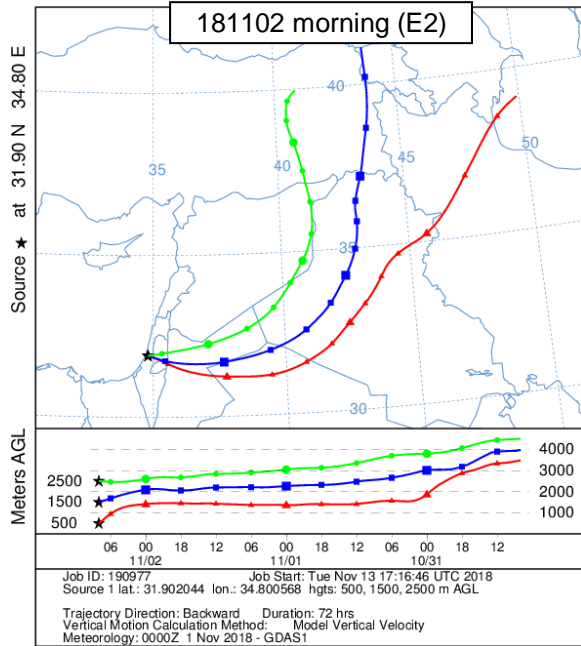
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Backward trajectories ending at 0800 UTC 01 Nov 18
GDAS Meteorological Data



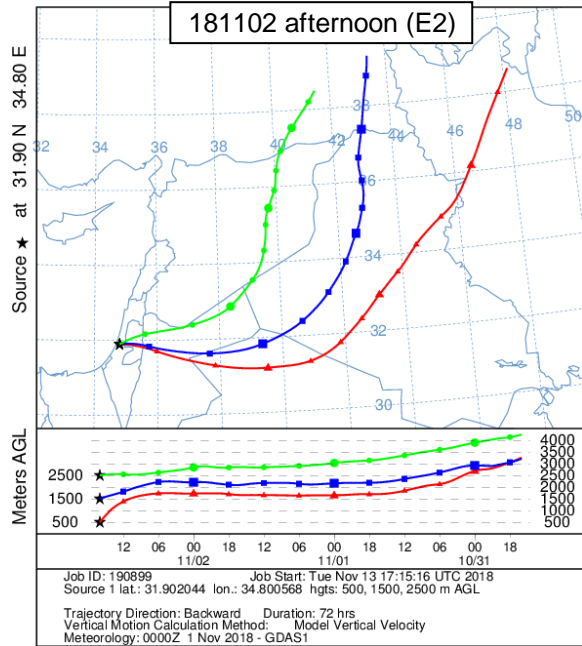
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Backward trajectories ending at 1600 UTC 01 Nov 18
GDAS Meteorological Data



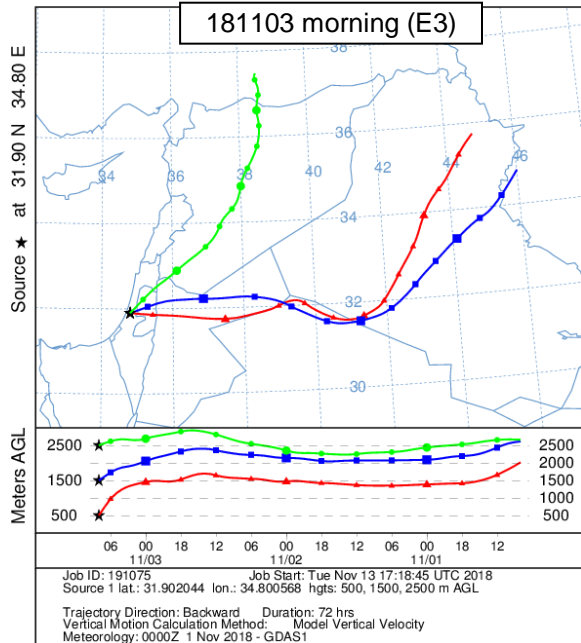
NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 02 Nov 18
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 02 Nov 18
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 03 Nov 18
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 03 Nov 18
GDAS Meteorological Data

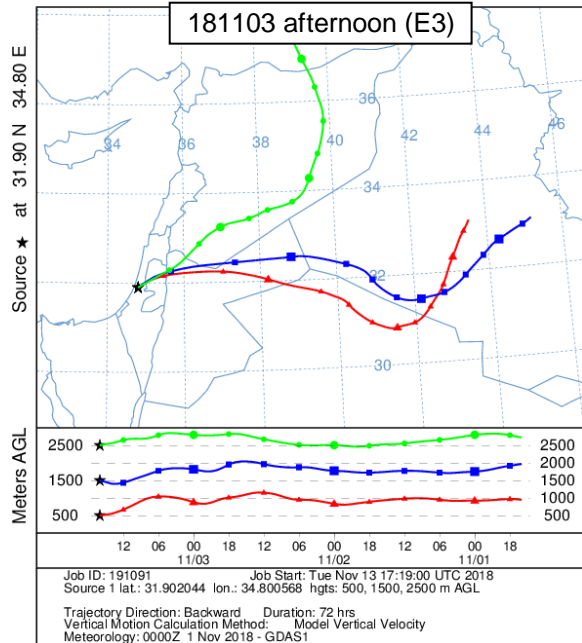


Figure S17: Air mass back trajectories (72 h) determined using the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model (<https://www.ready.noaa.gov>) (Stein et al., 2016; Rolph et al., 2017) throughout the campaign. The left column represents air masses reaching Rehovot, Israel, (represented by a star) in the mornings while the right column represents air masses reaching in the afternoons. Times are in UTC (Coordinated Universal Time), while the time in Israel is UTC+2, hence the end times for the air masses in the left column are 10:00 local time in Israel (08:00 UTC) and the end times in the right column are at 18:00 local time (16:00 UTC). Above ground level (AGL) altitudes of 500 m are shown in red, 1,500 m are shown in blue, and 2,500 m are shown in green.

S9 PM₁₀ vs. PM₁ for “181029 afternoon”

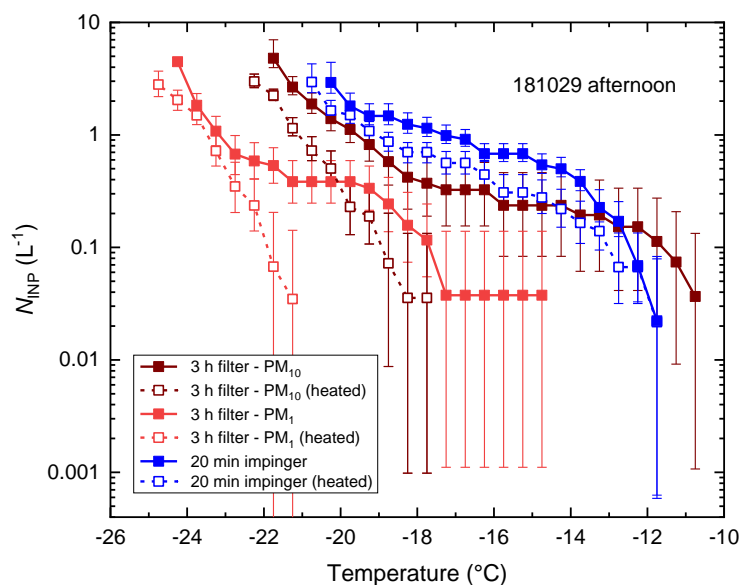


Figure S18: Background-corrected ice-nucleating particle concentrations, N_{INP} , for 3 h filter samples collected simultaneously using PM₁₀ and PM₁ inlet heads on two Mesa Labs PQ100 air sampling systems on the afternoon of 29 October 2018. A sample collected within the same time period for 20 min using a Bertin Technologies Coriolis® Micro impinger-based air sampler is also shown. All samples were subjected to heat treatment (95 $^{\circ}\text{C}$ for 30 min) to test for potential proteinaceous INPs, as per Daily et al. (2022), and those results are also shown here. The date in the figure is in the YYMMDD format.

S10 Heat treatments

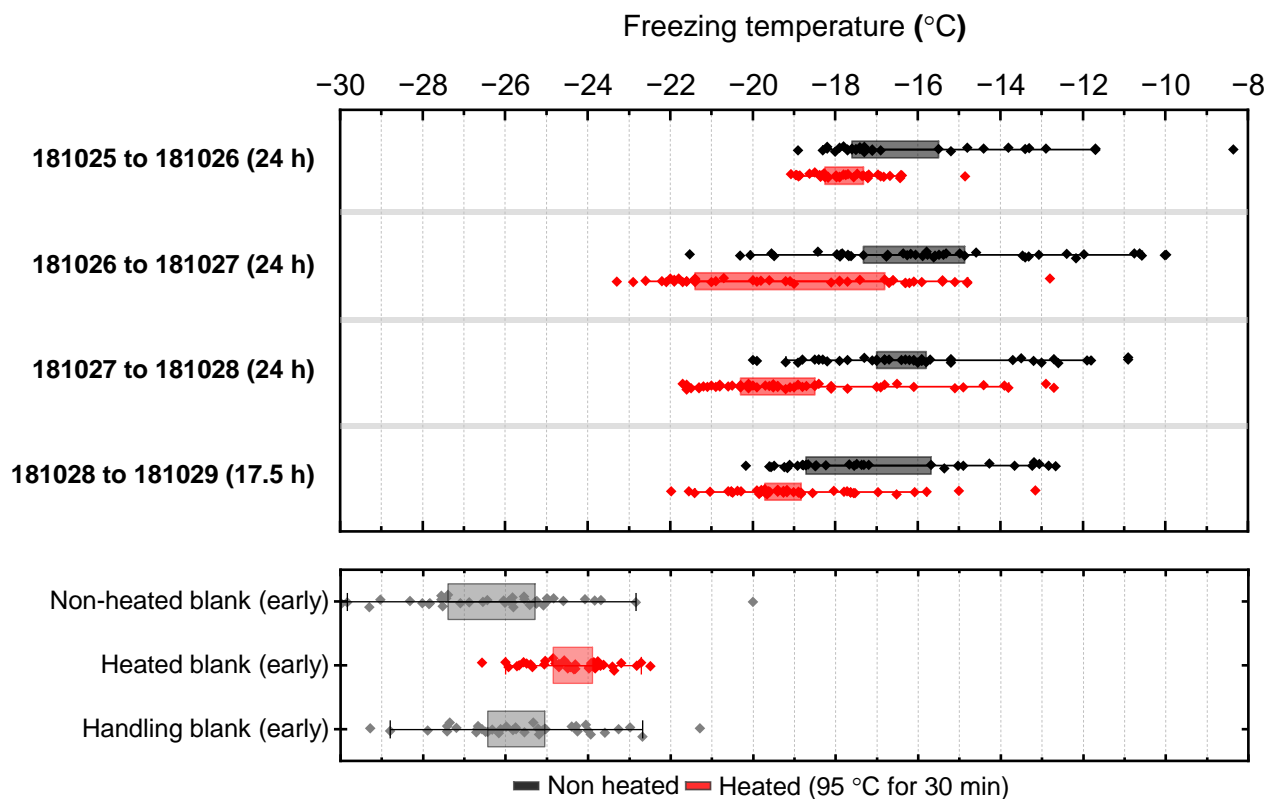


Figure S19: Box-and-whisker plots showing the effect of heat treatment (95 °C for 30 min, as per Daily et al. (2022)) on the ice-nucleating activity of aqueous particle suspensions obtained from 24 h filter samples as an indicator of potential proteinaceous INPs. Heat treatments of heated and non-heated purified water “early” blanks as control tests are provided, in addition to an “early” handling blank obtained using a HEPA filter. Boxes represent 1 standard deviation from the mean (1σ ; 68 %) while whiskers represent 2 standard deviations (2σ ; 95 %). Dates are given in the YYMMDD format.

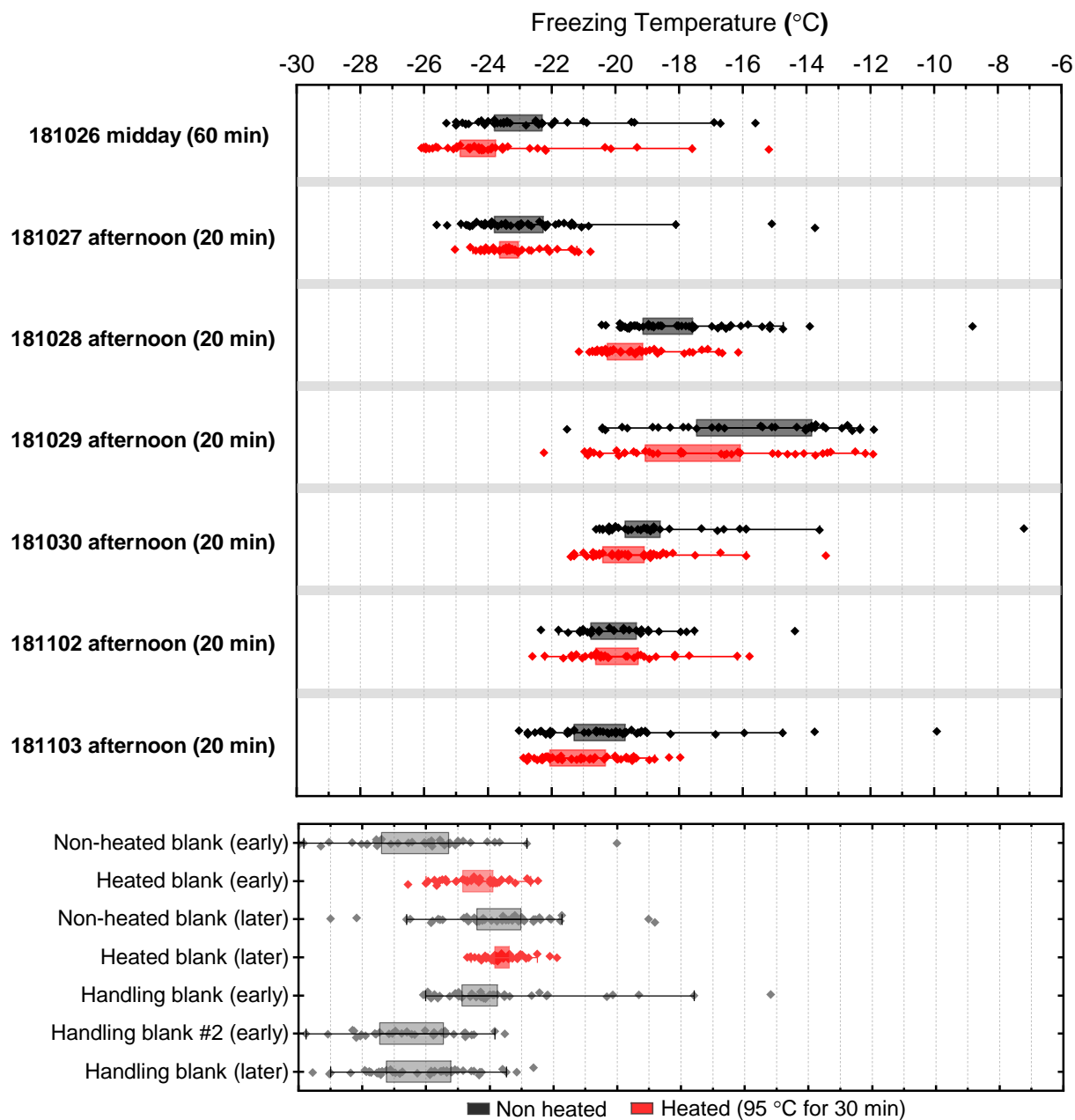


Figure S20: Box-and-whisker plots showing the effect of heat treatment (95 °C for 30 min, as per Daily et al. (2022)) on the ice-nucleating activity of aqueous particle suspensions obtained from 20-40 min impinger samples as an indicator of potential proteinaceous INPs. Heat treatments of heated and non-heated purified water blanks as control tests are provided for both “early” and “late” blanks alongside data for non-heated handling blanks. Boxes represent 1 standard deviation from the mean (1 σ ; 68 %) while whiskers represent 2 standard deviations (2 σ ; 95 %). Dates are given in the YYMMDD format.

S11 Data availability

The data sets for this paper are publicly available in the University of Leeds Data Repository (<https://doi.org/10.5518/1487>) (Tarn et al., 2024).

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