



## Supplement of

## Long-term variability in immersion-mode marine ice-nucleating particles from climate model simulations and observations

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D15	Aerosol property	Study area	Sample type	LIIIIIIauolis
	Number concentrations of dust	Saharan and	Mineral dust particles	Does not distinguish between
	particles with diameters greater than 500 nm $[m^{-3}]$	Asian deserts	larger than 0.5 $\mu$ m	different dust minerologies.
W15	Total organic carbon mass con-	Arctic (July-	Marine Organic	Does not include the differ-
	centration $[gm^{-3}]$	August 2013)	Aerosol (MOA)	ences in the
				emission and atmospheric
				chemistry of INPs
				between the airborne sea-
				spray (SSA) and sea surface
				microlayer water samples ;
				Entrainment of multiple INP
				species into a single SSA
				particle is not considered.
M18	Sea spray aerosol surface area	Mace Head	Sea salt aerosol	Derived for clean marine con-
	concentration $[m^{-2}m^{-3}]$	station (August		ditions. Only background sea-
		2015)		spray INPs are captured.
M18+D15	Sea spray aerosol surface area	Mace Head	Dust and Sea salt	Represents only background
	concentration $\left[\mathrm{m}^{-2}\mathrm{m}^{-3}\right]$ and	(August 2015)		INPs. Potential variations in
	number concentrations of dust	and South-		marine INPs due to ocean biol-
	particles [m <sup>-3</sup> ] with diameters	ern Ocean		ogy or dust mineralogy are not
	greater than 500 nm	(March-April,		captured.
		2016)		

Table S1. Descriptions of aerosol aware INP parameterizations



**Figure S1.** Climatology of dust concentrations from ground stations in the Northern Hemisphere compared against E3SM aerosol climatology. For the model, monthly average dust concentrations are shown for the period 2016–2018. Error bars in the model represent standard deviation of aerosol mass concentrations for 2016–2018. Error bars in the observations for each ground station represent standard deviation of measurements. Both CTL and EXP are shown for comparison with observations.



**Figure S2.** Climatology of sea salt concentrations from ground stations in the Northern Hemisphere compared against E3SM aerosol climatology. For the model, monthly average sea salt concentrations are shown for the period 2016–2018. Error bars in the model represent standard deviation of aerosol mass concentrations for 2016–2018. Error bars in the observations for each ground station represent standard deviation of measurements. Both CTL and EXP are shown for comparison with observations.



Figure S3. Differences in ratios of CNT and D15 INP parameterizations between  $-20^{\circ}$ C and  $-28^{\circ}$ C



**Figure S4.** Comparison of INP dependence on temperature in CNT and D15 INP parameterizations from E3SM simulations. Panel (a) shows INPs simulated by CNT dust for temperatures  $-28.5^{\circ}$ C (black), $-28^{\circ}$ C (magenta), $-20.5^{\circ}$ C (green), $-20^{\circ}$ C (blue), $-16.5^{\circ}$ C (grey), $-16^{\circ}$ C (red). For a given temperature, different data points shown here correspond to the days during the MICRE campaign. Panel (b) shows the INP temperature spectra for D15.



**Figure S5.** (Top) Aerial photograph of Macquarie Island isthmus, with the location of MICRE and RSV Aurora Australis (on which MAR-CUS was located) indicated. (Bottom) MICRE campaign filter on Macquarie Island. Waves, kelp and tussock upwind (toward the ocean) over which air must pass before reaching the filter are also visible in the photo. Photo credits: Andrew Klekociuk, Australian Antarctic Division.



**Figure S6.** Observed INP concentrations  $(L^{-1})$  at Macquarie Island from MICRE and simulated INPs using the updated dry deposition parameterization in E3SMv1. (a) and (b) : D15, (c) and (d) : M18, (e) and (f) : M18+D15, (g) and (h) : W15. INP concentrations are colored by activation temperatures used for measurements. Solid line in each panel represents 1:1 comparison, while dashed lines represent a factor of 2 and 10 from the observations. Error metrics in each panel include fractional gross error (FGE), modified normalized mean bias (MNMB), spearman correlation (R), percentage of model INPs within a factor of two from observations (2x), and percentage of model INPs within a factor of 10 from observations (10x). Scatter plots for austral summer (October-February) are shown in the left column and those for austral winter (March - September) are shown in the right column.