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Ice nucleating particles at a coastal marine boundary layer site: correlations with aerosol type and meteorological conditions

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S1 Measurements of CO, NO_x, and SO_2

CO (Thermo Fisher Scientific 48i-TL), NO_X (Thermo Fisher Scientific 42i) and SO_2 (Teledyne API T100U) monitors were located in the mobile laboratory operated by Environment Canada, the British Columbia Ministry of Environment, and Metro Vancouver, and sampled once a minute. Linear correlations between these trace gases and INP number concentrations are given in Tables S2 and S3.

S2 Empirical parameterizations of ice nucleation

The accuracy of six INP parameterizations from the literature was determined using data from this study. The parameterizations tested were those of Fletcher (1962), hereafter F62; Cooper (1986), hereafter C86; Meyers et al. (1992), hereafter M92; DeMott et al. (2010), hereafter D10; and two from Tobo et al. (2013), hereafter $T13_{total}$ and $T13_{fluorescent}$.

The parameterization of F62 used the following equation:

$$[\mathrm{INPs}(T)] = A\exp(-BT) \quad (1)$$

where [INPs(T)] is the number concentration of INPs in L^{-1} at temperature T in °C, and A and B are fitted constants with values of A = 0.00001 and B = 0.6. Another single-parameter function was proposed by C86 with the following form:

$$[\mathrm{INPs}(T)] = 10^{(C-D(-T))} \quad (2)$$

where E and F are fitted constants with values of C = -2.35 and D = 0.135. This parameterization was developed over a temperature range of -5 to -25 °C using measurements of ice crystal concentrations. The final single-parameter function that was evaluated is that of M92:

$$[\mathrm{INPs}(S_\mathrm{i})] = \exp(E + F[100(S_i - 1)]) \quad (3)$$

where E and F are fitted constants with values of E = -0.639 and F = 0.1296, and S_i is the ice supersaturation. This parameterization was developed using measurements between -7 to -20 °C.

D10 and T13 have recently proposed INP parameterizations to predict number concentrations of INPs as a function of both temperature and aerosol concentrations. Following work that showed a correlation between INP number concentration and aerosol particles larger than 0.5 μm (DeMott et al., 2006), D10 developed the following parameterization for mixed-phase cloud conditions:

$$[\mathrm{INPs}(T, \mathrm{AP}_{>0.5})] = a(-T)^b[\mathrm{AP}_{>0.5}]^{(-cT+d)} \quad (4)$$

where $[\mathrm{AP}_{>0.5}]$ is the number concentration of aerosol particles with diameters larger than 0.5 μm in cm^{-3}, and a, b, c, and d are fitted constants with values of a = 0.0000594, b = 3.33, c = 0.0264, and d = 0.0033. This parameterization was found to be more accurate than the earlier parameterizations of F62, C86, and M92.

T13 applied the same principle as D10 and used INP measurements from a forested site in Colorado to develop the following parameterization, $\mathrm{T13}_{\mathrm{total}}$:

$$[\mathrm{INPs}(T, \mathrm{AP}_{>0.5})] = [\mathrm{AP}_{>0.5}]^{(-\alpha T+\beta)}\exp(-\gamma T + \delta) \quad (5)$$

where α, β, γ, and δ are fitted constants with values of α = -0.074, β = 3.8, γ = 0.414, and δ = -9.671. As T13 found that INP number concentrations were strongly correlated to number concentrations of fluorescent bioparticles, they also proposed a second parameterization, $\mathrm{T13}_{\mathrm{fluorescent}}$:

$$[\mathrm{INPs}(T, \mathrm{FB}_{>0.5})] = [\mathrm{FB}_{>0.5}]^{(-\propto' T+\beta')}\exp(-\gamma' T + \delta') \quad (6)$$

where $[\mathrm{FB}_{>0.5}]$ is the number concentration of fluorescent bioparticles with diameters larger than 0.5 μm in cm^{-3}, and α', β', γ', and δ' are fitted constants with values of α' = -0.108, β' = 3.8, γ' = 0, δ' = 4.605.

S3 Size range of INPs used when comparing current results with parameterizations

The MOUDI-DFT used at the MBL site measures INP number concentrations for particles between 0.18–10 μm in size (50% cutoff aerodynamic diameter). The parameterizations of M92, D10, and T13 were formulated using INP measurements at particles sizes ≤ 3, ≤ 1.6, and ≤ 2.4 μm, respectively. To better match the size range of INPs measured in this study with those used to formulate the parameterizations of M92, D10, and T13, here we limit the measured INP data to sizes ≤ 3 μm when testing the parameterization of M92, ≤ 1.6 μm when testing the parameterization of D10, and ≤ 2.4 μm when testing the $\mathrm{T13}_{\mathrm{total}}$ and $\mathrm{T13}_{\mathrm{fluorescent}}$ parameterizations. As these sizes do not correspond to the size cut of any MOUDI stage, INP number concentrations smaller than a given particle size were calculated by the following method:

1) Identify the MOUDI stage that overlaps with the CFDC size range above. For example, the size range of ≤ 1.6 μm from D10 overlaps with the size range of MOUDI stage 5 (1.0–1.8 μm).

2) Multiply the INP number concentration found in this MOUDI stage by the fraction of the particle size range that overlaps with that of the CFDC. For example, using ≤ 1.6 μm from D10 this multiplication factor would be (1.6-1.0)/(1.8-1.0) = 0.75.

3) Add the INP number concentration found above to the total INP number concentration found on all lower MOUDI stages.

As an example using the above procedure, the INP number concentrations used for testing the parameterization of D10 were calculated using the following equation:

$$[\mathrm{INPs}(T)] = 0.75[\mathrm{INPs}(T)]_{1.0-1.8\ \mu m} + [\mathrm{INPs}(T)]_{0.18-1.0\ \mu m} \quad (7)$$

where $[\mathrm{INPs}(T)]_{1.0–1.8\mu m}$ is the number concentration of INPs 1.0–1.8 μm in size and $[\mathrm{INP}s(T)]_{0.18–1.0\mu m}$ is the number concentration of INPs 0.18–1.0 μm in size.

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Table S1. Details of the INP sampling periods. The meteorological parameters given have been averaged over the stated sampling duration.

Sample ID	Start date and time (PST)	End date and time (PST)	Sampling time (min)	Temp. (°C)	Relative Humidity (%)	Air mass category
N3	08/06 23:17:48	08/07 09:27:14	609	11.5	100	Coastal NW
D5*	08/08 11:43:23	08/08 22:36:01	645	13.7	100	Pacific Ocean
N5	08/08 23:09:19	08/09 09:46:40	637	12.6	100	Coastal NW
D6	08/09 10:52:52	08/09 19:41:50	529	12.9	99	Coastal NW
N6	08/09 23:37:17	08/10 06:06:32	389	12.1	100	Coastal NW
D7	08/10 10:16:37	08/10 18:16:00	479	12.8	100	Coastal NW
D8	08/11 10:50:02	08/11 18:50:03	480	13.3	99	Coastal NW
N8	08/11 22:15:02	08/12 06:15:02	480	12.7	100	Coastal NW
D9	08/12 11:30:02	08/12 19:30:02	480	14.2	97	Coastal NW
N9	08/12 22:00:02	08/13 06:00:02	480	13.9	98	Pacific Ocean
D10	08/13 10:52:02	08/13 18:52:02	480	13.8	97	Pacific Ocean
N10	08/13 22:00:03	08/14 06:00:03	480	15.0	91	Pacific Ocean
D11A	08/14 11:30:02	08/14 16:55:02	325	14.9	91	Coastal SE
D11B	08/14 17:16:02	08/14 22:05:30	289	14.3	98	Coastal SE
N11	08/14 22:32:02	08/15 06:32:02	480	14.0	100	Free troposphere
D12	08/15 11:05:06	08/15 19:05:07	480	14.9	97	Pacific Ocean
N13	08/16 23:01:33	08/17 07:00:33	479	14.4	100	Pacific Ocean
D16	08/19 10:30:02	08/19 18:30:02	480	15.1	94	Pacific Ocean
N16	08/19 22:00:02	08/20 06:00:03	480	13.4	96	Coastal NW
D17*	08/20 11:10:02	08/20 19:09:22	443	14.7	91	Coastal NW
N17	08/21 00:17:37	08/21 06:04:36	347	13.2	96	Coastal NW
D18*	08/21 10:50:02	08/21 18:49:54	445	15.7	89	Free troposphere
N18	08/21 22:02:02	08/22 06:02:02	480	13.4	100	Free troposphere
D19*	08/22 10:42:02	08/22 18:41:52	438	15.0	97	Free troposphere
N19	08/22 22:00:02	08/23 06:00:02	480	13.9	100	Pacific Ocean
D20*	08/23 10:15:02	08/23 18:14:25	462	13.9	100	Pacific Ocean
N20	08/23 22:00:02	08/24 06:00:02	480	13.5	100	Pacific Ocean
D21	08/24 11:40:02	08/24 19:40:02	480	13.8	99	Pacific Ocean
N21	08/24 22:00:02	08/25 06:00:03	480	13.2	100	Pacific Ocean
D22B	08/25 14:26:03	08/25 20:26:03	360	15.2	88	Pacific Ocean
N22	08/25 22:00:03	08/26 06:00:03	480	13.7	92	Pacific Ocean
D23	08/26 10:35:03	08/26 18:35:03	480	14.2	90	Coastal SE
N23	08/26 22:00:02	08/27 06:00:02	480	13.3	100	Coastal SE
D24*	08/27 11:22:02	08/27 17:57:43	367	14.2	99	Coastal SE

*Sampling was not continuous

Table S2. Correlation coefficients (R) for linear regression analyses of INPs versus CO, NO_x, and SO_2. No correlations had statistical significance ($P < 0.05$).

	Relation to the INP number concentration											
	-15 °C			-20 °C			-25 °C			-30 °C		
Measurement	R	P[a]	n[b]	R	P	n	R	P	n	R	P	n
CO	0.14	0.22	34	0.24	0.08	34	0.28	0.06	34	0.32	0.05	27
NO_x	-0.11	0.27	34	-0.25	0.08	34	-0.27	0.06	34	-0.06	0.38	27
SO_2	0.07	0.34	34	0.05	0.39	34	0.07	0.34	34	0.13	0.27	27

[a]The P-value is a conditional probability that is the probability of obtaining an R-value equal to or greater than the given R-value if there is no correlation between INPs and the given parameter.
[b]n represents the number of data points used in determining the correlation.

Table S3. Correlation coefficients (R) for linear regression analyses of INPs versus CO, NO_x, and SO_2 within category of air mass. Correlations with statistical significance ($P < 0.05$) have been underlined.

		Relation to the INP number concentration											
		-15 °C			-20 °C			-25 °C			-30 °C		
Air Mass	Measurement	R	P[b]	n[c]	R	P	n	R	P	n	R	P	n
Coastal NW	CO	<u>0.55</u>	0.04	11	0.52	0.05	11	<u>0.55</u>	0.04	11	0.42	0.17	7
	NO_x	0.01	0.49	11	-0.32	0.17	11	-0.29	0.19	11	-0.04	0.46	7
	SO_2	<u>0.62</u>	0.02	11	0.26	0.22	11	0.26	0.22	11	<u>0.76</u>	0.02	7
Coastal SE	CO	-0.59	0.15	5	-0.38	0.26	5	0.14	0.41	5	0.76	0.23	3
	NO_x	0.15	0.40	5	0.35	0.28	5	0.70	0.10	5	-0.05	0.48	3
	SO_2	-0.36	0.28	5	-0.26	0.34	5	0.12	0.42	5	-0.38	0.38	3
Pacific Ocean	CO	-0.03	0.46	14	0.07	0.41	14	0.04	0.45	14	<u>0.50</u>	0.04	13
	NO_x	-0.19	0.26	14	-0.23	0.22	14	-0.28	0.16	14	0.18	0.28	13
	SO_2	-0.21	0.24	14	-0.19	0.26	14	-0.32	0.13	14	0.05	0.44	13
Free troposphere	CO	0.74	0.13	4	<u>0.92</u>	0.04	4	<u>0.93</u>	0.03	4	<u>0.92</u>	0.04	4
	NO_x	-0.73	0.13	4	-0.74	0.13	4	-0.70	0.15	4	-0.67	0.17	4
	SO_2	0.08	0.46	4	0.24	0.38	4	0.30	0.35	4	0.33	0.34	4

[a]The P-value is a conditional probability that is the probability of obtaining an R-value equal to or greater than the given R-value if there is no correlation between INPs and the given parameter.
[b]n represents the number of data points used in determining the correlation.

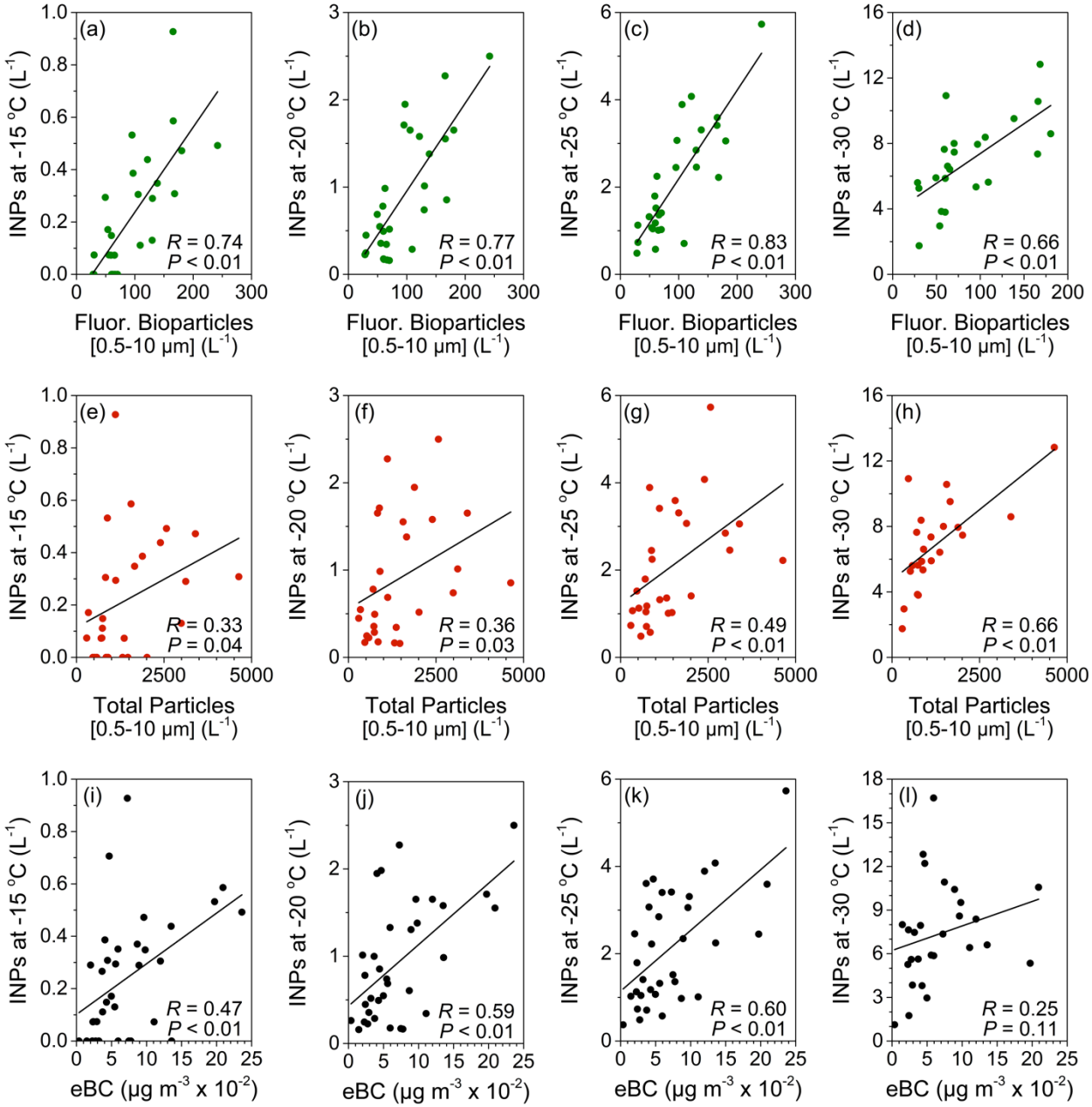

Figure S1. INP number concentrations from -15 to -30 °C (columns I–IV) plotted against total particle number concentrations (panels a–d), fluorescent bioparticle number concentrations (panels e–h), and eBC mass concentrations (panels i–l). Linear fits are given as solid lines with corresponding correlation coefficients (*R*) and probability values (*P*).

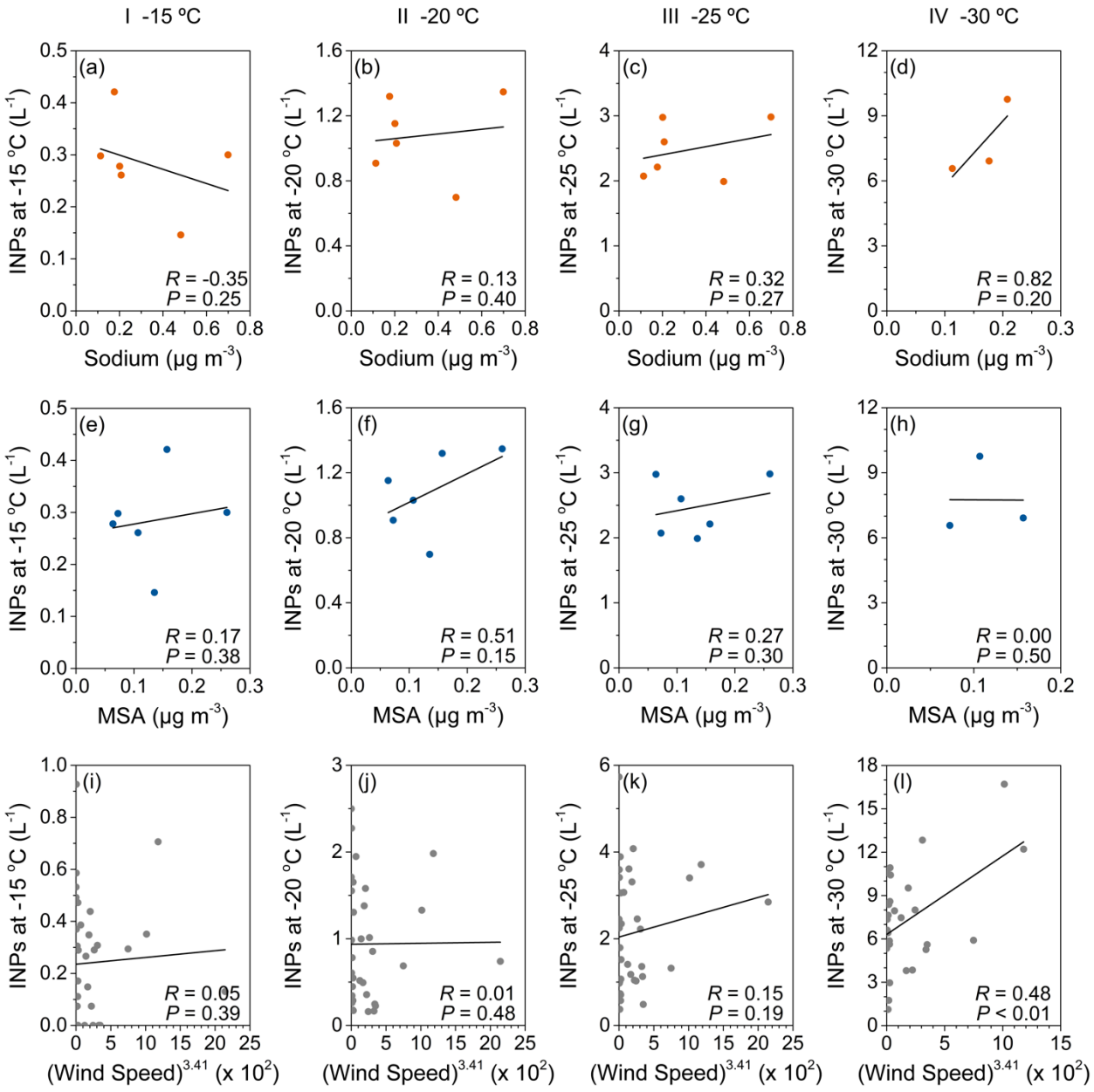

Figure S2. INP number concentrations from -15 to -30 °C (columns I–IV) plotted against sodium mass concentrations (panels a–d), MSA mass concentrations (panels e–h), and (wind speed)$^{3.41}$ based on the power law dependence of whitecap coverage on wind speed by Monahan and Muircheartaigh (1980) with wind speed in m s^{-1} (panels i–l). Linear fits are given as solid lines with corresponding correlation coefficients (R) and probability values (P).