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# Constraining the Surface Flux of Sea Spray Particles from the Southern Ocean

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## Key Points:

- Current-era GCMs over-estimate sea spray concentrations relative to measurements in the Ross Sea
- Better constraints for sea spray flux were found by tuning wind-speed based parameterizations to these observations
- Variations in sea surface temperature did not explain further variability within the temperature range studied

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## Abstract

Modeling the shortwave radiation balance over the Southern Ocean region remains a challenge for Earth system models. To investigate whether this is related to the representation of aerosol-cloud interactions, we compared measurements of the total number concentration of sea spray generated particles within the Southern Ocean region to model predictions thereof. Measurements were conducted from a container laboratory aboard the R/V *Tangaroa* throughout an austral summer voyage to the Ross Sea. We used source-receptor modeling to calculate the sensitivity of our measurements to upwind surface fluxes. From this approach, we could constrain empirical parameterizations of sea spray surface flux based on surface wind speed and sea surface temperature. A newly tuned parameterization for the flux of sea spray particles based on the near-surface wind speed is presented. Comparisons to existing model parameterizations revealed that present model parameterizations led to over-estimations of sea spray concentrations. In contrast to previous studies, we found that including sea surface temperature as an explanatory variable did not substantially improve model-measurement agreement. To test whether or not the parameterization may be applicable globally, we conducted a similar regression analysis using a database of in situ whitecap measurements. We found that the key fitting parameter within this regression agreed well the parameterization of sea spray flux. Finally, we compared calculations from the best model of surface flux to boundary layer measurements collected onboard an aircraft throughout the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES), finding good agreement overall.

## 1 Introduction

In the remote boundary layer of the Southern Ocean, continental sources of particulate matter such as black carbon, terrestrial monoterpenes, dusts, and pollen contribute very little to the population of suspended particulate (Murphy et al., 1998). As a result, the magnitudes of the direct and indirect shortwave radiative effects from the suspended particulate within the region are largely driven by local, marine sources (Carslaw et al., 2013; McCoy et al., 2015). There has been a considerable amount of work in recent years to understand the excess of shortwave radiation reaching the ocean surface in the Southern Ocean within climate-chemistry models (CCMs), especially regarding the representation of clouds within these models (Trenberth & Fasullo, 2010; Bodas-Salcedo et al., 2014). Since hygroscopic particulate matter are a necessary precursor to cloud formation, they can indirectly exert a substantial influence on the radiation balance through modification of cloud brightness (Twomey, 1977) and cloud phase through the availability of ice nuclei (DeMott et al., 2010).

The natural sources of airborne particles in the region are the production of sea-spray generated particles (SSPs) from wind-wave interactions and ultra-fine particles from the homogeneous nucleation of sulfuric acid and other volatile vapours. However, the rate of production of SSPs remains an open problem: the number of particles entering the atmosphere of a given droplet size and at a given wind speed has been shown to vary by over an order of magnitude among existing parameterizations for the production of SSPs (Ovadnevaite et al., 2014). If one also accounts for the uncertainties related to predicting the dependence of the surface flux on the wind speed over the water, estimates for the intensity of the surface flux diverge further. As a result, both the concentration and seasonal cycle of SSPs remain poorly constrained in the Southern Ocean (Henzing et al., 2006; Revell et al., 2019). Several studies have shown that the lack of prediction accuracy for the flux of SSPs results in large biases between observed and modelled mass concentrations of SSPs in

67 the marine boundary layer (MBL), particularly in regions with cold waters (Jaeglé  
68 et al., 2011; Grythe et al., 2014; Witek et al., 2016).

69 In general, sea spray is the dominant source of particulate matter in the South-  
70 ern Ocean in terms of mass (Murphy et al., 1998); however, during ice formation in  
71 coastal Antarctica, wind-blown frost flowers and snow from sea-ice can also become  
72 locally prominent sources (Kaleschke et al., 2004; Yang et al., 2008). Since these  
73 particles are the largest in the region (Quinn et al., 2017), they are also a substantial  
74 contributor to the local aerosol optical depth (AOD) (Shindell et al., 2013). While  
75 the contribution of SSPs to the regional AOD is much more significant than its con-  
76 tribution to cloud albedo in the Northern Hemisphere, over the Southern Ocean it  
77 is precisely the opposite (Ayash et al., 2008). This highlights that SSPs are a re-  
78 gionally important component of cloud formation over the Southern Ocean. This is  
79 not surprising: SSPs are mainly comprised of highly soluble sea salt and so they are  
80 very efficient cloud condensation nuclei (CCN) (Petters & Kreidenweis, 2007). While  
81 SSPs form only a small fraction of CCN globally, they can make up  $\sim 65\%$  of CCN  
82 over the Southern Ocean (Quinn et al., 2017).

83 Recent studies have also shown that SSPs can act as ice nucleating particles  
84 (INPs), which encourage the phase transition of cloud droplets to ice (DeMott et  
85 al., 2016; McCluskey et al., 2018). Since the Southern Ocean is far removed from  
86 continental sources of INPs (e.g. dust), SSPs may be the only source of INPs in  
87 the region. Ice nucleation sites within the droplets are likely a result of suspended  
88 amounts of organic material within the sea surface microlayer which became en-  
89 trained within the droplets during formation (DeMott et al., 2016). However,  
90 organic materials form very little of the mass composition of the resulting SSPs  
91 (Murphy et al., 1998); hence, the ice-nucleating potential of sea spray is very weak  
92 relative to continental sources such as mineral dusts (McCluskey et al., 2018). Still,  
93 the capacity for SSPs to modulate cloud phase represents an additional mechanism  
94 through which they can affect the local radiation balance.

95 While we have emphasized the potential radiative effects SSPs might have  
96 on the Southern Ocean region, there are other ways in which they can perturb the  
97 Earth system. Several studies have shown that the largest SSPs are non-negligible  
98 contributors to the exchange of latent and specific heat across the ocean–atmosphere  
99 interface (Richter & Sullivan, 2013; Ortiz-Suslow et al., 2016). In a bulk flux model  
100 of the air–sea exchange of heat, Andreas et al. (2015) showed that these large,  
101 “shear” sea-spray droplets accounted for fluxes of sensible and latent heat on the  
102 same order of magnitude as fluxes directly from the ocean–atmosphere interface  
103 at high wind speeds ( $U_{10} > 15 \text{ m s}^{-1}$ ). Observations and model simulations have  
104 shown that the rate of momentum transferred from the atmosphere to the ocean  
105 starts to decrease after a critical threshold wind speed is passed ( $30 \text{ m s}^{-1}$ ; (Powell  
106 et al., 2003; Bao et al., 2011; Hwang, 2018)). Theoretical work has suggested that  
107 this change is driven by the exchange of sensible heat between the largest droplets  
108 and the atmosphere, which become more abundant at high wind speeds (Bao et al.,  
109 2011). This leads to considerable biases in the prediction of storm intensity (Bao et  
110 al., 2011). The ability to predict the abundance of SSPs is therefore vital to fully  
111 understanding many macroscopic processes within the region.

112 To constrain the potential role sea spray may have on the regional radiation  
113 budget, it is first necessary to validate current parameterizations for its flux against  
114 in situ observations of its abundance. However, there is currently a dearth of such  
115 observations over the Southern Ocean. In this work we present measurements of  
116 the total number concentration of airborne particles recorded throughout an austral  
117 summer voyage to the Ross Sea aboard the R/V *Tangaroa*. We use these mea-  
118 surements to test existing empirical parameterizations which describe the flux of  
119 particles from wave breaking events in open seas. Measurements from instruments

120 onboard the High-Performance Instrumented Airborne Platform for Environmental  
 121 Research (HIAPER) throughout the Southern Ocean Clouds, Radiation, Aerosol  
 122 Transport Experimental Study (SOCRATES) were also used to validate these pa-  
 123 rameterizations. Since the winds throughout both of these experiments included the  
 124 extremes of surface conditions encountered at the air–sea interface, understanding  
 125 the flux of SSPs in this highly dynamic region will be valuable in constraining both  
 126 current and future flux estimates.

## 127 2 Methods

### 128 2.1 Measurements

129 The voyage aboard the R/V *Tangaroa* began on February 9<sup>th</sup> and ended on  
 130 March 21<sup>st</sup>, 2018 departing and returning to Wellington, New Zealand (41°17' S,  
 131 174°46' E). The bulk of the voyage was spent in waters south of 60°S, with 17 days  
 132 of the voyage spent in seas between 60–70°S and 13 days of the voyage south of  
 133 70°S.

134 In situ measurements of boundary layer aerosol were conducted from a con-  
 135 tainer laboratory on the shelter deck of the R/V *Tangaroa* (2 m a.s.l.). The instru-  
 136 ments within the container laboratory drew a continuous air sample through 40 m of  
 137 100 mm ID anti-static tubing (EOLU PU; IPL Ltd.) from the mast of the R/V *Tan-*  
 138 *garoa* (15 m a.s.l.). For the purposes of this study, we have primarily focussed on  
 139 measurements from the passive cavity aerosol spectrometer probe (PCASP-100X;  
 140 Droplet Measurement Technologies) with supplementary data from a differential  
 141 mobility particle sizer (DMPS; TSI). The PCASP-100X is an optical particle counter  
 142 which measured the number concentration size spectra of particles within the air  
 143 sample. The instrument is capable of detecting particles with optical diameters  
 144 between 0.1–3.0  $\mu\text{m}$  in 30 size bins at 1 Hz. The DMPS measured the number con-  
 145 centration size spectra of particles within the air sample with mobility diameters  
 146 between 0.02–0.3  $\mu\text{m}$  in 32 size bins once every 10 minutes. We have corrected  
 147 the number concentration measurements according to calculations of the sampling  
 148 and transport efficiency from Brockman (2001). These calculations accounted for  
 149 anisokinetic sampling conditions, diffusion of the particles toward the tube walls,  
 150 and gravitational settling of the particles. All of these calculations were based on  
 151 empirical parameterizations of these losses in a turbulent flow. Across the spectrum  
 152 of sizes we measured, we estimate that the total sampling efficiency was at most  
 153 93%, but no less than 90%.

154 Throughout the voyage, a cavity ring-down spectrometer (CRDS; Picarro  
 155 G2301) measured mole fractions of CO<sub>2</sub>, CH<sub>4</sub>, and H<sub>2</sub>O from a separate sampling  
 156 line. The sampling line of the CRDS was within 5 m of the main sampling line used  
 157 for the particulate sampling. Intrusions of ship exhaust from the rear of the ship  
 158 would have been sufficiently well-mixed in the turbulent air around the ship super-  
 159 structure so as to affect both sampling lines. We used a threshold limit of 405 ppm  
 160 of CO<sub>2</sub> to detect when ship exhaust contaminated our main sampling line. This  
 161 threshold was well above the trend line of the [CO<sub>2</sub>] mole fraction time-series. After  
 162 removing these outliers, we used 1 Hz sub-samples of the particle number concen-  
 163 trations to calculate 1-minute averages of the number concentration size spectra and  
 164 its standard deviation. When the standard deviation of the 1 Hz samples deviated  
 165 significantly from Poisson counting statistics the sample was removed.

166 This study also incorporated measurements from the New Zealand Met Ser-  
 167 vice’s Automated Weather Station (AWS) aboard the R/V *Tangaroa*. The AWS  
 168 anemometer was positioned at 22.5 m a.s.l. on the mast of the ship, while the rest  
 169 of the AWS was positioned at 15 m a.s.l. The AWS measured: atmospheric pres-

170 sure, atmospheric temperature, relative humidity, wind speed, wind direction, and  
171 accumulated precipitation. Measurements of the average relative wind speed and  
172 wind direction were made using a pair of ultrasonic anemometers (Gill WindSonic)  
173 and reported at 1-minute intervals. The measurements of wind speed were corrected  
174 according to directionally-dependent acceleration factors, based on a model of air  
175 flow around the R/V *Tangaroa*'s superstructure (Popinet et al., 2004; Smith et al.,  
176 2011). The wind speeds were then corrected according to the ship heading and speed  
177 to derive the true wind speed and wind direction. Finally, the acceleration-corrected,  
178 true wind speed at 22.5 m was scaled to the 10 m reference level using the bulk flux  
179 algorithms developed from the Coupled Ocean-Atmosphere Response Experiment  
180 (COARE) (Edson et al., 2013). In employing the COARE bulk flux algorithms, we  
181 have not accounted for differences in the height of the AWS due to heave with re-  
182 spect to mean sea level, which may amount to  $\pm 4$  m in heavy seas. If for a given  
183 measurement, the pitch or roll of the ship was significant with respect to the mean  
184 wind vector, then the measured wind speed would have been systematically biased  
185 low. However, throughout the voyage, the pitch of the ship was  $< 20^\circ$ , and so these  
186 corrections would be less than 6%.

187 Measurements of the boundary layer number concentration size spectra were  
188 also conducted onboard HIAPER, a modified Gulfstream V aircraft, from January  
189 16<sup>th</sup>–February 24<sup>th</sup>, 2018. These measurements were part of the SOCRATES ex-  
190 periment. Over the course of the experiment, there were 15 flights, which departed  
191 and returned to Hobart, Australia. We have focused on the flights which coincided  
192 with our observational record, namely Research Flights (RF) 10–15 which took  
193 place between February 7<sup>th</sup>–24<sup>th</sup>, 2018. Two Ultra-High Sensitivity Aerosol Spec-  
194 trometers (UHSAS; Droplet Measurement Technologies) were used throughout the  
195 experiment to measure the number concentration size spectra of particles within the  
196 surrounding air; however, for this study we focused solely on the measurements from  
197 the UHSAS mounted inside of the aircraft. The UHSAS sampled ambient air via a  
198 counterflow virtual impactor inlet mounted outside of the aircraft. This ensured that  
199 the internal flow rate of the UHSAS was isokinetically matched to the exterior flow  
200 around HIAPER. Like the PCASP-100X, the UHSAS is an optical particle counter  
201 which can detect particles with optical diameters between 0.059–1.022  $\mu\text{m}$  in 100  
202 discrete size bins at 1 Hz. It was determined that corrections to the number concen-  
203 tration size spectra from the additional ram pressure of sampling the aerosol from a  
204 moving aircraft would amount to less than 1%. This was substantially less than the  
205 observed variability in both the number concentration time series and the volume  
206 flow rate of the pump that provided the flow through the UHSAS. For each flight  
207 we identified 3–6 periods when the altitude was stable, there was little precipitation,  
208 and the observed number concentration size spectra were relatively stable. In each  
209 of these periods, we averaged the number concentration size spectra over 5–10 min-  
210 utes. This resulted in 28 unique measurements between 69–6,100 m a.s.l, 17 of which  
211 were in the boundary layer.

## 212 2.2 FLEXPART-WRF

213 The FLEXPARTicle transport model (FLEXPART), FLEXPART-WRF,  
214 is a Lagrangian particle dispersion model designed to model particle trajectories  
215 within mesoscale meteorological fields from the Weather Research & Forecasting  
216 Model (WRF) (Brioude et al., 2013). For this study, we used meteorological fore-  
217 casts from the real-time Antarctic Mesoscale Prediction System (AMPS) (Polar  
218 Meteorology Group, Byrd Polar and Climate Research Center, 2018). AMPS uses  
219 a variety of data sources to constrain these forecasts, including near-real-time sea-  
220 ice concentrations measured from the Special Sensor Microwave/Imager (SSM/I)  
221 radiometer and sea surface temperature (SST) data from the National Center for  
222 Environmental Prediction (NCEP) (Bromwich et al., 2005). Initial and boundary

223 conditions for AMPS were specified according to near-real-time forecasts from the  
 224 NCEP Global Forecasting System (Bromwich et al., 2005). We used the AMPS  
 225 output with the widest spatial coverage, domain 1, which has a horizontal resolu-  
 226 tion of  $24 \times 24$  km, a vertical resolution of 61  $\eta$  levels, and a temporal resolution of  
 227 three hours. The AMPS forecasts used throughout this study were downloaded from  
 228 <https://www.earthsystemgrid.org/project/amps.html>.

229 We initialized 100,000 particle trajectories from the geographic location of  
 230 the R/V *Tangaroa* every three hours to match the temporal resolution of AMPS.  
 231 Additional simulations were run for every hour in between the meteorological time  
 232 steps if the R/V *Tangaroa* had entered a new grid cell in the AMPS domain. These  
 233 two criteria resulted in 651 unique simulations. To trace losses due to deposition  
 234 throughout the simulation, FLEXPART assigned each particle a unit mass dis-  
 235 tributed over a log-normal size distribution. To match our observations, we centered  
 236 this distribution around a geometric dry diameter of  $0.20 \mu\text{m}$  ( $D_{p,g} = 0.4 \mu\text{m}$  at  
 237 80% relative humidity), with a geometric standard deviation of 2.00, and a dry den-  
 238 sity of  $1.84 \text{ g cm}^{-3}$ . FLEXPART-WRF used the discretized Langevin equation to  
 239 describe the turbulent dispersion of these particles through the atmosphere in re-  
 240 verse time with an adaptive time-step strictly less than 180 s. The particles were  
 241 advected through the meteorological fields specified by AMPS from the time of mea-  
 242 surement up to five days prior in reverse time. Throughout the trajectory, losses  
 243 of particle mass due to dry deposition were calculated according to the resistance  
 244 method (Hicks et al., 1987). To improve these calculations we modified FLEXPART-  
 245 WRF to account for hygroscopic particle growth according to the ambient relative  
 246 humidity (Gerber, 1985), since changes in particle size can significantly affect a  
 247 particle’s settling velocity and dry deposition velocity. FLEXPART-WRF also ac-  
 248 counted for losses of particle mass from precipitation and droplet activation. Within  
 249 clouds, FLEXPART-WRF calculated the scavenging rate of particles from droplet  
 250 activation according to the parameterization of Hertel et al. (1995). For scavenging  
 251 by precipitation below cloud, loss rates were estimated from the following empirical  
 252 relationship:

$$253 \quad \Lambda = AI_s^B \quad (1)$$

254 where the scavenging rate,  $\Lambda$ , was calculated as a function of the rain-equivalent  
 255 snow intensity,  $I_s$  ( $\text{mm hr}^{-1}$ ), and user-set scavenging coefficients, A and B. While  
 256 fairly good representations of particle scavenging from rain exist in the litera-  
 257 ture, there is substantially more uncertainty with regards to the scavenging from  
 258 snow (Slinn, 1977). Recent parameterizations of below-cloud scavenging in the non-  
 259 WRF version of FLEXPART recommend applying an empirical fit to a set of snow  
 260 scavenging rates measured in southern Finland (Kyrö et al., 2009; Grythe et al.,  
 261 2017). However, this parameterization doesn’t explicitly account for increases in  
 262 scavenging with increasing snow intensity. We observed that the differences in scav-  
 263 enging rates Kyrö et al. (2009) observed across the particle size spectra were small  
 264 compared to the difference in median snow scavenging rates they observed between  
 265 their median observed snow intensity ( $0.2 \text{ mm hr}^{-1}$ ) and peak snow intensity ( $5$   
 266  $\text{mm hr}^{-1}$ ). We used the median scavenging rates and snow intensity values they  
 267 reported to estimate the following scavenging coefficients for snow:  $A = 4 \times 10^{-5}$   
 268 and  $B = 0.43$ . For reference, the typical values used by FLEXPART-WRF for rain  
 269 are  $5 \times 10^{-6}$  and 0.62, respectively. Together with the dry deposition velocity, the  
 270 mass concentration loss rate could be described at any point in the simulation by  
 271 the following:

$$272 \quad \frac{dm}{dt} = - \left( \frac{v_d}{h} + \Lambda \right) m \quad (2)$$

273 where  $m$  is the mass of the particle,  $v_d$  is the dry deposition velocity, and  $h$  was the  
 274 height of the layer in which dry deposition occurred (30 m a.s.l.).

275 In reverse mode, FLEXPART-WRF calculated the residence time of the parti-  
 276 cles within the lowest 100 m of the atmosphere. The residence time calculation was  
 277 weighted by the local air density and the residual mass of the particles within the  
 278 grid-cell. The weighted residence time was normalized by the initial mass of the par-  
 279 ticles such that the resulting residence time accounted for losses from wet and dry  
 280 deposition as described above. Finally, FLEXPART-WRF integrated the weighted  
 281 residence time over the duration of the meteorological time-step for each grid-cell of  
 282 the AMPS domain.

### 283 2.3 Quantifying the contribution of sea spray

284 Previous studies have shown that in pristine marine environments, the contri-  
 285 bution of SSPs to the number concentration size spectra can be characterized by a  
 286 single log-normal number concentration size distribution (Modini et al., 2015; Quinn  
 287 et al., 2017). Distributions derived from this method have been shown to agree well  
 288 with number concentration size spectra measured during laboratory wave-breaking  
 289 experiments and the “canonical sea spray size distribution” derived from other  
 290 studies (Prather et al., 2013; Lewis & Schwartz, 2004). This also agrees with the  
 291 mass composition of the particles measured in the Southern Ocean boundary layer  
 292 during the ACE-1 campaign, which found that particles larger than  $0.3 \mu\text{m}$  were  
 293 composed almost entirely of sea-salt (Murphy et al., 1998). We applied this method-  
 294 ology to our own measurements of the number concentration size spectra from the  
 295 PCASP-100X; i.e. we fit a number concentration size spectra,  $dn(d \log D_p)^{-1}$ , of the  
 296 following form:

$$297 \frac{dn}{d \log D_p} = \frac{N}{\sqrt{2\pi} \log(\sigma_g)} \exp \left[ -\frac{\log^2 \left( \frac{D_p}{D_{p,g}} \right)}{2 \log^2(\sigma_g)} \right] \quad (3)$$

298 where  $D_p$  is the particle diameter,  $N$  is the total number concentration of particles,  
 299  $D_{p,g}$  is the geometric mean diameter of the distribution, and  $\sigma_g$  is the geometric  
 300 standard deviation of the distribution. After fitting, the retrieved spectra were cor-  
 301 rected to a relative humidity of 80% (Gerber, 1985). In the process of fitting, the  
 302 geometric standard deviation ( $\sigma_g$ ) of the mode was fixed to a value of 2, which best  
 303 fit our data. While Modini et al. (2015) and Quinn et al. (2017) allowed  $\sigma_g$  to freely  
 304 vary in their regression analysis, the variance-covariance matrix from our regression  
 305 indicated that the resulting parameters were significantly correlated, since the data  
 306 very weakly constrained  $\sigma_g$ .

307 To calculate the total number concentration of sea spray from the  
 308 FLEXPART-WRF residence time, we assumed that the surface flux of SSPs also  
 309 followed a log-normal distribution:

$$310 \frac{\partial f}{\partial \log D_p} = \frac{F}{\sqrt{2\pi} \log(\sigma_g)} \exp \left[ -\frac{\log^2 \left( \frac{D_p}{D_{p,g}} \right)}{2 \log^2(\sigma_g)} \right] \quad (4)$$

311 where  $f$  is the partial particle flux in  $\text{m}^{-2} \text{s}^{-1}$ ,  $F$  is the total particle flux in  $\text{m}^{-2}$   
 312  $\text{s}^{-1}$ , and  $D_p$  is the particle diameter.

313 The most widely used empirical approach for constraining the particle flux  
 314 from the ocean surface is the “whitecap method”. It results from the following as-  
 315 sumptions: first, that the total flux of particles entering the atmosphere from the  
 316 ocean surface can be determined from the fractional surface coverage of whitecaps,  
 317  $W$  (“whitecap fraction”); second, that the whitecap fraction can be adequately de-  
 318 termined from the 10 m scalar wind speed over the ocean,  $U_{10}$ ; and third, that the  
 319 shape of the SSP size distribution is not a function of wind speed. Laboratory and  
 320 field experiments have shown that all of these assumptions are reasonable (Monahan



321 & Ó Muircheartaigh, 1980; Monahan et al., 1986). Hence, the total number of par-  
 322 ticles entering the atmosphere,  $F$ , can be predicted from just the 10 m wind speed,  
 323  $U_{10}$ :

$$324 \quad \begin{aligned} F &= \frac{E}{\tau} W(U_{10}) \\ F &= \alpha W(U_{10}) \end{aligned} \quad (5)$$

325 where  $W$  is a function that models how the surface coverage of whitecaps increases  
 326 as a function of wind speed,  $E$  is the number of particles produced per whitecap,  
 327 and  $\tau$  is the lifetime of the whitecaps. Since we can only hope to constrain one con-  
 328 stant pre-factor, we combine both  $E$  and  $\tau$  into a single parameter  $\alpha$ , which we  
 329 assume to be constant. Historically, the whitecap function  $W$  has been assumed  
 330 to be a simple power-law, based on early field observations of whitecap forma-  
 331 tion (Monahan, 1971). However, it has since been well-established that whitecaps  
 332 do not form in the open ocean until the 10 m wind speed exceeds 3–4 m s<sup>-1</sup>, which  
 333 is a feature that cannot be described by a power-law model (A. Callaghan et al.,  
 334 2008; Schwendeman & Thomson, 2015; Bell et al., 2017). We considered three other  
 335 wind-dependent models of the surface flux that incorporated a threshold wind speed  
 336 below which very few SSPs are produced:

$$337 \quad \begin{aligned} W_{PL} &= a_1 U_{10}^{a_2} \\ W_C(U_{10}) &= \begin{cases} b_1 (U_{10} - b_2)^3, & U_{10} \geq b_2 \\ 0, & U_{10} < b_2 \end{cases} \\ W_F(U_{10}) &= 1 - \Phi\left(\frac{c_1}{\sqrt{U_{10}}}\right) \\ W_{LLPL}(U_{10}) &= \frac{d_1 U_{10}^{d_2}}{1 + \left(\frac{U_{10}}{d_3}\right)^{-d_4}} \end{aligned} \quad (6)$$

338 where  $a_x, b_x, c_x$  and  $d_x$  and are empirical parameters determined from regression  
 339 analysis with our observations, and  $\Phi$  is the error function. The first function,  
 340  $W_{PL}$  (‘PL’ = ‘Power-Law’), is as previously introduced. The second function,  $W_C$   
 341 (‘C’ = ‘Cubic’), has been used to match more recent field observations of white-  
 342 caps (A. Callaghan et al., 2008; Schwendeman & Thomson, 2015)). The third func-  
 343 tion,  $W_F$  (‘F’ = ‘Fetch’), was based on the theoretical work of Snyder and Kennedy  
 344 (1983), who developed a model of whitecap production based on a fetch dependent  
 345 threshold for wave breaking. While the work of Xu et al. (2000) showed that the  
 346 whitecap fraction could be fully determined from the model of Snyder and Kennedy  
 347 (1983) if both the wind speed and fetch were known, the fetch was typically unlim-  
 348 ited throughout our observation period. In high fetch regimes, the coverage of white-  
 349 caps is only very weakly dependent on variations in fetch (Piazzola et al., 2002). As  
 350 a result, we treated  $c_1$ , which is a function of the fetch, as a free parameter to be  
 351 determined through regression, since a single value should accurately describe the  
 352 data. The last function,  $W_{LLPL}$  (‘LLPL’ = ‘Log-Logistic Power-Law’) combined  
 353 the power-law with a log-logistic curve to emulate the threshold mechanism. While  
 354  $W_{PL}$ ,  $W_C$  and  $W_{LLPL}$  predict that the surface flux will continue increasing as a  
 355 function of wind speed, the Fetch model ( $W_F$ ) is the only model which predicts that  
 356 there exists an upper bound on the particle flux at high wind speeds.  
 357

358 There has also been some debate as to how the temperature of the sea water  
 359 might moderate whitecap formation (Mårtensson et al., 2003; Sellegri et al., 2006;  
 360 Jaeglé et al., 2011; Zábori et al., 2012; A. H. Callaghan et al., 2014; Grythe et al.,  
 361 2014). This was tested directly with SST data from NCEP, which was available  
 362 from the AMPS forecasts. Thus, the model of surface flux was expanded to:

$$363 \quad \begin{aligned} F &= \alpha(T_w) W(U_{10}) \\ \alpha(T_w) &= \alpha_0 (1 + \alpha_1 T_w) \end{aligned} \quad (7)$$

364

365 where the coefficient  $\alpha$ , which describes both the lifetime of the whitecaps and  
 366 the number of particles produced per whitecap, is now a function of the SST,  $T_w$ .  
 367 Note that, in reality, parameters  $\alpha_0$  and other scaling coefficients within  $W$  (e.g.  
 368  $a_1, b_1, d_1$ ) cannot be determined independently, so they are combined into a single  
 369 parameter for each regression (e.g.  $a_1^* = a_1\alpha$ ). Finally, we assumed that the surface  
 370 flux was well-mixed within the lowest atmospheric grid cell in FLEXPART-WRF,  $h$   
 371 = 100 m. Following these assumptions, the number concentration of particles in the  
 372 SSP mode,  $\hat{N}$ , was calculated according to:

$$373 \quad \hat{N}_i = \frac{1}{h} \int_{-t_0}^0 \iint_O F(1 - C_{ice}) d\mathcal{P}_i \quad (8)$$

374 where  $t_0$  was the length of the FLEXPART-WRF simulation,  $C_{ice}$  was the fractional  
 375 surface coverage of sea ice,  $O$  denotes that the integral was only integrated over  
 376 oceans, and  $\mathcal{P}_i$  was the map of footprint residence times for the observation  $i$ .

377 A non-linear least-squares regression analysis optimized the set of parameters  
 378 for each surface flux model,  $W$ , being tested. Parameter optimization was achieved  
 379 with the Gauss-Newton algorithm, where the goodness-of-fit was measured by the  
 380 Nash-Sutcliffe model efficiency coefficient (NSE):

$$381 \quad NSE = 1 - \frac{\sum_{i=1}^m (N_i - \hat{N}_i)^2}{\sum_{i=1}^m (N_i - \bar{N})^2} \quad (9)$$

382 where  $m$  was the total number of observations and  $\bar{N}$  represents the average of ev-  
 383 ery measurement of  $N_i$  within the dataset (Nash & Sutcliffe, 1970). To account for  
 384 differences between the number of parameters between models, we also calculated  
 385 the Akaike Information Criterion (AIC), which penalized models with more param-  
 386 eters (Akaike, 1974):

$$387 \quad AIC = 2k + m \log \left( \frac{1}{m} \sum_{i=1}^m (N_i - \hat{N}_i)^2 \right) \quad (10)$$

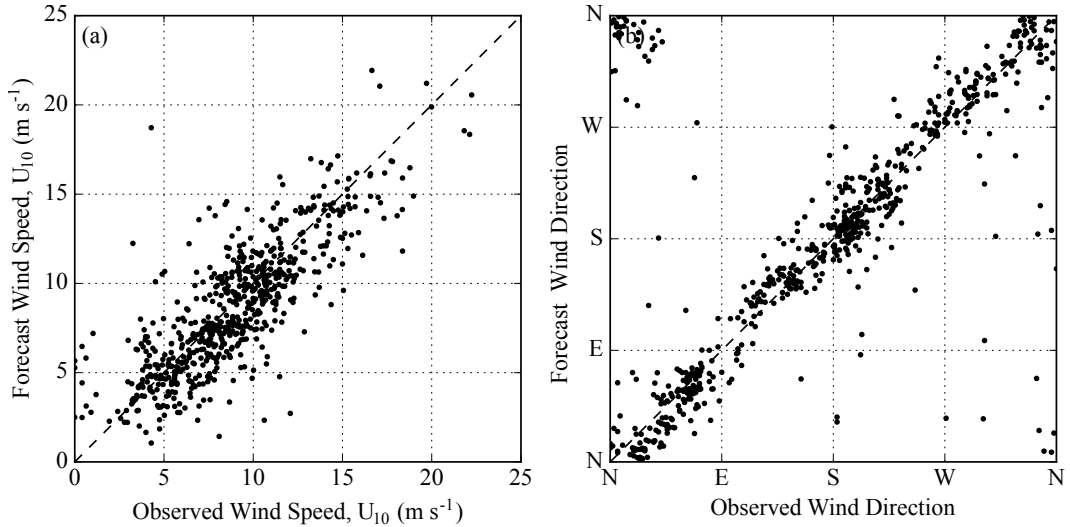
388 where  $k$  was the total number of parameters for a given model. The best model of  
 389 surface flux was the model which minimized the AIC.

### 390 **3 Results**

#### 391 **3.1 Comparisons Between Surface Meteorological Measurements** 392 **and Model Forecasts**

393 To demonstrate that the transport simulations produced a meaningful link  
 394 between the observations and surface fluxes, it was necessary to first validate the  
 395 Antarctic Mesoscale Prediction System's meteorological fields against the record of  
 396 observations from the Automated Weather Station (AWS) aboard the R/V *Tan-*  
 397 *garoa*. As described in the Section 2.1, the AWS measured wind speeds at 22.5 m,  
 398 which were corrected to the 10 m reference height according to the COARE 3.5 bulk  
 399 flux algorithms. The corrected wind speeds were compared to the 10 m wind speeds  
 400 predicted by the AMPS forecasts by matching the location of the R/V *Tangaroa* to  
 401 the nearest grid-cell within AMPS. This comparison is presented in Fig. 1. The cor-  
 402 relation coefficients calculated for both the wind speed ( $R = 0.81$ ) and wind direc-  
 403 tion ( $R_o = 0.78$ ) between observations and forecasts were both significant ( $p < 0.01$ ),  
 404 where  $R_o$  represents the circular correlation coefficient (Fisher & Lee, 1986).

405 Despite the good agreement we found between the measured and forecast  
 406 winds, there was no spatio-temporal correlation between the rain-equivalent snow



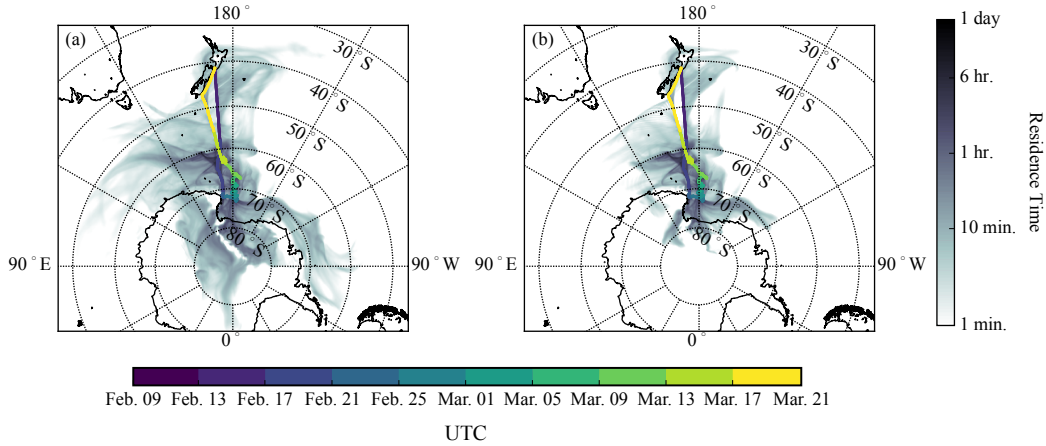
**Figure 1.** (a) The hourly 10 m scalar wind speeds from the AMPS forecasts were compared to the corrected wind speed (see Section 2.1) observed on the R/V *Tangaroa*. (b) The hourly 10 m wind direction.

407 rate measured aboard the R/V *Tangaroa* and the precipitation fields forecast by  
 408 AMPS. However, the climatological distribution of the rain-equivalent snow rate was  
 409 at least consistent between measurements and forecasts. This suggests that even if  
 410 precipitation was not spatially consistent with our observations, it was at least as  
 411 frequent, and of the right intensity within the AMPS forecasts. While comparing  
 412 localized, discrete events like precipitation can be challenging, even comparing our  
 413 measurements to grid cells within 100 km and within 6 hours of the R/V *Tangaroa*  
 414 measurements did not produce a significant correlation.

### 415 3.2 Source–Receptor Modeling

416 In Fig. 2a we show the cumulative five-day, near-surface, residence time for  
 417 all of the source–receptor simulations described in Section 2.2. The track of the  
 418 R/V *Tangaroa* throughout the voyage has also been shown for reference. As ex-  
 419 pected, the near-surface residence time was greatest near the R/V *Tangaroa*. This  
 420 indicated that our measurements were most sensitive to surface fluxes near the ship.  
 421 To understand how dry deposition might govern the concentration of SSPs, we also  
 422 ran several FLEXPART-WRF simulations in which the dry deposition velocity was  
 423 set to a fixed rate. However, the consistent turbulence of the atmosphere over the  
 424 Southern Ocean meant that the simulated particles were often very evenly dispersed  
 425 throughout the boundary layer. As a result, dry deposition was severely limited  
 426 throughout all of the simulations. Predicted surface flux sensitivities within these  
 427 simulations only began to diverge after 1–2 days had elapsed in simulation time;  
 428 however, by then the residence time was typically less than 5% of what it was near  
 429 the ship. Hence, dry deposition was not a strong factor controlling the concentration  
 430 of SSPs.

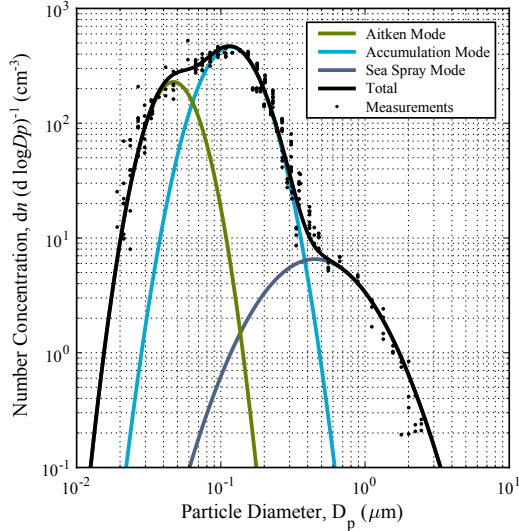
431 It was evident from Fig. 2a, however, that our observations near the Ross Ice  
 432 shelf were sensitive to non-marine sources. The fraction of the time the particles  
 433 spent above non-marine surfaces throughout their five-day simulations increased  
 434 rapidly as the R/V *Tangaroa* approached Cape Adare, Antarctica. Throughout this  
 435 period we observed strong, southerly winds, which brought continental, Antarctic air



**Figure 2.** (a) A map of the cumulative near-surface residence time derived from FLEXPART-WRF particle dispersion simulations. The model simulated the transport of  $1 \times 10^5$   $0.2 \mu\text{m}$  particles in reverse time from the time of measurement to five days prior. The near-surface residence time is simply the total amount of time the particle spent below 100 m a.g.l. Within FLEXPART-WRF the transport of the particles was calculated according to meteorological forecasts from the Antarctic Mesoscale Prediction System (AMPS). (b) The same map, but now for a 48-hr. simulation. The colored line in both panels marks the track of the R/V *Tangaroa* throughout the voyage.

436 across the Ross Ice Shelf. Intrusions of continental air into the MBL are a common  
 437 phenomenon within the region (Coggins et al., 2014; Coggins & McDonald, 2015).  
 438 Near the end of the observation period, after March 18<sup>th</sup>, 2018, the source-receptor  
 439 simulations showed that our measurements were also sensitive to surface fluxes from  
 440 the South Island of New Zealand. This was a direct result of the strong northerlies  
 441 we observed throughout our return transect.

442 As we identified in Section 3.1, the rain-equivalent snow rates forecast by  
 443 AMPS were not well-correlated with snowfall intensity measured onboard the R/V  
 444 *Tangaroa*. While the frequency of occurrence of these events seemed consistent, it  
 445 is important to note that the wet deposition scheme used by FLEXPART-WRF  
 446 implicitly assumed that in-cloud activation events only occurred within a precipitat-  
 447 ing cloud. Hence, the frequency of in-cloud scavenging events was almost certainly  
 448 under-estimated within the source-receptor simulations. As Hertel et al. (1995)  
 449 note, the magnitude of the loss of particles to in-cloud activation is almost always  
 450 greater than either below-cloud scavenging or dry deposition. In fact, an activation  
 451 event is always strong enough to terminate a particle trajectory within FLEXPART-  
 452 WRF. Therefore, it was expected that the source-receptor modeling vastly over-  
 453 estimated the near surface residence time by continuing to advect particles that  
 454 should have been completely scavenged by cloud. However, a lack of boundary  
 455 layer cloud within the simulation did not stem from this issue alone. It has been  
 456 well-established that there is currently a large shortwave radiation bias over the  
 457 Southern Ocean (Bodas-Salcedo et al., 2014). Observations of cloud base height  
 458 from radiosondes and ceilometer measurements throughout this same voyage showed  
 459 that the shortwave radiation bias is related to the lack of low-level cloud and fog  
 460 predicted within atmospheric models (Kuma et al., 2019). Therefore, it is reason-  
 461 able to expect that even with an improved in-cloud activation scheme (e.g. Grythe  
 462 et al. (2017)), FLEXPART-WRF still would have under-estimated the frequency  
 463 of droplet activation events. This would have a substantial impact on our source-

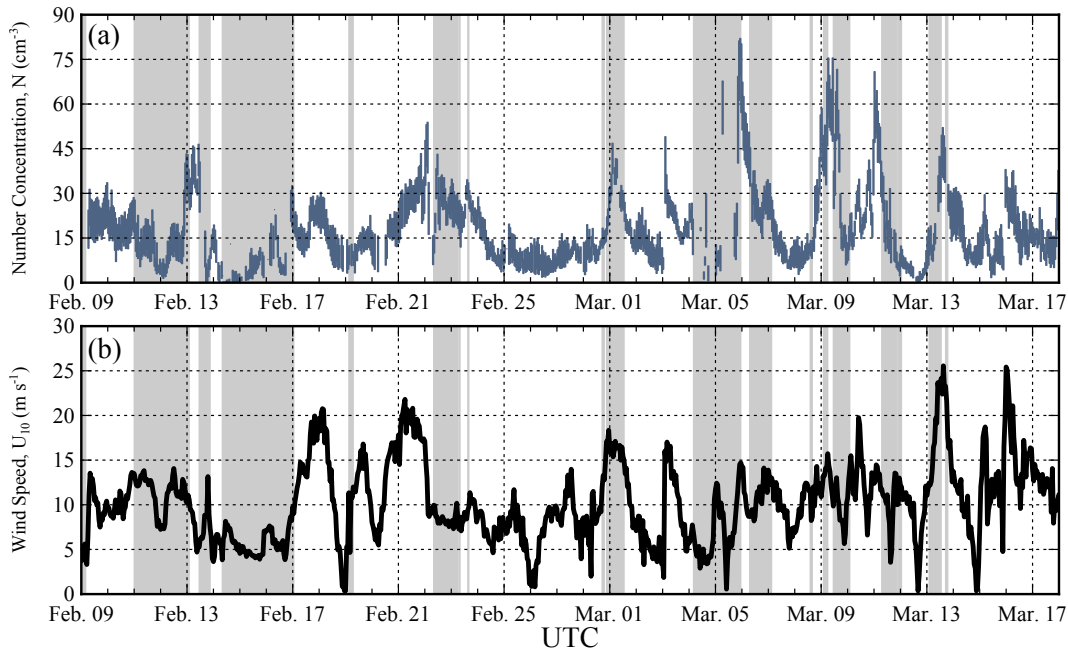


**Figure 3.** Number concentration size spectra measured from the PCASP-100X (0.1–3 $\mu\text{m}$ ) and the DMPS (0.02–0.3 $\mu\text{m}$ ) on February 20<sup>th</sup>, 2018 at 1300 UTC. Particle sizes have been corrected to 80% relative humidity (Gerber, 1985). Number concentrations for particles larger than 0.5  $\mu\text{m}$  were used to constrain a single log-normal number concentration size distribution (“SSP mode”). This method has been used by other researchers (e.g. Modini et al. (2015) and Quinn et al. (2017)) to constrain the contribution of SSPs to the total number concentration size spectra. The Aitken and accumulation modes are shown for reference.

464 receptor calculations, resulting in significantly less ocean surface area contributing  
 465 to the integral in Eq. (8). To visualize this effect, we have also shown the cumula-  
 466 tive near surface residence time for a two-day simulation in Fig. 2b, instead of the  
 467 five-day simulation shown in Fig. 2a. This is addressed further in Section 3.4.

### 468 3.3 Number Concentrations of Sea Spray

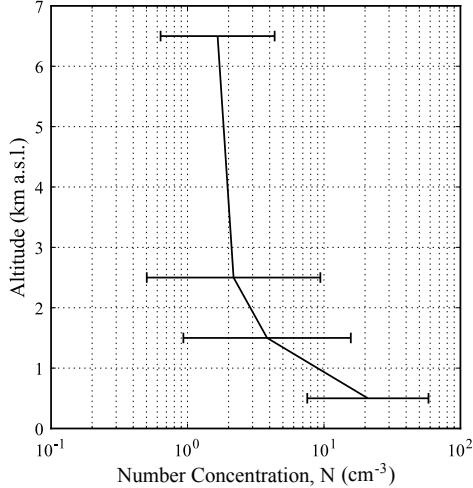
469 In Fig. 3 we show an example modal analysis of a number concentration size  
 470 spectrum measured by the PCASP-100X and DMPS aboard the R/V *Tangaroa*. The  
 471 size spectra were used to constrain three log-normal “modes” which represented the  
 472 entire size distribution. The largest of these modes, the SSP mode, is so-named as it  
 473 has been shown to be comprised almost solely of SSPs (Modini et al., 2015; Quinn  
 474 et al., 2017). At each hour of observation throughout the voyage we constrained the  
 475 SSP mode from the spectral measurements shown in Fig. 3, resulting in the time  
 476 series of the total number concentration of SSPs shown in Fig. 4. In general it was  
 477 sufficient to constrain the SSP mode from just the PCASP-100X measurements,  
 478 so the measurements from the DMPS were not used in this study, but are shown  
 479 for reference. Throughout the entire voyage, we observed the median and standard  
 480 deviation of the geometric mean diameter of the SSP mode to be  $0.4 \pm 0.05 \mu\text{m}$   
 481 at a relative humidity of 80%. This agreed well with the observations of Quinn et  
 482 al. (2017) in the Southern Ocean. This also agreed with the median dry diame-  
 483 ter of SSPs measured from laboratory generated waves, 140–200 nm (Prather et  
 484 al., 2013), since SSPs are twice as large at 80% relative humidity compared to dry  
 485 conditions (Gerber, 1985).



**Figure 4.** (a) The average hourly number concentration of sea spray particles is shown for the entire campaign with periods of fog ( $RH > 98\%$ ) or rain ( $> 1 \text{ mm hr}^{-1}$ ) marked by the shaded areas. (b) The hourly 10 m wind speed.

486 In Fig. 4a, the total number concentrations of SSPs and the 10 m scalar wind  
 487 speeds measured are shown from the beginning of the voyage, February 9<sup>th</sup>, 2018,  
 488 until March 18<sup>th</sup>, 2018. Throughout the voyage, there were several periods when  
 489 either fog or precipitation was observed at the ship. As expected, fog very efficiently  
 490 scavenged the particles in the SSP mode through droplet activation processes, much  
 491 more so than precipitation. However, the lack of observed particles during such  
 492 events meant that the SSP mode could not be constrained. This is particularly evi-  
 493 dent around March 5<sup>th</sup>, 2018. In the last three days of the voyage, March 18<sup>th</sup>–21<sup>st</sup>,  
 494 2018, we encountered strong northerly winds along the coast of New Zealand, which  
 495 transported terrestrial particles to the R/V *Tangaroa*. The addition of these non-  
 496 SSPs resulted in number concentration size spectra from which the SSP mode could  
 497 not be constrained. As a result, measurements when fog or precipitation was ob-  
 498 served at the ship, or when there was a significant influence from New Zealand were  
 499 excluded from the regression analysis presented in the following section.

500 In Fig. 4b we also show the 10 m wind speed throughout the same period of  
 501 measurement. We observed that when winds were light ( $U_{10} < 4 \text{ m s}^{-1}$ ) the total  
 502 number concentration of particles in the SSP mode was no more than  $10 \text{ cm}^{-3}$ , and  
 503 had a median of  $7 \text{ cm}^{-3}$ . Light-wind periods ( $U_{10} < 4 \text{ m s}^{-1}$ ) occurred 14% of the  
 504 time in upwind conditions, as weighted by the near surface residence time. However,  
 505 during a light-wind period on March 12<sup>th</sup>, 2018, there appeared to be no particles at  
 506 all. This agreed well with the transport modeling in Fig. 2, which showed that dur-  
 507 ing all light-wind periods, except the period occurring on March 12<sup>th</sup>, the particles  
 508 had a significant near surface residence time over Antarctica. While the boundary  
 509 layer over Antarctica is generally a very pristine environment, human activity from  
 510 research stations near the Ross Ice shelf and exposed mountain faces both represent  
 511 potential sources for the concentration of particles observed at low wind speeds. In  
 512 the regression analysis that followed, the median concentration of SSPs observed at



**Figure 5.** The total number concentration of sea spray generated particles is shown as a function of altitude over the course of several flights aboard HIAPER, (RF 11–15). The ranges within each altitude bin show the minimum and maximum number concentration observed. Flights RF 11–15 took place on the following days, in order: Feb. 17<sup>th</sup>, 18<sup>th</sup>, 20<sup>th</sup>, 21<sup>st</sup> and 24<sup>th</sup>, 2018.

513 low wind speeds was removed from the observations (except for the period around  
 514 March 12<sup>th</sup>, 2018). After removing this background concentration, we calculated the  
 515 hourly-averaged number concentration of particles in the SSP mode to be  $9 \text{ cm}^{-3}$ ,  
 516 with a maximum of  $62 \text{ cm}^{-3}$ .

517 In Fig. 5 we show the total number concentration of SSPs derived from the  
 518 UHSAS number concentration size spectra onboard HIAPER. These measurements  
 519 were taken over the course of several flights in February 2018 to illustrate the range  
 520 of concentrations observed within each 1 km bin. The measurements within one  
 521 kilometer of the Earth’s surface were always determined to be below-cloud (if any  
 522 cloud was present) and within the boundary layer. In contrast, all other bins were  
 523 determined to be above cloud (if any cloud was present) and above the boundary  
 524 layer. From Fig. 5 we can also identify that there was always at least  $5\text{--}10 \text{ cm}^{-3}$  of  
 525 SSPs in the boundary layer, which is consistent with the measurements at low wind  
 526 speeds on board the R/V *Tangaroa*. As expected, there were also very few SSPs  
 527 above the cloud, indicating that nearly all of these particles had been consumed  
 528 during cloud formation.

### 529 3.4 Regression Analysis

530 Predicted SSP concentrations can be obtained by integrating Eq. (8); however,  
 531 we have already identified that particle losses from in-cloud scavenging represented  
 532 the greatest uncertainty to our source–receptor modeling. To address this within  
 533 the regression analysis we allowed the simulation length,  $t_0$ , to vary as a free pa-  
 534 rameter within Eq. (8). In effect, this allowed the regression to estimate the return  
 535 rate of a droplet activation event within a boundary layer cloud (e.g. fog or ma-  
 536 rine stratocumulus) or a significant precipitation event ( $>10 \text{ mm hr}^{-1}$ ). Either of  
 537 these events would have efficiently scavenged the particle, thereby terminating its  
 538 trajectory. This approach is similar to the Statistical Wet Deposition method used  
 539 by other researchers, which prescribes the length of time it takes a surface flux of

540 particles to fully mix into the boundary layer after a precipitation event or cyclone  
 541 (e.g. Ovadnevaite et al. (2014)).

**Table 1.** The values listed in this table are the relative likelihood that a given parameterization of surface flux correctly predicted the observed number concentration of sea spray generated particles relative to the best parameterization. These values were calculated from the difference between the Aikake Information Criterion (AIC) of each parameterization and the AIC of the best parameterization ( $AIC_b$ ) according to the following:  $\exp(-0.5(AIC - AIC_b))$  (Burnham & Anderson, 2002). The AIC measures the log-likelihood that a given surface flux parameterization minimizes the residual sum of squares between predicted and observed concentrations while also penalizing parameterizations which include large numbers of parameters (Akaike, 1974). See Eq (6) for parameterization definitions.

Surface Flux parameterization	$W_{PL}^\ddagger$	$W_C^\dagger$	$W_F^\S$	$W_{LLPL}^{\dagger\dagger}$
$F(U_{10})$	$<1 \times 10^{-3}$	$<1 \times 10^{-3}$	1	0.2
$F(U_{10}, T_w)$	$<1 \times 10^{-3}$	$<1 \times 10^{-3}$	0.6	$<1 \times 10^{-3}$

$^\ddagger$ Power-Law;  $^\dagger$ Cubic;  $^\S$ Fetch;  $^{\dagger\dagger}$ Log-Logistic Power-Law.

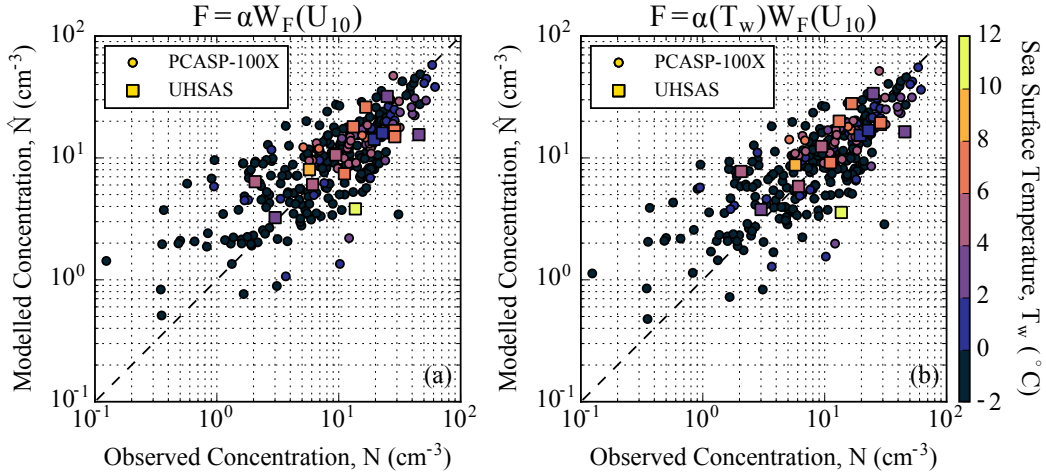
542 In Table 1 we have calculated the relative likelihood that a given surface flux  
 543 parameterization fit the data as compared to the best parameterization,  $W_F$ . We  
 544 used the relative probabilities in Table 1 to compare two parameterizations: for in-  
 545 stance, modelling the flux with  $W_F$  and a function of sea surface temperature was  
 546 only 60% as likely to optimally fit our data as using  $W_F$  alone. We also found that  
 547 regardless of the surface flux parameterization, the optimal simulation length,  $t_0$  was  
 548  $48 \pm 3$  hours. This is similar to the “filling time” Ovadnevaite et al. (2014) used to  
 549 characterize surface fluxes of SSPs from their measurements in the North Atlantic.  
 550 The filling time is a characteristic timescale used in the Statistical Wet Deposition  
 551 Method for determining sea spray fluxes from a concentration time series (Lewis &  
 552 Schwartz, 2004). Definition of the filling time varies by author. In Ovadnevaite et al.  
 553 (2014), they interpret the filling time as “...the time between the cyclone formation  
 554 and subsequent arrival to [the measurement location]” instead of “the time since the  
 555 last precipitation event as considered in (Lewis & Schwartz, 2004)”. The filling time  
 556 we determined is consistent with the average time that elapsed between the passage  
 557 of seven separate cyclones we encountered throughout March, 2018. These cyclones  
 558 provided widespread high winds and boundary layer cloud, resulting in high fluxes,  
 559 but relatively short lifetimes for any suspended particulate. Hence, our finding is  
 560 consistent with the definition of filling time given by Ovadnevaite et al. (2014).

561 According to the AIC, the best parameterization for the surface flux of SSPs,  
 562  $F$ , used the fetch parameterization for whitecaps,  $W_F$  (Snyder & Kennedy, 1983; Xu  
 563 et al., 2000):

$$\begin{aligned}
 \frac{\partial f}{\partial \log D_p} &= \frac{F}{\sqrt{2\pi} \log(2)} \exp \left[ -\frac{\log^2 \left( \frac{D_p}{0.4} \right)}{2 \log^2(2)} \right] \\
 F &= \alpha W_F(U_{10}) \\
 \alpha &= 3.6 \times 10^7 \\
 W_F(U_{10}) &= 1 - \Phi \left( \frac{6.5}{\sqrt{U_{10}}} \right)
 \end{aligned}
 \tag{11}$$

566 where  $D_p$  is the particle diameter in  $\mu\text{m}$  at a relative humidity of 80%.





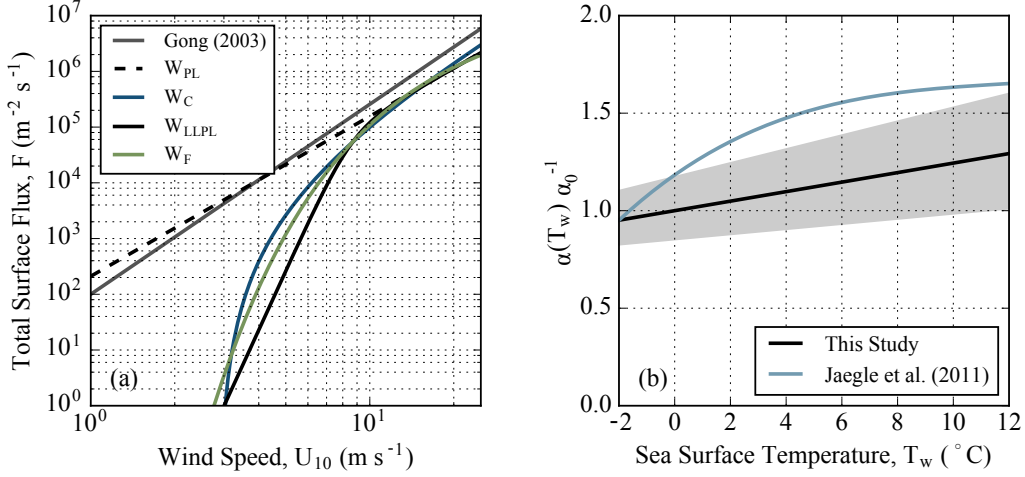
**Figure 6.** (a) Predicted concentrations of sea spray generated particles from the best parameterization of surface flux are compared to measurements from two observation platforms: a PCASP-100X aboard the R/V *Tangaroa* and the UHSAS onboard HIAPER. The best parameterization for the surface flux,  $F$ , of these particles was a function of the wind speed over the ocean surface. Each model–measurement pair is colored according to the average SST, weighted by the near-surface residence time. (b) As in (a), but for a parameterization of surface flux which incorporated a linear function of SST in addition to the wind speed dependence.

567 In Fig. 6a we show the model–measurement residuals for the best parameter-  
 568 ization of surface flux. The model–measurement residuals have been color-coded  
 569 according to the average SST. The average values of SST were weighted by the two-  
 570 day, near-surface residence time. On average, the predicted concentrations did not  
 571 appear to be biased positive or negative. However, a recent study by Jaeglé et al.  
 572 (2011) showed that particle fluxes may significantly depend on SST. Changes in SST  
 573 result in changes to the water viscosity. This is thought to modify the length of time  
 574 for the whitecap to dissipate,  $\tau$ , which is part of the constant,  $\alpha$ . Hence, in order to  
 575 compare to their result, we fit a linear correction term for the constant  $\alpha$  presented  
 576 in Eq. (11) as a function of the SST,  $T_w$ , finding:

$$577 \alpha(T_w) = 3.6 \times 10^7 (1 + 0.024T_w) \quad (12)$$

579 The model–measurement residuals of the temperature-corrected parameterization,  
 580 are shown in Fig. 6b. From Table 1 we can see that this did not substantially  
 581 improve model fidelity. In both Figs. 6a and b we have also shown the model–  
 582 measurement pairs for the SOCRATES observations within the boundary layer.  
 583 Since these measurements were not included within the regression framework, the  
 584 good agreement in both of these figures provides a measure of confidence that  
 585 Eq. (11) produces reasonable results within the Southern Ocean region.

586 In Fig. 7a we show the predicted flux of SSPs from each of the models in  
 587 Eq. (6) as a function of near-surface wind speed. The total particle flux predicted  
 588 by Gong (2003) has also been shown for reference. In Fig. 7b we have compared the  
 589 linear function of SST we recovered from the regression analysis to the polynomial  
 590 function fit by Jaeglé et al. (2011). We have shifted the values of the polynomial  
 591 function so that it matches our linear function at  $T_w = -2^\circ\text{C}$ . The slope of our linear  
 592 function predicts that SST is not as significant a control of SSP surface flux as  
 593 shown by Jaeglé et al. (2011). This may be a result of the distribution of our mea-



**Figure 7.** (a) The best parameterization of the total surface flux of sea spray generated particles,  $W_F$ , is compared to calculations from the Gong (2003) parameterization. (b) The linear bias correction function found by the regression is compared to the function reported by Jaeglé et al. (2011), where their function has been shifted vertically to match the linear bias correction function at  $T = -2^{\circ} \text{C}$ . The shaded region shows how uncertainty in the retrieved regression parameters propagated to prediction uncertainty.

594 measurements across different SST regimes since 78% of our observations were related  
 595 to waters in a narrow temperature range ( $-2$ – $0^{\circ}\text{C}$ ). Hence, there would be little im-  
 596 provement to either the NSE or AIC for these samples. Still, as shown in Fig. 7b,  
 597 the bias correction curve presented by Jaeglé et al. (2011) is clearly outside of the  
 598 uncertainty bounds for the modest temperature dependence we observe within our  
 599 dataset.

600 Finally, we compared the Nash-Sutcliffe model efficiency coefficient (NSE) for  
 601 the best parameterization we found within our regression framework to two differ-  
 602 ent parameterizations of SSP surface flux. The NSE is generally equivalent to  $R^2$ ,  
 603 but can also become negative when the average observed concentration provides a  
 604 better fit to the data than the proposed parameterization. From Table 2 it is clear  
 605 that Eq. (11) predicted concentrations of SSPs that were more consistent with our  
 606 observations than predictions from either the Gong (2003) or the Jaeglé et al. (2011)  
 607 parameterizations. Comparisons showed that the Gong (2003) parameterization  
 608 produced too many SSPs at all wind speeds.

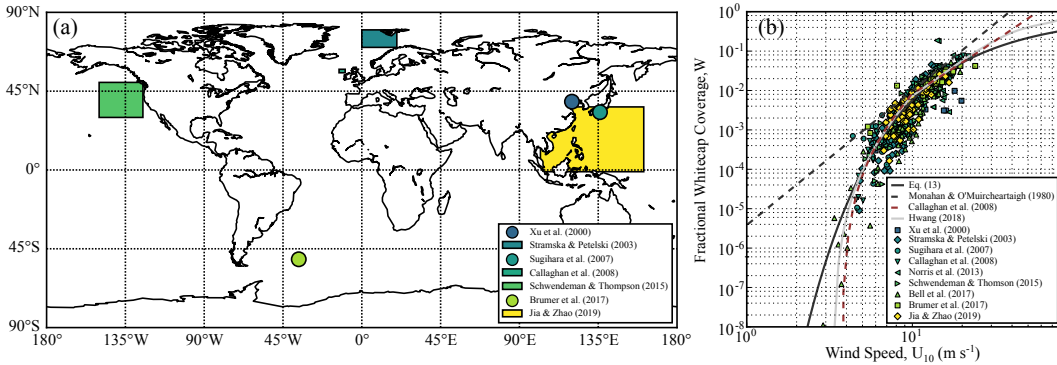
609 For reference, we also performed the regression for the entire five-day simula-  
 610 tion. In all of the parameterizations presented, the NSE decreased significantly for  
 611 the longer simulation, consistent with our hypothesis that in-cloud droplet activation  
 612 was not accurately simulated. In addition, surface fluxes predicted by  $W_F$  when  
 613 constrained by the five-day simulations were strictly smaller than surface fluxes pre-  
 614 dicted by  $W_F$  when constrained by the two-day simulations, for all wind speeds.  
 615 Therefore, our finding that the parameterization of Gong (2003) over-predicted the  
 616 surface flux of SSPs was robust.

### 617 3.5 Meta-Analysis of Whitecap Data

618 In order to assess the conditions under which the parameterization presented  
 619 above may be applicable, we analyzed global whitecap data from the literature. In

**Table 2.** The Nash-Sutcliffe model efficiency coefficient between the number concentration of SSPs predicted by a given surface flux parameterization and the observation conducted aboard the R/V *Tangaroa*. The time,  $t_0$ , is the length of time for which the Lagrangian particle dispersion parameterization simulated the movement of SSPs back in time. A negative value for the NSE implied that the mean of the observations was better at predicting the observed variance than the given parameterization, whereas a value of 1 would imply a perfect model-measurement fit.

Surface Flux parameterization	NSE ( $t_0 = 48$ hours)	NSE ( $t_0 = 120$ hours)
(Gong, 2003)	<0	<0
(Jaeglé et al., 2011)	0.22	<0
Eq. (11)	0.67	0.52



**Figure 8.** (a) The spatial extent of shipborne and tower whitecap observations within the database assembled for the whitecap meta-analysis. Studies where the coordinates of the observations were not specified all took place in the North Atlantic. (b) The fractional coverage of the sea surface by whitecaps as a function of 10 m wind speed.

620 Fig. 8b we have amalgamated 527 in situ ship-borne and tower observations of the  
 621 surface coverage of whitecaps as a function of the 10 m wind speed,  $U_{10}$  (Xu et al.,  
 622 2000; Stramska & Petelski, 2003; Sugihara et al., 2007; A. Callaghan et al., 2008;  
 623 Norris et al., 2013; Schwendeman & Thomson, 2015; Bell et al., 2017; Brumer  
 624 et al., 2017; Jia & Zhao, 2019). Studies published after the year 2000 were used since  
 625 they all employed some form of automated image processing. This meant that each  
 626 whitecap measurement was a result of  $>10^2$  images, a necessary minimum to have  
 627 a convergent mean (A. H. Callaghan & White, 2009). The spatial coverage of these  
 628 studies is shown in Fig. 8a, indicating that there is a good degree of coverage across  
 629 surface temperature regimes within the database. In Fig. 8b we have shown three  
 630 parameterizations for the whitecap coverage from the literature overlying the in  
 631 situ measurements (Monahan & Ó Muircheartaigh, 1980; A. Callaghan et al., 2008;  
 632 Hwang, 2018). We used the AIC to compare the existing parameterizations shown  
 633 in Fig. 8b to the fetch model, which was fit to the data via non-linear least-squares  
 634 regression. We found that the fetch model for whitecap development captured the  
 635 variability in the database best, with the relative likelihood that the other models  
 636 accurately captured the variability being  $<10^{-3}$ . It also did so with a continuous  
 637 function, whereas the other models were piece-wise. The best fit for the whitecap  
 638 data was as follows:

$$639 \quad W_F = 1 - \Phi \left( \frac{6.2}{\sqrt{U_{10}}} \right) \quad (13)$$

640 We also sought to validate the dependence of SSP fluxes on SST. However, SST data  
 641 within the works cited were either not included or simply summarized as a range  
 642 of values encountered. Still, most voyages made their measurements within a fairly  
 643 narrow SST band. Hence, we could test whether or not there was any dependence  
 644 by using a voyage-average SST for each study. Using the AIC as a measure of the  
 645 goodness of fit, we found that SST did not improve the regression. This is evident  
 646 by visually comparing the observations of Jia and Zhao (2019), which took place in  
 647 extremely warm seas ( $T_w \sim 28^\circ\text{C}$ ), to the rest of the data points.

## 648 4 Discussion

### 649 4.1 Meteorological Measurements

650 In the previous section, we presented total number concentrations of SSPs  
 651 within the Southern Ocean marine boundary layer as measured from two separate  
 652 measurement platforms. These were compared to estimations of the total number  
 653 concentration of SSPs derived from a source–receptor analysis. To assess the validity  
 654 of the meteorological fields used within the source–receptor analysis, we compared  
 655 the near-surface winds forecast by AMPS to our observations aboard the R/V *Tan-*  
 656 *garooa*. We found that the near-surface winds forecast by AMPS compared favorably  
 657 to our observed winds with respect to both magnitude and direction. Previous stud-  
 658 ies have found large biases between AMPS forecasts and the true, local winds over  
 659 the complex coastal topography of the Antarctic coastline (Bromwich et al., 2005;  
 660 Jolly et al., 2016). However, ocean waves have much less surface roughness in com-  
 661 parison to the coastal topography of Antarctica, which would suggest that this may  
 662 not be as substantial an issue over the ocean.

663 While the precipitation fields forecast by AMPS did not correlate well with  
 664 our measurements, the climatological distribution of precipitation events within  
 665 AMPS was consistent with our observations. However, within FLEXPART-WRF  
 666 a lack of precipitation “trickled up” to the cloud layer: in the current version of  
 667 FLEXPART-WRF, clouds are only present within a simulation if they are precip-  
 668 itating. As a result, scavenging of SSPs from in-cloud activation was likely poorly  
 669 modeled within the AMPS–FLEXPART-WRF framework. As our own observational  
 670 record showed, SSPs were strongly scavenged by boundary layer cloud (e.g. fog),  
 671 particularly through February 15–17<sup>th</sup> and March 4–6<sup>th</sup>, 2018. It is useful, how-  
 672 ever, to recall that clouds over the Southern Ocean are not very well represented  
 673 within modern atmospheric models (Trenberth & Fasullo, 2010; Schuddeboom et  
 674 al., 2019). Current era atmospheric models systematically under-predict the amount  
 675 of low-lying cloud and fog relative to the true cloud observed over the Southern  
 676 Ocean (Kuma et al., 2019). Hence, even if a state-of-the-art microphysical param-  
 677 eterization of in-cloud scavenging had been present in FLEXPART-WRF, it is likely  
 678 that the scavenging of sea spray still would have been under-estimated.

### 679 4.2 Source–Receptor Modelling

680 Once we had established that there was a missing sink of sea spray within our  
 681 source–receptor framework, it was necessary to decouple this sink from each of the  
 682 parameterizations of surface flux we tested within the regression analysis. This was  
 683 accomplished by allowing the simulation length to vary as a free parameter within  
 684 the regression analysis. The simulation length can be interpreted as the average  
 685 length of time since a boundary layer in-cloud activation event (e.g. fog or low-  
 686 cloud). From Table 2 it is evident that our observations were better reproduced for  
 687 a fixed simulation length of two days, rather than the five days originally simulated.  
 688 This agreed well with the “filling time” of 1.5–2 days used by Ovadnevaite et al.  
 689 (2014) to constrain the surface flux of sea spray in the North Atlantic. While setting

690 a voyage-wide simulation time may have been a gross approximation, it was likely  
 691 the only approach in light of the present systematic cloud biases over the Southern  
 692 Ocean.

### 693 4.3 Regression Analysis

694 By constraining the missing sink of sea spray within our model framework, we  
 695 could finally compare how well the near-surface wind speed and SST predicted our  
 696 observational record. We found that the fetch parameterization presented by Xu  
 697 et al. (2000) and Snyder and Kennedy (1983) performed the best in our compar-  
 698 ison as measured by the Nash-Sutcliffe model efficiency coefficient, NSE, and the  
 699 Akaike Information Criterion, AIC. The parameterization,  $W_F$ , is so-named since  
 700 the parameter  $c_1$  is a function of the fetch. In our analysis we have assumed that  
 701 this parameter was constant, since fetch does not significantly influence the degree  
 702 of whitecapping in open seas (Hsu, 1986; Piazzola et al., 2002). Later, when we  
 703 performed a similar regression analysis with a database of whitecap coverage ob-  
 704 servations, we found a slightly smaller value for  $c_1$ . When we compared Eq. (11)  
 705 to Eq. (13), we found that  $c_1$  retrieved from the whitecap regression was  $6.2 \pm$   
 706  $0.2$ , which was consistent with the value of  $6.5 \pm 0.2$  we retrieved from the SSP re-  
 707 gression. The sensitivity of  $W_F$  means that Eq. (11) will under-estimate whitecap  
 708 coverage globally and subsequently result in under-estimations of sea spray fluxes.  
 709 However, we can compare to the value for  $c_1$  retrieved when we only consider South-  
 710 ern Ocean whitecap data from Brumer et al. (2017) ( $c_1 = 6.4 \pm 0.1$ ). Combined  
 711 with the goodness of fit to the SOCRATES data (Fig. 7a), this provides a secondary  
 712 measure of validation for the parameterization over the Southern Ocean. We can  
 713 only conclude that for a global study, a value for  $c_1$  of 6.2 may be more appropriate.  
 714 For Southern Ocean specific studies a value for  $c_1$  of 6.5 should be used.

715 Finally, we compared results from two other parameterizations for the surface  
 716 flux of SSPs to our observations. We found that neither the Jaeglé et al. (2011)  
 717 nor the Gong (2003) parameterization could predict the concentration of SSPs we  
 718 observed over the Southern Ocean as well as Eq. (11). This is connected to how  
 719 the Gong (2003) parameterization (which Jaeglé et al. (2011) re-scaled) scales the  
 720 surface flux of SSPs with increasing wind speed. Within this parameterization,  
 721 the scaling is estimated via a power-law relationship between the surface coverage  
 722 of whitecaps and near-surface wind speed (Monahan & Ó Muircheartaigh, 1980).  
 723 However, as we show in Fig. 8b, the parameterization presented by Monahan and  
 724 Ó Muircheartaigh (1980) results in consistent over-estimations of the whitecap cov-  
 725 erage. These over-estimations propagate through the SSP flux parameterization  
 726 of Gong (2003) and lead to the over-estimations in concentrations we observe. In  
 727 addition, the power-law predicts that there will always be a flux of sea spray from  
 728 the ocean surface, despite it being well-established that whitecaps do not form until  
 729 the wind speed over the ocean exceeds  $3\text{--}4 \text{ m s}^{-1}$ . Even the re-scaled Gong (2003)  
 730 parameterization presented in Jaeglé et al. (2011) did not match our observations  
 731 well, either.

### 732 4.4 The Effect of Sea Surface Temperature

733 To understand differences between whitecapping in different regions, previous  
 734 research has focused on wave parameters and SST. As Sugihara et al. (2007) and  
 735 Goddijn-Murphy et al. (2011) have shown, there is a marked difference between ob-  
 736 servations of whitecaps in a pure windsea vs. a swell dominated sea. Indeed, when  
 737 we fit  $W_F$  to the whitecap data from Sugihara et al. (2007) we retrieved a value  
 738 for  $c_1$  of  $6.1 \pm 0.1$  in a pure windsea (indicating higher spatial coverage of white-  
 739 capping) compared to  $6.7 \pm 0.1$  when the winds were following swell or counter  
 740 swell (indicating lower spatial coverage of whitecapping). This could potentially

741 explain the difference between the value of  $c_1$  we retrieved from our measurements  
 742 and the one retrieved from the entire whitecap database. However, conversely, in a  
 743 satellite-derived whitecap database, Albert et al. (2016) found that whitecaps were  
 744 not dependent on wave parameters, but were actually modestly dependent on SST.  
 745 They noted that the lack of dependence on wave parameters may have been a result  
 746 of using wind history as a proxy for wave age and spatial averaging. However, we  
 747 found that there was no dependence on SST within the database of in situ whitecap  
 748 observations.

749 Of course, even if SST does not affect the fractional coverage of whitecaps,  
 750 it can still affect the surface flux of particles through changes to viscosity. Re-  
 751 sults from laboratory studies are mixed: while two studies have clearly shown that  
 752 the surface flux of sea spray should increase in warmer waters (Mårtensson et al.,  
 753 2003; Sellegri et al., 2006), others found that that differences in seawater compo-  
 754 sition (A. H. Callaghan et al., 2014) and wave characteristics (A. H. Callaghan et  
 755 al., 2012) could be much more important. Other laboratory results have even shown  
 756 that increases in water temperature led to decreases in sea spray fluxes (Zábori et  
 757 al., 2012). To test whether or not changes in SST affected our own observations, we  
 758 used SST as a second independent variable within the regression analysis. We found  
 759 that the impact to the model-measurement fit was more modest than predicted by  
 760 Jaeglé et al. (2011) (see Fig. 7b), and that the parameterization which only used  
 761 wind speed (Eq. (11)) performed just as well. As we noted, this may have been a  
 762 result of making observations in a very narrow range of SSTs, which would result  
 763 in very small changes to the regression metrics we analyzed. However, from Fig. 6b  
 764 we can see that the model-measurement residuals don't appear to be significantly  
 765 biased from the 1:1 line at warm temperatures.

766 Ultimately, we should be cautious when implementing temperature correction  
 767 functions for SSP fluxes. Consider that field observations have already clearly shown  
 768 that the presence of swell inhibits the surface fraction of whitecaps for a given wind  
 769 speed (Sugihara et al., 2007). Hence, global climatologies of swell could potentially  
 770 explain the latitudinal trends in SSP flux expected by (Jaeglé et al., 2011): swell  
 771 rarely occurs in the tropics (where fluxes are expected to be higher), whereas swell  
 772 frequently is present at high latitudes (where fluxes are expected to be lower) (Jiang  
 773 & Chen, 2013). As a result, the resulting bias correction curve Jaeglé et al. (2011)  
 774 derived from in situ and satellite observations of aerosol may be partially dependent  
 775 on the presence (or absence) of swell. Yet, the curve attributed the latitudinal varia-  
 776 tions in flux necessary to fit their observations completely to variations in SST. Our  
 777 own observations, which were made exclusively in the presence of swell, showed that  
 778 the dependence of SSP fluxes on SST was much weaker than anticipated by Jaeglé  
 779 et al. (2011), leading to a very negligible effect on model performance. We conclude  
 780 that a more comprehensive global study of sea spray which fully controls for upwind  
 781 wave and SST conditions is needed in order to decouple these two effects. In the in-  
 782 terim, models should be cautious in implementing functions which could potentially  
 783 over-exaggerate radiative feedback loops.

#### 784 4.5 The Direct Radiative Effect

785 As the goal of this study was to understand how SSPs might influence the lo-  
 786 cal radiation budget, it would be useful to evaluate whether or not changes to the  
 787 parameterization of SSP fluxes result in substantial changes within existing climate-  
 788 chemistry models (CCMs). A recent study comparing the winter-time AOD over  
 789 the Southern Ocean found that current era parameterizations (e.g. Gong (2003))  
 790 of sea spray within a climate-chemistry model (CCM) resulted in over-estimations  
 791 of the AOD relative to satellite observations (Revell et al., 2019). However, in the  
 792 austral summer, the opposite was observed: namely, a lack of particles formed from

793 the nucleation of sulfate-gasses resulted in under-estimates of AOD. Within this  
 794 same study, the parameterization of surface flux,  $W_{PL}$ , was implemented within  
 795 the CCM to better constrain the contribution of SSPs to the total particle popula-  
 796 tion (Revell et al., 2019). While  $W_{PL}$  was not the best function determined by this  
 797 work, it was similar in form to the Gong (2003) currently implemented within the  
 798 CCM being studied, so it was an easy substitution. Since  $W_{PL}$  had a Nash-Sutcliffe  
 799 coefficient of 0.6 it also produced results that were consistent with Eq. (11). It is  
 800 important to note that  $W_{PL}$  was used to re-scale the size distribution of the Gong  
 801 (2003) parameterization, so any changes would be related to differences in the scal-  
 802 ing function and not to differences between size distributions. Results conclusively  
 803 showed that the more conservative estimates of the surface flux of SSPs generated  
 804 by  $W_{PL}$  completely removed the bias in winter-time AOD that was previously ob-  
 805 served. Therefore, we are confident that the parameterization for the surface flux of  
 806 SSPs presented in Eq. (11) will result in better predictions of the abundance of SSPs  
 807 within the Southern Ocean region. More importantly, Revell et al. (2019) show that  
 808 it helps disentangle the potential compensating errors in predicting the AOD for  
 809 studies interested in the more complicated gas phase and aqueous phase chemistry  
 810 which produces sulfate particles from volatile marine precursors like dimethylsulfide.

811 As we have emphasized throughout this study, the MBL over the South-  
 812 ern Ocean region is home to the strongest surface winds over open ocean on  
 813 Earth (I. Young, 1999). Surface winds also appear to be getting stronger: at Mac-  
 814 quarie Island, winds have increased in intensity by  $3 \text{ cm s}^{-1}$  per year from 1973–  
 815 2011, with satellite data showing that winds over the Ross Sea increased by 0.5–1%  
 816 through 1991–2008 (Hande et al., 2012; I. R. Young et al., 2011). Within the Ross  
 817 Sea region, this increase is related to the deepening of the Amundsen Sea low, an  
 818 area of climatologically low pressure in the Southern Ocean which influences regional  
 819 winds, sea-ice extent and temperature (Coggins & McDonald, 2015; Raphael et al.,  
 820 2016). As we show throughout this study, sea spray has a highly non-linear rela-  
 821 tionship with wind speed. Given their large contribution to the CCN population  
 822 (10–65%; Quinn et al. (2017)), AOD (Murphy et al., 1998; Revell et al., 2019), and  
 823 cloud phase (McCluskey et al., 2018) over the Southern Ocean, these particles can  
 824 have a significant buffering effect on the local climate. We would therefore encourage  
 825 future studies interested in climate projections for the Southern Ocean to make use  
 826 of Eq. (11) when predicting the surface flux of sea spray generated particles.

## 827 5 Conclusions

828 In this study, we described and optimized an existing parameterization for  
 829 the surface flux of sea spray generated particles (SSPs) based on the 10 m wind  
 830 speed in Eq. (11). Within our regression framework we found that the dependence  
 831 of SSP fluxes on SST was very weak in the temperature range of our observations  
 832 ( $T_w < 12 \text{ }^\circ\text{C}$ ) and that it did not help to constrain additional variability in our  
 833 data set. An external database of previously published whitecap observations was  
 834 exploited to test the parameterization we used in this analysis and found no tem-  
 835 perature dependence at all. While others have shown that temperature-dependent  
 836 flux parameterizations seem to explain known latitudinal variations in SSP flux, the  
 837 correction functions derived from such an analysis could potentially be a proxy for  
 838 latitudinal variations in wave characteristics. Given the potential links between SSPs  
 839 and the Southern Ocean radiation budget, we should be cautious to add feedback  
 840 loops where none may exist.

841 Finally, the parameterization presented in this study is already being used  
 842 to model the AOD and concentration of CCN in the region. Research has shown  
 843 that the new parameterization vastly improved regional calculations of AOD, com-  
 844 pared with previous parameterizations which over-predicted the surface flux of

845 SSPs (Revell et al., 2019). We recommend that studies interested in aerosol–cloud  
 846 interactions implement the parameterization as it has been shown to better con-  
 847 strain the contribution of SSPs to the CCN population.

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