



Supplement of

Marine productivity and synoptic meteorology drive summer-time variability in Southern Ocean aerosols

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Figure S1: Example 7-day back trajectories for air masses which reached the RV Investigator at hourly intervals between 0000 and 0600 (UTC) on 4 February 2015. An ensemble of 27 back trajectories were generated for each air mass and the depicted trajectories represent the mean of each ensemble. Coloured data points represent the air mass location at each hour along the back trajectory and the black diamond shows the mean ship position. The colour scale gives the altitude of the air mass relative to the estimated boundary layer, and grey data points represent hours when the air mass passed through the free troposphere. Similar back trajectory plots are available for the full voyage (1st – 15th February 2015) on request.



Figure S2: Comparison between observed sea salt mass concentrations (coloured points) and concentrations modelled using the sea salt source functions (black lines) given in Ovadnevaite et al. (2012). The dashed black line corresponds to the sea salt flux parameterisation derived from periods with increasing wind speeds. The solid black line corresponds to the sea salt flux parameterisation that was not differentiated for increasing or decreasing wind speeds. The colour scale represents the number of consecutive hours that each air mass spent within the MBL, with rainfall rates <0.25 mm h⁻¹, prior to reaching the ship. Gaps in the modelled data indicate that these conditions were not met during the hour prior to the air mass reaching the ship. Periods influenced by different air masses are labelled and delimited by dotted grey lines.



Figure S3: (a) Geometric mean diameters of aerosol in the fitted nucleation, Aitken and accumulation modes. (b) Concentrations of aerosol with diameters larger than 10 nm and CCN concentrations at 0.5% supersaturation. Periods influenced by different air masses are labelled and delimited by dashed lines.

Table S1: Physical properties observed for each sampling period corresponding to the three air masses: continental Antarctic (cAA), continental Australian (cAU), and maritime Southern Ocean (mSO). The values represent the medians, with the interquartile range provided in parentheses.

Air mass	mSO-I	cAU	mSO-II	cAA-I	mSO-III	cAA-II	mSO-IV
Latitude extents (°S)	43.3–54.0	54.2–58.8	59.3-64.0	64.0–65.1	59.9–64.1	57.5–59.9	46.1–57.5
Number concentrations							
N ₁₀ (cm ⁻³)	405 (289–498)	239 (166–259)	506 (466–586)	304 (294–328)	484 (407–646)	315 (290–328)	764 (463–863)
CCN _{0.5%} (cm ⁻³)	203 (136–236)	200 (127-225)	198 (106–294)	252 (233–258)	179 (132–195)	198 (154–211)	195 (167–215)
CCN0.5% activation ratio	0.50 (0.40–0.59)	0.79 (0.72–0.89)	0.32 (0.22–0.56)	0.80 (0.76–0.83)	0.34 (0.25–0.40)	0.65 (0.54–0.69)	0.31 (0.20–0.53)
Number size distributions							
Nucl. conc. (cm ⁻³)	93 (51–150)	13 (9.8–15)	180 (120-400)	15 (12–16)	310 (170–560)	60 (32–170)	590 (480-620)
Aitken conc. (cm ⁻³)	150 (100–230)	65 (43–77)	140 (79–230)	72 (49–81)	110 (100–140)	78 (70–83)	640 (240-820)
Accum. conc. (cm ⁻³)	160 (110–190)	170 (130–180)	130 (73–170)	210 (200–230)	130 (87–150)	160 (130–180)	140 (110–170)
Nucl. peak diameter (nm)	19 (18–22)	23 (22–25)	27 (23–29)	17 (11–21)	20 (16–21)	13 (11–15)	27 (22–28)
Aitken peak diameter (nm)	41 (37–43)	51 (45–57)	45 (44–46)	37 (36–41)	37 (35–39)	37 (35–38)	34 (32–36)
Accum. peak diameter (nm)	125 (110–138)	172 (169–181)	114 (107–124)	114 (112–118)	122 (112–131)	96 (94–102)	121 (116–126)
Nucl. spread (nm)	1.40 (1.26–1.50)	1.33 (1.10–1.45)	1.32 (1.26–1.43)	1.32 (1.15–1.43)	1.28 (1.23–1.38)	1.42 (1.31–1.50)	1.38 (1.29–1.53)
Aitken spread (nm)	1.32 (1.25–1.36)	1.45 (1.34–1.57)	1.22 (1.17–1.26)	1.28 (1.24–1.35)	1.30 (1.25–1.40)	1.27 (1.25–1.31)	1.31 (1.28–1.34)
Accum. spread (nm)	1.59 (1.51–1.69)	1.62 (1.53–1.70)	1.57 (1.49–1.65)	1.53 (1.49–1.55)	1.70 (1.64–1.70)	1.68 (1.61–1.70)	1.50 (1.42–1.66)
Composition							
Org (ng m ⁻³)	< 50*	76 (47–110)	< 50*	< 50*	< 50*	< 50*	< 50*
SO ₄ (ng m ⁻³)	88 (68–110)	180 (120-250)	87 (64–120)	130 (110–140)	92 (62–110)	120 (87–140)	180 (130-210)
SSA (ng m ⁻³)	100 (41–130)	85 (40–170)	88 (71–140)	25 (19-41)	270 (110–380)	140 (100–170)	400 (270–530)
Continental / anthropogenic influences							
Radon (mBq m ⁻³)	25.7 (16.0–40.2)	72.9 (59.6–88.5)	47.6 (38.7–53.6)	89.8 (63.6–97.4)	43.8 (32.3–54.1)	61.5 (54.5–74.1)	44.3 (38.4–54.4)
BC (ng m ⁻³)	< 8*	10.5 (7.6–15.0)	< 8*	< 8*	< 8*	< 8*	< 8*

* Below detection limit



Figure S4: Estimated total precipitation experienced by air masses during the previous 48 hours prior to reaching reaching the ship. Air mass locations and corresponding precipitation rates were determined on an hourly basis from ensembles of 27 back trajectories generated with the HYSPLIT modelling system. Periods influenced by different air masses are labelled and delimited by dashed lines.



Figure S5: Influence of (a) ultrafine aerosol and (b) SSA on the non-accumulation mode CCN0.5% concentrations. Periods influenced by different air masses are labelled and delimited by dashed grey lines. Grey data points indicate times when sea salt measurements were unavailable.



Figure S6: Ocean surface Chl-a concentrations along the averaged 7-day air mass trajectories corresponding to three episodes of CCNrelevant Aitken mode aerosol, projected on a map of the Southern Ocean (a,c,e) and as air mass altitude profiles (b,d,f). For each of the map projections, the ship's position is indicated by the black diamond. In the altitude profiles, the black line represents the height of the inversion layer, while the coloured line represents the altitude of the air mass during the hours prior to reaching the ship. In all panels, the colour scale represents the local ocean surface Chl-a concentration.



Figure S7: Ocean surface Chl-a concentrations for an averaged subset of 7-day air mass trajectories (a) projected on a map of the Southern Ocean and (b) as air mass altitude profiles. The trajectories correspond to a strong peak in nucleation mode aerosol concentrations observed between 17:00–20:00 on 10th February 2015. For the map projection, the ship's position is indicated by the black diamond. In the altitude profile, the black line represents the height of the inversion layer, while the coloured line represents the altitude of the air mass during the

hours prior to reaching the ship. In both panels, the colour scale represents the local ocean surface Chl-a concentration.



Figure S8: Mass concentrations of the dominant compositional species observed in sub-micron aerosol throughout the voyage. A six-hour rolling average has been applied to assist in identification of temporal trends. The shaded regions represent the standard deviation within each six-hour average. Periods influenced by different air masses are labelled and delimited by dashed lines.



Figure S9: 7-day air mass back trajectories generated at hourly intervals during the *cAU* sampling period. Coloured data points represent the air mass location at each hour along the back trajectory and the ship's path is shown in black.

The colour scale gives the altitude of the air mass relative to the estimated boundary layer, and grey data points represent hours when the air mass passed through the free troposphere.



5 Figure S10: 7-day air mass back trajectories generated at hourly intervals during the *cAA-I* sampling period. Coloured data points represent the air mass location at each hour along the back trajectory and the ship's path is shown in black. The colour scale gives the altitude of the air mass relative to the estimated boundary layer, and grey data points represent hours when the air mass passed through the free troposphere.



Figure S11: Meteorological observations at the southern-most extent of the voyage, demonstrating a pronounced decrease in air temperature (a) and absolute humidity (b) below 64 °S, likely indicating a transition from the Ferrel atmospheric cell into the colder and dryer polar cell.

References

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