Response to reviewer comments for manuscript "Observations of high droplet number concentrations in Southern Ocean boundary layer clouds"

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$_{\circ}$ 1 Anonymous Referee #1

7 Received and published: 14 October 2015

Review of Observations of high droplet number concentrations in Southern Ocean
boundary layer clouds by Chubb et al.

¹⁰ Recommendation: Requires minor revision before acceptance in ACP.

11 **1.1 General Comments**

This paper presents some interesting observations from microphysical probes and the 12 UHSAS aboard the NSF G-V aircraft during the HIPPO project from over the Southern 13 Ocean (SO). Analyzing data from one case study of boundary layer clouds sampled during 14 15 the wintertime, the authors show that the observed cloud droplet number concentrations and sub-micron aerosol concentrations observed in the southern most profiles were 16 exceptionally high compared to expectations given background aerosol concentrations 17 in this region. By combining their data with some chemistry observations and back 18 trajectory analysis, the authors show that although there was some evidence of continental 19 influence for the profiles, the data and trajectories are not consistent with the long range 20 transport of continental aerosols explaining the observed cloud and aerosol concentrations. 21 Thus, they infer that the high surface winds were most likely responsible for the high 22 observed concentrations. 23

Given the paucity of observations over the Southern Ocean and the contradictions from 24 some previous studies that surface winds were not necessarily correlated with sea salt 25 aerosol production, I certainly believe that this paper should be published. Even though 26 a fairly limited data set is presented in the study, the results are of sufficient merit 27 that they should guide future studies and in fact, should motivate further observations 28 in this region to better explore the relationship between cloud and aerosol properties. 29 Nevertheless, there are a few changes which I suggest should be incorporated into the 30 manuscript to better improve the flow of the manuscript and to better emphasize that 31 the limitations in the data mean that that their results are consistent with the high 32 surface winds causing the observed concentrations rather than proving that the high 33 surface winds cause these concentrations. 34

Author Response: Thank you for taking the time to provide a thoughtful review of our paper. We will address the comments below point by point. There is a version of the revised manuscript with changes tracked since the original submission accompanying this

38 response.

I think the paper could be shortened and improved if Section 5 on the evaluation of 39 uncertainties was incorporated into the sections of the manuscript where the relevant 40 results were described earlier. When I was reading through the manuscript for the 41 first time, I was wondering about some of the issues introduced in Section 5 and how 42 they affected the presented analysis. If this material was explained (before or at the 43 same time) as the relevant results, it will be much easier for the reader to interpret the 44 observations and trajectories. Right now, for example, the basis of the calculation of the 45 back trajectories are presented in Section 2.4, the back trajectories themselves described 46 in Section 4.1, and the uncertainties in Section 5.3. There is necessarily some repetition 47 in the manuscript because these calculations are repeated three times. Thus, the paper 48 could be made much more tight if the back trajectories were only discussed in Section 4.1 49 (with maybe a quick introduction that they will be considered in Section 2). Similarly, 50 the uncertainties in the CDP (Section 5.1) and UHSAS (Section 5.2) should be described 51 in Section 2.2 so that the analysis of the flight level data in Section 3 can be better 52 interpreted. 53

Author Response: Thank you for this comment. We respectfully disagree with your 54 suggestion of working the discussion of the uncertainties into the results section. We have 55 confidence in the data and were forthright with the discussion of the uncertainties in 56 section 5, which we referred to in the description of the instrumentation and in the results 57 section itself. Discussing the uncertainties alongside the results would be distracting, 58 and the argument that we have presented is already complex enough. However, we agree 59 that the discussion about the uncertainties in the back trajectories in particular was 60 somewhat repetitive and there was an opportunity to make the manuscript more concise. 61

In response, section 5.3 (Uncertainties in back trajectories), which was a general discussion
of the uncertainties in the use of back trajectories, has been removed and the content
from there was worked in to section 2.4 (Calculation of back trajectories). Overall this
resulted in a reduction of about 250 words, so it was clearly worthwhile.

We have left sections 5.1 and 5.2 in place, with minor modifications to address various
reviewer comments. There was no scope to significantly reduce the overall length of
the manuscript by moving these elsewhere. In addition, both of these sections included
details specific analysis that we wanted to keep separate from our results.

My second major comment can be best described by reviewing the final sentence of the 70 manuscript, namely we conclude that local production of sea spray aerosol through the 71 high winds in the southernmost regions of the flight is the most likely explanation for 72 these observations. I think it would be better to state that the observations are consistent 73 with the high winds causing the production of the sea salt aerosol, because this is really 74 inferred from the data rather than establishing a relationship between these variables. I 75 think this change in language is needed because the authors do admit that there is some 76 uncertainties in the trajectory analysis. 77

Author Response: Thank you for this comment. Even though there are some uncertainties in our analysis, in part due to missing data and in part to the absence of instrumentation that would make the argument unequivocal, we believe that we have presented a strong case for our the hypothesis that the elevated aerosol and droplet concentrations are due to sea spray aerosol. However, comments from both Reviewer #1 and Reviewer #2 requested a dilution of the language, so we have changed section 7 (the only paragraph): In this paper, ... (*snip*) ... We conclude that <u>these observations are</u> <u>consistent with the</u> local production of sea spray aerosol through the due <u>to</u> high winds in the southernmost regions of the flightis the most likely explanation for these observations. ... (*snip*)

One other thing that would be nice to add to the manuscript is a description of how often the unusual winter-time microphysical conditions in the boundary layer over the Southern Ocean occur. Other flights are given a cursory inspection to determine how often the pollutants might be present over the Southern Ocean, but can any comments be made about how often the gale force winds might be expected in the boundary layer?

Author Response: Thank you for this comment. The occurrence of strong winds over the Southern Ocean has received some attention in the literature, which we do address (e.g. references to Korhonen et al., 2010; Hande et al., 2012a). In addition, we specifically commented in the original manuscript (P. 25515, lines 8–10) on how often gale force winds occurred at Macquarie Island, where there is a weather station operated by the Australia Bureau of Meteorology. Upon reflection, this sentence would be better placed in the discussion, so we have made the following changes to the manuscript:

1. Edited section 3.1 (final sentences of this section):

The boundary layer wind speeds ... (snip) ... Using a log scaling law to translate this to surface conditions, the ten meter winds would have been in the range of 17 to 20 m s^{-1} . Gale force winds speed gale force $(\geq 17 \text{ m s}^{-1})$ occur regularly over the SO; weather station data from Macquarie Island, which is nearby in the storm track region, had half hourly average surface wind speeds greater than this on about 15% of days between 2008 and 2011.

¹⁰⁸ 2. Edited section 6.2 (moved the discussion from 3.1 to here):

109This result is of interest ... (snip) ... over the SO. Strong boundary layer110Gale force winds, such as those encountered in HIPPO-4 RF06, are111a regular occurrence occur regularly over the SO...; weather station data112from Macquarie Island, which is nearby in the storm track region, had113half hourly average surface wind speeds greater than this on about 15%114of days between 2008 and 2011. Moreover, ... (snip)

¹¹⁵ In terms of the microphysics data and the uncertainties, I was surprised that there were ¹¹⁶ no statements about how the bulk water content derived from the size distributions ¹¹⁷ compared to that measured by a bulk water probe. I am assuming there must have been ¹¹⁸ some sort of King or hotwire probe on the G-V. This would be a basic test that could help ¹¹⁹ verify that the CDP is well calibrated (especially since some of the channel boundaries ¹²⁰ can sometimes be shifted). Can this be done and added to uncertainty analysis section?

Author Response: Thank you for this comment. There was indeed a PMS-King type hot wire probe installed on the GV, and it was operational during HIPPO-4 RF06, and 122 naturally compared the values derived from this instrument with those from the CDP. The two values were highly correlated (R = 0.98) but initially the CDP values were 124 approximately twice that of the King probe. This was in spite of the standard calibra-125 tion using glass beads during the HIPPO-4 campaign and subsequent post-processing 126 Romashkin (2012). This was highly concerning for us and the resultant investigation led to the beam mapping of CDP #16, which was the one used in HIPPO-4. The beam 128 mapping is a relatively new technique which evaluates the true sample area of the specific 129 instrument (as opposed to the "theoretical" sample area which had been used previously) 130

with water droplets. This was performed by DMT, the instrument manufacturer, who
 were intentionally kept unaware of the discrepancy that we had identified.

The original data were processed using a theoretical sample area of $0.240 \,\mathrm{mm^2}$, but the

¹³⁴ subsequent beam mapping of CDP #16 showed that the sample area was $0.309 \,\mathrm{mm^2}$.

¹³⁵ We recalculated the droplet concentration and liquid water content using the measured

136 beam area for this paper.



Data from profile 1 plotted against altitude (ma.s.l.). Left: LWC derived from various instruments (colored lines), with adiabatic (solid gray) and 0.68 times adiabatic (dashed gray). The shaded region indicates where the cloud was sub-adiabatic, probably due to entrainment from above at the boundary between two overturning cells. Right: temperature and dew-point temperature (colors) and lifted parcel (gray lines).

As mentioned in the original manuscript, we evaluated the CDP liquid water content against theoretical values for profile 1, which we believe to be close to truly adiabatic (see figure). We used a parcel model (conserved θ_e) initialized near cloud base to calculate a theoretical LWC profile. If the values in the shaded region—where there appears to be entrainment of dry air from above—are excluded, a very good match with the CDP data is obtained ($\rho_{L,CDP} = 1.01\rho_{L,Adiabatic}$). The King probe appears to be measuring about 68% of the adiabatic amount according to this analysis, although this is not outside the range of possibilities for stratocumulus clouds (see Boers et al., 1996).

Finally, we note the comments of Romashkin (2012) pertaining to the use of the PMS-King
 probe data on the GV during HIPPO-4:

147 Significant improvements have been made to the King probe processing code

- to better quantify changes in the heat transfer related to the changes in
- the airspeed. However, rapid fluctuations in the PLWCC baseline are still

observed in the PLWCC that are not realistic. Please compare the PLWCC
data with PLWCD_* that is calculated from the cloud droplet probe to assess
the quality of the liquid water data from the King probe.

In conclusion, there are some uncertainties about the LWC data from both the CDP and the PMS-King probe on the GV and the true value was probably somewhere between the two. Assuming for a moment that the PMS-King probe were accurate, there could be two hypotheses to explain the difference in the CDP observations:

- The CDP sample area was even larger than the beam mapping analysis suggested,
 in which case then the cloud droplet number concentrations would have been affected too, resulting in a mean CDNC for profile 1 of about 32 cm⁻³. This is quite low but not impossible given previous observations.
- 2. There was a binning error resulting in systematic mis-sizing of the droplets. This 161 could affect $\rho_{L,CDP}$ without affecting the CDNC. Neither of these possibilities 162 change our conclusions in any way, because in the case of (1.), we would still need 163 to explain the factor of five (or more) difference in the CDNC for profiles 1 and 164 4, and in the case of (2.), the CDNC data are unaffected. Following our intensive 165 quality control of the CDP data we elected to present these in order that our results 166 could be fully reproducible. However, we agree that it is worth mentioning that 167 these analyses have been performed in the manuscript, in such a way that it does 168 not distract from our message. 169
- ¹⁷⁰ We have made the following changes to the manuscript:
- 171 1. Item added to section 2.2: to describe bulk water measurements:

172 173 174 175 176	• <i>PMS-King "hot-wire" probe.</i> Total cloud liquid water content can be directly measured by exposing a temperature-controlled element to the flow outside the aircraft (King et al., 1978). Within cloud, the power required to maintain a constant temperature is compared to that required in clear air to derive $\rho_{\rm L,King}$.
177	2. Sentences added to section 5.1:
178 179 180 181	The accuracy of the CDP is typically stated as $\pm 10\%$ due to uncertainties in the true sample volume and in the sizing of small particles through Mie scattering. However, the PMS-King probe consistently showed about 0.68 of ρ_L from the CDP. Using a parcel ascent (snip)
	Even if the CDP did significantly overestimate as which we believe to

- ¹⁸² Even if the CDP did significantly overestimate ρ_L —which we believe to ¹⁸³ be unlikely—it may have also overestimated N_C by the same fraction, ¹⁸⁴ depending on the cause of the error. However, this would ultimately ¹⁸⁵ have little impact on our conclusions, because it would still be necessary ¹⁸⁶ to explain a factor of five increase in N_C between profiles 1 and 4.
- ¹⁸⁷ 3. New bibliography item added for King et al. (1978).

188 1.2 Specific Comments

Abstract: standard cloud physics payload. Although there may be a standard payload
for the G-V, in general there are so many different cloud physics probe that there really
is no such thing as a standard payload. Recommend listing instruments.

Author Response: We'd prefer to leave the instrument list to section 2.2, but accept
 your comment about the terminology. The leading sentence of the abstract has been
 changed as follows:

- ¹⁹⁵ Data from the standard cloud physics payload Cloud physics data collected
- during the NSF/NCAR High-performance Instrumented Airborne Platform
- ¹⁹⁷ for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO)
- campaigns provide ... (snip)

¹⁹⁹ Page 25509, line 14, first word should be clouds rather than cloud.

200 Author Response: Accepted.

Page 25510, line 9. There are some uncertainties with the depth of field in 2DC probes,
especially for particles smaller than 125 micrometers (Baumgardner and Korolev 1997).
This should be commented upon when discussing the uncertainties for this probe.

²⁰⁴ Author Response: Accepted. Please note the following changes in the manuscript.

²⁰⁵ 1. Changes to section 2.2:

206	• Particle Measurement Systems (PMS) 2-D Cloud Imaging Probe
207	(2DC). Precipitation particles larger than about (snip)
208	individual particle images. Here, as for most applications of
209	the 2DC, we only use This type of probe is susceptible to
210	uncertainties in depth-of-field for particles with diameters less than
211	$200\mu\mathrm{m}$ (Baumgardner and Korolev, 1997), but we made no specific
212	correction for this other than only using particles with diameters
213	(snip)

214 2. New bibliography entry for (Baumgardner and Korolev, 1997).

Page 25510, lines 19-21: Given this calibration was done in 2015 and the HIPPO observations were obtained earlier, is this relevant to the presented observations? Was this sample area used in the computation of the microphysical quantities? Make clear.

Author Response: Thanks for this comment. The beam area is not expected to have changed in the interval between HIPPO-4 and the subsequent beam mapping. The beam mapping technique is relatively new and provides the best estimate of the true sample area that is available. As for the $\rho_{\rm L}$ and $N_{\rm C}$ data, we recalculated these ourselves using the new sample area. We have made this clear in the manuscript by adding a sentence to the item in section 2.2:

- Droplet Measurement Technologies (DMT) Cloud Droplet Probe (CDP). The CDP operates by ... (snip) ... beam mapping by the manufacturer in June 2015. We recalculated $\rho_{\rm L}$ and $N_{\rm C}$ using the updated sample area. Further discussion of the uncertainties associated with this instrument
- is provided in Sect. 5.1.

Page 25514, line 18 or so: How long of a horizontal distance was traveled during the time the profiles were obtained? To what degree could some horizontal inhomogeneity in the clouds be affecting the observed profiles?

Author Response: This is always a problem with aircraft data, and there is a trade-off to be made between artefacts due to high vertical speeds and inhomogeneities due to horizontal distance travelled. For HIPPO-4 RF06, the profiles were performed with a vertical speed of $7.5 \,\mathrm{ms^{-1}}$ at altitudes above 600 m, and $2.5 \,\mathrm{ms^{-1}}$ below this, at a true air speed of $130 \,\mathrm{ms^{-1}}$. The distance covered was about 38 km between 1500 m and 160 m (the lowest altitude reached) for each profile. However, as mentioned in the original

manuscript, there weren't significant differences between the ascending and descending 238 profile data, except in profile 2 where there was no cloud sampled in the ascending profile. 239 We do not anticipate major effects from this factor, but have highlighted these details in 240 the revised manuscript. Changes: 241

1. Inserted sentences in section 2.1 (final paragraph) to describe air speeds and vertical 242 motion: 243

244	The aircraft performed four descent/ascent profiles \dots $(snip)$ \dots cloud
245	top conditions are provided in Fig. 3. The mean true air speed varied
246	during the profiles, but it was consistently about $130 \mathrm{m s^{-1}}$ at altitudes
247	below 1500 m a.s.l. (well above the boundary layer). The vertical speed
248	of the HIAPER was about $7.5 \mathrm{ms^{-1}}$ for altitudes above $600 \mathrm{ma.s.l.}$
249	and $2.5\mathrm{ms^{-1}}$ below this. A total distance of about 38 km was covered
250	between the lowest level and 1500 m a.s.l. for each profile. Conditions
251	were quite varied between (snip)

2. Edited first paragraph of section 3.1 to highlight typical concerns with aircraft 252 profiles: 253

254	Figure 4 shows thermodynamic observations from each of the descend-
255	ing profiles from the 1 Hz dataset. The <u>slantwise nature of aircraft</u>
256	profiles leaves open the possibility of horizontal inhomogeneity limiting
257	the analysis, but the values for the ascending profiles were not (<i>snip</i>)

Page 25516, line 19: Would it be also useful to show/quote more of these maximum 258 values as well as the mean values in the plots? 259

Author Response: The maximum values for $N_{\rm C}$ were included for profiles 3 and 4 but 260 not for profiles 1 and 2 simply because there was much more variability in profiles 3 261 and 4. The maximum values are particularly important for profile 4 because $N_{\rm C}$ was 262 correlated with $\rho_{\rm L}$ for this profile only, indicating that entrainment was important. We 263 are reluctant to introduce more values in to the descriptions for profiles 1 and 2 because 264 they could be distracting, and the values could be read from the graphs if readers are 265 interested. 266

We have made no specific changes to the manuscript in response to this comment. 267

Page 25516, line 23-25: Could there be any influence (e.g., seeding) of the higher cloud 268 layers on the lower cloud layers that could complicate the observed trends? 269

Author Response: If you are referring to the cloud layer above 2400 m a.s.l. in profile 3, 270 we think that this is highly unlikely. There was no evidence of any precipitation particles 271 above the boundary layer cloud top, and there was a vertical displacement of nearly

272

1500 m between the cloud layers in this instance. 273

We have made no specific changes to the manuscript in response to this comment. 274

Page 25516, line 22: I assume that some of the observations of the UHSAS were obtained 275 at different humidities, resulting in different amounts of growth of particles. Could this be 276 affecting the comparison of concentrations at different flight legs? Were any corrections 277 made for this? 278

Author Response: This issue was also raised by Reviewer #2 and is addressed more 279 thoroughly in our response to their comments. In summary, due to the combined effect of 280 decelerating the air, anti-ice heating and optics block heating, it is fairly safe to assume 281 that the observed particle sizes are close to dry sizes. There is precedence for this in 282

the literature, and we have made this more clear in the manuscript with the following changes:

1. Added sentence to UHSAS item in section 2.2:

286	• DMT Ultra High Sensitivity Aerosol Spectrometer (UHSAS). The
287	UHSAS measures sizes of aerosol (snip). Due to the
288	combined effect of electrical anti-ice and internal heating, and
289	adiabatic heating of decelerated inlet air, we assumed that the
290	measured particle diameters were close to their dry diameters
291	(e.g. Blot et al., 2013; Kassianov et al., 2015) $(snip)$

292 2. Bibliography item added for Kassianov et al. (2015).

²⁹³ 2 Anonymous Referee #2

²⁹⁴ Received and published: 29 October 2015

²⁹⁵ 2.1 General comments

The paper presents data from one HIPPO flight over the Southern Ocean where high 296 aerosol number and cloud droplet concentrations were observed in the boundary layer. 297 Based on 3-day back trajectories, concentrations of CO and BC, and high wind speeds, 298 the authors conclude that the enhanced concentrations were likely due to sea spray 299 aerosol. However, direct evidence for this conclusion, e.g., aerosol composition measure-300 ments or thermal analysis, is lacking. In addition, as pointed out below, the CO and 301 BC supporting data are missing at some altitudes making the case for non-combustion 302 sources less certain. The only direct aerosol available for assessing the contribution of 303 sea spray aerosol to the high number and cloud droplet concentrations is the number 304 size distribution measured with the UHSAS. It should be possible to apply a lognormal 305 fit to these data and estimate the number concentration of sea spray aerosol as was 306 done by Modini et al., JGR, vol. 120, doi:10.1002/2014JD022963, 2015. Based on size 307 distributions generated in a wave tank (Prather et al., PNAS, 2013) and the canonical 308 number size distribution of sea spray aerosol defined by Lewis and Schwartz (2004), 309 Modini et al. fit a sea spray aerosol mode with the constraint of a 200 nm +/-30% mean 310 diameter and a geometric standard deviation between 2.5 and 3. They then integrated 311 the number concentration within that mode to estimate the number concentration of sea 312 spray aerosol. For comparison to that analysis, the UHSAS data in this paper would 313 have to be shifted to the same RH. I assume the data shown in Figure 6 are at ambient 314 RH. If they are at ambient RH and they are shifted to a dry diameter, the peak diameter 315 of the mode would be smaller than previously reported SSA size distributions. This 316 analysis would help to assess whether the measured aerosols were primary marine aerosol. 317 The paper should be published because there is a lack of in-situ aerosol and cloud data 318 over the Southern Ocean. That said, the above size distribution analysis should be 319 performed to assess the potential contribution of sea spray aerosol to the total number 320 concentration. In addition, given the lack of direct evidence and the uncertainties in 321 the analysis (e.g., back trajectory calculations, UHSAS data, missing CO and BC data), 322 the conclusion should be softened to "sea spray aerosol is a POSSIBLE explanation for 323 these observations". It would also benefit the community if a strong recommendation for 324 aerosol chemical composition measurements on future flights over the Southern Ocean 325

were included in the conclusion section. Additional issues to be addressed are listed below.

Author Response: Thank you for taking the time to provide a thoughtful review of our paper. We will address the comments above point by point (our underlined sections of the reviewer comment). There is a version of the revised manuscript with changes tracked since the original submission accompanying this response.

332 2.1.1 Direct evidence for the conclusion is lacking

Direct evidence for our conclusionthat sea-spray aerosol is the most likely (or at least a possible) explanation for the elevated NC/NU observations—can ultimately only be provided by compositional analysis of the observed aerosol. For example, Blot et al. (2013) identified concentrations of "non-volatile" aerosol (predominantly sea salt) by heating the inlet of a Condensation Nuclei (CN) counter to 360°C. No such observations were made during the HIPPO campaigns because the mission priorities were to sample trace gases.

We have been up front about the limitations of the available data for our analysis. Specifically on P. 25512, lines 1-5, we stated that there was no compositional analysis of the aerosol data. However, we agree that it is worth stressing the caveat that our argument is primarily founded on indirect evidence based on the elimination of alternative hypotheses. We have made the following changes to the manuscript:

 Changes to Section 1 (last paragraph) to highlight that we are eliminating alternative hypotheses:

This paper focuses on ... (*snip*) ... with the approach of a strong cold front. Our-Direct evidence for this hypothesis—in the form of observations of aerosol composition—is not available, so our objectives are firstly to verify and analyze the available in-flight microphysics observations, which were not intensive due to their secondary importance for the HIPPO missions, and secondly to ... (*snip*)

Changes to Section 6.1 (last paragraph) to concede that other aerosol sources
 cannot be completely ruled out:

355	While $N_{\rm C}$ values of (<i>snip</i>) observed by the UHSAS, which
356	probably includes most of the CCN, was produced locally. We While
357	alternative sources for the CCN cannot be completely ruled out without
358	compositional analysis of the aerosol, we showed through (snip)

359 2.1.2 Missing CO and BC data

Firstly, there are no BC data for elevations below 1200 m because the SP2 data are not reliable in cloud or rain, so they were removed by the PI based on the CDP and other diagnostics. The same is true for the BC data in profile 3 between 2500 and 4200 m a.s.l, but it is not clear why there is missing data for profile 4. This has been clarified by changing the wording of paragraph 3 of section 4.2 (see below).

Within the boundary layer we rely on CO as a marker for combustion, while acknowledging the possibility of marine sources (BC could have ruled these out if it were available). We found that the concentration increased towards the surface. Assuming that the CO were anthropogenic, we would expect to see a similar increase in NU as was observed at higher altitudes. However, the increase is much larger, which supports our argument that SSA is the dominant contributor to $N_{\rm U}$.

However, we refute the suggestion (if it was intended) that missing CO data detracts from our conclusions because the CO data is in fact very complete, with only a small amount of missing data for profile 1 that does not affect the features of that profile.

- ³⁷⁴ Changes to manuscript to address these concerns:
- 1. Change wording of paragraph 3 of section 4.2:

If there was terrestrial interaction for the air sample in profile 3, the signals in the observations were weak. Due to the presence of cloud and precipitation. BC observations were unfortunately missing unavailable at 3000-4000 m a.s.l., but slightly elevated CO concentrations at about 4000 m a.s.l. correspond to increased $N_{\rm U}$ of about 50 cm⁻³ from values of 10-20 cm⁻³ just above the boundary layer.

Section 4.2, final paragraph modified to clarify these points (and strengthen our argument). Some minor errors in the quoted CO concentration were also fixed in this paragraph.

At 4000 m a.s.l. in profile 4, there was a slight ... (snip) ... to a maximum 385 of 57 ppbv at 167 m a.s.l. This negative CO gradient could be argued 386 to correspond to the group of trajectories that passed near land, but 387 a marine source below a poorly-mixed boundary layer could also account 388 for this. In any Without BC observations at these levels, it is difficult 389 to attribute the source of this CO to human activity (combustion) or 390 natural marine sources. In either case, $N_{\rm U}$ varied much more in the boundary layer than it did near 4000 m a.s.l. for a similar variation in 392 CO, so it is difficult to attribute the elevated $N_{\rm U}$ to long-range transport. 393

³⁹⁴ 2.1.3 Application of lognormal fits to UHSAS data

Thank you for this excellent suggestion. The analysis of the PSDs in the original manuscript was somewhat perfunctory and by applying the methods of Modini et al. (2015), we have been able to provide additional evidence to support our hypothesis that SSA are especially important for the aerosol population in profile 4, and we feel that the revised manuscript is stronger as a result.

As we argue in the next point, it is not necessary to modify the size distribution to
 account for the relative humidity of the sampled air because of heating of the UHSAS
 inlet and internal components.

We used the method described by Modini et al. (2015) to fit a PMA mode (we used 403 the terminology SSA mode to be consistent with the rest of our manuscript while 404 acknowledging there are other contributors), with one minor modification. Modini et al. 405 (2015) fit the PMA mode to particles with $D > 0.5 \,\mu m$. However, with the logarithmically 406 spaced bins of the UHSAS (0.06 to $1 \,\mu$ m) and the poor sensitivity for the largest bins, we 407 found the fits obtained could be relatively poor. We relaxed this parameter and sought 408 the best overall fit using a variable cut-off for 0.2;D; $0.5 \mu m$. The actual values used were 409 $0.391, 0.359, \text{ and } 0.235\,\mu\text{m}$ for profiles 1, 2, and 4 respectively, but the sensitivity of the 410 calculated SSA component to these cut-off values was low. 411

⁴¹² The changes to the manuscript as a result of the new analysis are as follows:

413 414	1.	New section (section 2.4 in revised manuscript) summarizing the methods used Please see accompanying tracked changes document.
415 416 417	2.	Figure 6 of revised manuscript now contains four panels: One for the combined $CDP/2DC$ PSDs (as previously), and one panel for the UHSAS PSD for each profile.
418 419 420	3.	We changed the averaging interval for the lowest leg to 60 s to reduce noise. This had the effect of slightly changing some of the values quoted in section 3.2 for $N_{\rm U}$ and so on.
421 422 423	4.	In addition, we aggregated the UHSAS bins into groups of 3 so that they remained logarithmically spaced. This did not affect the results of the analysis, but it makes the graphs somewhat easier to interpret.
424 425	5.	Changes to discussion in section 3.2 to describe the new results. Please see accompanying tracked changes document.
426	6.	Sentence added to section 6.1, last paragraph:
427 428 429 430 431 432		While $N_{\rm C}$ values of $(snip)$ was produced locally. We While alternative sources for the CCN cannot be completely ruled out without compositional analysis of the aerosol, we showed through modal decomposition of the UHSAS PSD for profile 4 that the mode representing primary marine aerosol, which is dominated by SSA, to be the main contributor to the aerosol population. We thus consider sea
433		spray aerosol (snip)

434 2.1.4 Shift of UHSAS data to dry diameters

Thank you for this suggestion. We understand the importance of relative humidity 435 in determining the diameter of hygroscopic particles, especially sea salt, in ambient 436 conditions. At very high humidity, the ambient particle diameters can be 2-4 times the 437 "dry" diameters. However, we need to stress that in spite of the high relative humidity 438 of the environment (especially for profile 4 where it was close to 100%), the UHSAS 439 measurements are not of the ambient particle diameters. Firstly, the sampled air has 440 been decelerated prior to entering the inlet and heated adiabatically (ram rise) by about 441 4–9°C. This depends on the speed of the aircraft and the recovery factor of the inlet, 442 which is an unknown quantity. Secondly, the sensor inlet is electrically heated to prevent 443 icing, and finally, the optics block is maintained at 30°C. If ambient air at around 5°C 444 and 100% RH were heated by only 10° C (a conservative estimate given the heat sources 445 mentioned), the resultant RH would be about 50%. This is why we, like other authors, 446 do not believe that a correction to the particle size distribution would be necessary 447 for the UHSAS data. However, in the revised manuscript, we have made the following 448 changes to clarify this point: 449

⁴⁵⁰ 1. Added sentence to UHSAS item in section 2.2:

451	• DMT Ultra High Sensitivity Aerosol Spectrometer (UHSAS). The
452	UHSAS measures sizes of aerosol (snip). Due to the
453	combined effect of electrical anti-ice and internal heating, and
454	adiabatic heating of decelerated inlet air, we assumed that the
455	measured particle diameters were close to their dry diameters
456	(e.g. Blot et al., 2013; Kassianov et al., 2015) $(snip)$

457 2. Bibliography item added for Kassianov et al. (2015).

458 2.1.5 Softening of conclusions

⁴⁵⁹ Thank you for this comment. Even though there are some uncertainties in our analysis, ⁴⁶⁰ in part due to missing data and in part to the absence of instrumentation that would ⁴⁶¹ make the argument unequivocal, we believe that we have presented a strong case for ⁴⁶² our the hypothesis that the elevated aerosol and droplet concentrations are due to sea ⁴⁶³ spray aerosol. However, comments from both Reviewer #1 and Reviewer #2 requested ⁴⁶⁴ a dilution of the language in the conclusion, so we have made the following changes:

465 1. Section 7.1 (only paragraph):

In this paper, we have presented ... (snip) ... much higher altitudes in
the profiles. We conclude that these observations are consistent with the
local production of sea spray aerosol through the due to high winds in
the southernmost regions of the flightis the most likely explanation for
these observations... (snip)

471 2.1.6 Strong recommendation for aerosol chemical composition measure 472 ments on future flights over the Southern Ocean

⁴⁷³ Thanks for this comment. We couldn't agree more. We have added a sentence to the ⁴⁷⁴ final paragraph of the new manuscript:

475 1. Section 7.1 (only paragraph):

In this paper, we have presented ... (snip) ... these observations. In order
 to reduce ambiguities such as those discussed in this paper, we strongly
 recommend the inclusion of aerosol chemical composition measurements
 for future cloud physics observational missions over the Southern Ocean.

480 2.2 Specific comments

P. 25511, Section 2.3. These measurements quantify refractory BC and CO. It is mentioned that they do not measure mineral dust. Long range transport of anthropogenic sulfate and organic carbon would also be missed.

Author Response: The purpose of including the BC and CO measurements was primarily 484 to identify anthropogenic signatures (specifically combustion) in the data, and to relate 485 these to the aerosol concentrations. Sulfur dioxide measurements were not made in real 486 time, so we were unable to test for the likelihood of significant quantities of sulfate 487 aerosol. However, we would anticipate that both sulfur dioxide and sulfate aerosol, from 488 either anthropogenic or natural sources, would only be found in the presence of CO. In 489 addition, the CO data was one of the most gap-free and reliable datasets from the flight 490 and as such it was the best choice. 491

The BC data, as you point out, has some issues with missing values in and below cloud, but it was the only aerosol composition data available, and is also an excellent tracer for anthropogenic activity.

⁴⁹⁵ We do not believe that any specific changes to the manuscript are necessary to address ⁴⁹⁶ this comment.

⁴⁹⁷ P. 25525, Lines 11–14: It is stated in the text that during profile 4, $N_{\rm C}$ was in the range ⁴⁹⁸ of 6–10 cm⁻³ with particles of mean diameter 6–7 μ m. Based on Figure 5, below cloud

$_{499}$ N_C for profile 4 was around 450 cm⁻³ with a mean diameter of 40 μ m. Am I reading the Figure incorrectly?

Author Response: Thank you for this comment. It appears some clarification is needed. Firstly, figures 4, 5, 9, and 10 have a profile-dependent offset (specified in the legend of each panel) so that the lines do not overlie one another. For the panel you refer to (Fig. 5 panel 2), there is an offset of 100 cm⁻³ between the successive lines. For profile 4 one must subtract 300 from the value indicated by the line. We understand that this is somewhat confusing but have tried other ways of visualizing these data and haven't come up with a better way to do it.

Secondly, we got the values you quoted from the sub-cloud leg (the same interval as used to calculate the PSD for the UHSAS data in Fig. 6 panel 1), which occurred immediately after the descending profile was completed. It is not really possible to read the values from the line in Fig. 5 panel 2, but you should be able to see that the bottom of the line (where it is dashed because $\rho_{\rm L}$ was below 0.05) goes to 300 (i.e. zero).

To reduce the confusion around this point, we have repeated the note about the offsets for the different profiles from Fig. 4 in the caption of Fig. 5, and we have removed the offsets altogether from Figures 9 and 10.

P. 25527, lines 18–25: It is stated that "For profiles 3 and 4... the most likely signature of anthropogenic influence was well above and decoupled from the boundary layer..." But there are no BC data for these flights below 1200 m (at least according to Figure 9). And CO is not significantly lower (65 and 70 ppbv for flights profiles 3 and 4 at altitudes <1000 m) than the air masses tagged as anthropogenic during Flight 2 (~ 65 ppbv at 4000–5000 m).

Author Response: Thank you for this comment. Please note that we have assumed you meant to write "profiles" instead of "flights" as indicated by our change above.

Secondly, unfortunately, we believe that you have again misunderstood our offsets between 524 the lines for each profile. Subtracting the offsets specified in the panel legend, the CO 525 values for profiles 3 and 4 below 1000 m were about 55 and 57 ppbv, respectively. You 526 are correct in pointing out that these are not too different from the values above the 527 boundary layer in RF07 ("Flight 2"), which were about 55 ppbv. However, we would 528 argue that the air above the boundary layer in RF07, in spite of having a recent history 529 over the Australian continent, is very clean based on the absence of BC and extremely 530 low NU. We present RF07 as an example of a clearly polluted boundary layer against 531 the background of a pristine airmass. 532

Our main argument here is that there is a weak anthropogenic signal at 3000–400 m a.s.l. 533 in profiles 3 and 4 of RF06, but there is a layer between about 1250 and 2000 m a.s.l 534 which is as clean as any other data that we have seen (especially for profile 4). This 535 brings into question the origin of the slightly elevated CO at the bottom of the profiles, 536 since there are natural maritime sources as well as anthropogenic ones. In any case, even 537 if it were anthropogenic, given the tiny increase in $N_{\rm U}$ at 3000–4000 m a.s.l. for a much 538 larger increase in CO, anthropogenic sources could not account for the observed increase 530 in $N_{\rm U}$ at the lowest levels. 540

We have made some changes to the manuscript to address this comment and clarify our arguments:

543
 1. Changes to section 4.2, last paragraph (some minor errors in the quoted CO concentration were also corrected):

545

At 4000 m a.s.l. in profile 4, there was ... (snip) ... evidenced by

546		$N_{\rm U}$ values strictly below $20 {\rm cm}^{-3}$ and CO concentration of 52 ppby
547		between 1250 and 2000 m a.s.l. Within the boundary layer itself, CO
548		concentrations decrease with height from 54 ppby-increase towards the
549		surface to a maximum of 57 ppbvat 167 m a.s.l. to 50 ppbv at 1250 m a.s.l.
550		This negative CO gradient could be argued to correspond to the
551		group of trajectories that passed near land, but a marine source below
552		a poorly-mixed boundary layer could also account for this. In any
553		Without BC observations at these levels, it is difficult to attribute the
554		source of this CO to human activity (combustion) or natural marine
555		sources. In either case, $N_{\rm U}$ varied much more in the boundary layer than
556		it did near 4000 m a.s.l. for a similar variation in CO, so it is difficult to
557		attribute the elevated $N_{\rm U}$ to long-range transport.
558	2.	Sentence added to section 4.3, paragraph 1:
559		It is useful to provide some context $(snip)$ CO concentrations $(R =$
560		0.87 and $R = 0.95$ respectively). On the other hand, the free troposphere
561		was extremely clean based on the CO values of about 55 ppby, BC of
562		virtually zero, and $N_{\rm U}$ of 20–40 cm ⁻³ .
563	3.	Changes to section 6.1, paragraph 3:
564		For profiles 3 and 4 we have argued that the most likely signature
565		of anthropogenic/continental influence was at 3000-4000 m a.s.l., which
566		was well above and decoupled from the boundary layer , and that by a
567		layer of clean air. Furthermore, $N_{\rm U}$ at those levels (about $80 {\rm cm}^{-3}$) was
568		insignificant in comparison to values within the boundary layer (nearly
569		400 cm^{-3}). The trajectories that arrived (<i>snip</i>) profile 1. The very
570		small increase in CO in the boundary layer may equally suggest a highly
571		diluted anthropogenic signature , but or a natural maritime source, but
572		$\underline{in any case}$ it is not nearly of sufficient magnitude to (<i>snip</i>)

573 **3** Anonymous Referee #3

574 Received and published: 8 November 2015

575 3.1 General comments

I have reviewed the manuscript "Observations of high droplet number concentrations 576 in Southern Ocean boundary layer clouds" by Chubb et al. The work presents results 577 from a small subset of HIPPO flights and examines the microphysical properties of 578 boundary-layer clouds from a small set of observations made near Tasmania. The work 579 highlights that a wide range of cloud droplet number concentrations were observed during 580 these flights. The authors hypothesize that the large number of drops could be related 581 to either anthropogenic emissions or sea-spray aerosol. Based on their analysis of model 582 back trajectories the authors argue that the most likely cause of the large numbers of 583 particles is the generation of sea-spray aerosol associated with very strong winds. It 584 is important to note that this finding is not based on direct observations, but rather 585 on the elimination of a number of other potential sources of the particles. Overall, the 586 manuscript provides a clear and concise description of the results, and I feel that the 587 manuscript would likely be acceptable for publication in Atmospheric Chemistry and 588 Physics with some minor changes. That said, the study would be much more convincing 589

(and useful to the community) if additional data sets were available to give an idea of the importance of the contribution of sea-spray aerosol in a larger sense, if more measurements of aerosol chemistry could be used to help highlight the nature of aerosol that is observed, and application of a more detailed chemistry model to better understand the aerosol sources (including the potential role of mineral dust). I have provided some more detailed comments below.

Author Response: Thank you for taking the time to review our manuscript. The main points raised in your summary above (underlined by us) are that additional data would 597 have had the potential to make our conclusions much stronger, which we of course 598 completely agree with. This point was also raised by Reviewer#2, who additionally 599 requested that we soften our conclusions as a result. Reviewer#1 also requested a 600 softening of our conclusions based on the ambiguities relating to aerosol composition. 601 We have made two several changes to the manuscript as a result of those requests 602 which should address your comment, including a recommendation for aerosol chemical 603 composition measurements on future flights over the Southern Ocean. Please note that 604 there is a version of the revised manuscript with changes tracked since the original 605 submission accompanying this response. 606

Changes to Section 1 (last paragraph) to highlight that we are eliminating alternative hypotheses:

609	This paper focuses on (<i>snip</i>) with the approach of a strong cold front.
610	Our Direct evidence for this hypothesis—in the form of observations of
611	aerosol composition—is not available, so our objectives are firstly to
612	verify and analyze the <u>available</u> in-flight microphysics observations, which
613	were not intensive due to their secondary importance for the HIPPO
614	missions, and secondly to (snip)

- Changes to Section 6.1 (last paragraph) to concede that other aerosol sources
 cannot be completely ruled out:
- ⁶²¹ 3. Changes to section 7 to soften final conclusion:

In this paper, ... (*snip*) ... We conclude that <u>these observations are</u> <u>consistent with the</u> local production of sea spray aerosol through the <u>due</u> to high winds in the southernmost regions of the flightis the most likely <u>explanation for these observations</u>. ... (*snip*)

4. Changes to section 7 to recommend inclusion of additional instrumentation on future flights:

628	In this paper, (<i>snip</i>) most likely explanation for these observations.
629	In order to reduce ambiguities such as those discussed in this paper,
630	we strongly recommend the inclusion of aerosol chemical composition
631	measurements for future cloud physics observational missions over the
632	Southern Ocean.

Page 25505, line 7. I would suggest adding the word "observed" after "droplet sizes."

⁶³⁴ Author Response: Accepted; please see tracked changes document.

Page 25505, line 13 (and other locations). The use of high and low to mean large and small could be confusing to the reader. I would suggest changing the occurrences with large and small to be more accurate.

Author Response: Thanks for this comment. We would prefer to retain our terminology in this case because $N_{\rm C}$ and $N_{\rm U}$ have units of concentration (cm⁻³). For this example, if we substituted words for our symbols and used "small" instead of "low", it would read as follows: "Droplet number concentration was found to be small," which sounds odd.

⁶⁴² Page 25505, line 29. "Provide" should be "provided".

643 Author Response: Accepted; please see tracked changes document.

Page 25508, line 5. I would suggest adding "associated" after "low-level winds".

645 Author Response: Accepted; please see tracked changes document.

Page 25508, line 16. It would be helpful, at some point in the document, to indicate the
airspeed of the aircraft. That would make it easier to understand the impacts of the
sampling speed on the results.

Author Response: Accepted; this was addressed in response to a comment by Reviewer#1.
 The changes to the manuscript were:

Inserted sentences in section 2.1 (final paragraph) to describe air speeds and vertical
 motion:

The aircraft performed four ... (snip) ... provided in Fig. 3. The mean true air speed varied during the profiles, but it was consistently about 130 m s⁻¹ at altitudes below 1500 m a.s.l. (well above the boundary layer). The vertical speed of the HIAPER was about 7.5 m s^{-1} for altitudes above 600 m a.s.l., and 2.5 m s^{-1} below this. A total distance of about 38 km was covered between the lowest level and 1500 m a.s.l. for each profile. Conditions were quite ... (snip)

Page 25509, line 18. Additional detail about the ascent/decent profiles would be helpful. For example, what was the approximate ascent/decent rate of the aircraft? Were these profiles designed to overlap each other?

Author Response: The design of the profiles was essentially to sample the atmosphere from about 8000m down to near the surface during a series of flights that constituted a global transect (see Wofsy 2011). The aircraft was either climbing or descending almost continuously so there was no overlap in the profiles. Details of the descent rate were included in response to comments by Reviewer#1, and are included in the change for the previous item (see above).

Page 25513, line 20-25. The text argues that the potential temperature profile shown for profile 2 is more complex than that for profile 1, but that isnt clear to me from the figure unless the authors are referring to the buffer layer. Perhaps the small spatial extent of the figure hides the relative complexity? The text also states that large values of CDP liquid water content are consistent with a cumulus cell. Given the aircraft speed, how large would this cell have to be to provide the continuous profile shown in the figure? Is that reasonable?

676 Author Response:

Thank you for this comment. The buffer layer is precisely the complexity to which we refer. We agree that there is a lot of information in figure 4, but the main point is that the change in specific humidity (and also NU, but this discussion follows later) occurs at



Time series data for profile 2. Blue line: altitude (left axis). Green lines: 1Hz and 11-point smoothed LWC.

a different level to the v increase (temperature inversion). It seems that we neglected to
 include this point in that paragraph, so we have added it in the revised manuscript (see
 below).

The second point was about the size of the putative cumulus cell. We mentioned that 683 there was a relatively consistent $\rho_{\rm L}$ in a layer about 250 m, which is about right based 684 on our Figure 4. When viewed as a time series and smoothed, as in Figure 2 of this 685 document, there is a local maximum near t = 18865. The slope of this feature drops of 686 sharply after about t = 18870, even though the descent rate decreased dramatically at 687 about this time, suggesting that the aircraft is indeed exiting a convective feature through 688 the side (nearly horizontally) rather than through the base. A reasonable estimate for the 689 traversal time of the convective core might be 10 s (1300 m), which is not unreasonable 690 for this type of feature. 691

The 1 Hz data appears to have some periodicity in this region with 6 peaks spaced by roughly 5 s (650 m), but they are not clearly separated by clear air. There may well be some complexity to the feature (entrainment and/or multiple cores) that we do not attempt to explain.

⁶⁹⁶ 1. Changes to section 3.1 (paragraph 2). Description of change in humidity with
 ⁶⁹⁷ altitude.

698	Profile 2 shares a number of (snip) intermediate layer of about
699	200 m, with in which the specific humidity (q_v) remained similar to the
700	in-cloud values. There was a weaker θ_v increase at 1320 m a.s.l., which
701	was coincident with a sharp drop in q_v to nearly zero. At cloud top
702	(snip)

Changed section 3.1 (paragraph 2) to improve our description of the convective feature:

705Profile 2 shares a number of ... (snip) ... cloudy layer. Below706thisthere was a layer about 250 m deep with relatively consistent values707of $\rho_{\rm L} \simeq 0.25 \,{\rm gm}^{-3}$, which we interpret, the 10-second smoothed $\rho_{\rm L}$ 708(not shown) reached a minimum of $0.25 \,{\rm gm}^{-3}$ before increasing briefly709to $0.35 \,{\rm gm}^{-3}$ and then dropping rapidly as the aircraft continued to710descend. We interpret this feature as a cumulus cell rising into ... (snip)

Page 25516, line 7. It would be good to add a note to the caption of Figure 5 about the
 offset applied to some profiles.

Author Response: Accepted; this was done in the response to a comment by Reviewer #2.
We have also removed the offsets from Figures 9 and 10 to improve clarity.

Page 25517, line 3. The text states that there is a peak in $N_{\rm U}$ near cloud top, but that isn't clear to me from the figures.

Author Response: Apologies; the dashed line (representing in-cloud values, which are absolutely not to be trusted due to droplets splashing on the inlet) disappeared from this panel somehow. This has been rectified in figure 5 of the new manuscript.

Page 25517, line 27. You might want to add "thermodynamically" before the word
 "stable".

722 Author Response: Accepted; please see tracked changes document.

Page 25522, line 11. The text in this paragraph states that HIPPO-4 RF06 is not a
good example of a pristine flight nor a polluted one, but early in the section, Profile 1 is
descried as "very clean". This description appears to be a bit inconsistent.

Author Response: Thank you for this comment. It was somewhat paradoxical that the profile closest to the continent was in many ways the cleanest. Upon reflection, we find the sentence describing profile 1 as "very clean" is somewhat redundant so we have removed it from the revised manuscript. Please see the tracked changes document.

Page 25523, line 9. The text states that the trajectories from 500 and 1500 m are very
similar, and if dust is a major issue than the aerosol loading should be the same (or
at least close in value). Is this due to deeper boundary layers (and associated vertical
mixing) over Australia?

Author Response: We showed that trajectories arriving at 500 and 1500 m for profile 734 3 actually had similar histories both spatially and vertically (at least for those that 735 originated over land). Many of the members for the 1500 m ensemble were close to the 736 surface when over land. Therefore, as we mentioned, the aerosol loading should be similar 737 for these levels at the location of profile 3, but it was not (suggesting, again, a local 738 aerosol source). Presumably the mixed layer would have been deeper over the continent 739 than over the ocean, but this is speculative and not necessary for our argument. We 740 have not made any specific changes to the manuscript in response to this comment. 741

742 **References**

Baumgardner, D. and Korolev, A.: Airspeed corrections for optical array probe sample
 volumes, Journal of Atmospheric and Oceanic Technology, 14, 1224–1229, 1997.

Blot, R., Clarke, A. D., Freitag, S., Kapustin, V., Howell, S. G., Jensen, J. B., Shank, L. M.,
 McNaughton, C. S., and Brekhovskikh, V.: Ultrafine sea spray aerosol over the
 southeastern Pacific: open-ocean contributions to marine boundary layer CCN, Atmos.

- ⁷⁴⁸ Chem. Phys., 13, 7263–7278, 2013.
- ⁷⁴⁹ Boers, R., Jensen, J., Krummel, P., and Gerber, H.: Microphysical and short-wave
 ⁷⁵⁰ radiative structure of wintertime stratocumulus clouds over the Southern Ocean, Q. J.
 ⁷⁵¹ Roy. Meteor. Soc., 122, 1307–1339, 1996.
- Hande, L., Siems, S., and Manton, M.: Observed trends in wind speed over the Southern
 Ocean, Geophys. Res. Lett., 39, L11802, 2012.

Kassianov, E., Berg, L. K., Pekour, M., Barnard, J., Chand, D., Flynn, C., Ovchinnikov,
 M., Sedlacek, A., Schmid, B., Shilling, J., et al.: Airborne aerosol in situ measurements
 during TCAP: A closure study of total scattering, Atmosphere, 6, 1069–1101, 2015.

King, W., Parkin, D., and Handsworth, R.: A hot-wire liquid water device having fully
calculable response characteristics, Journal of Applied Meteorology, 17, 1809–1813,
1978.

Korhonen, H., Carslaw, K. S., Forster, P. M., Mikkonen, S., Gordon, N. D., and
 Kokkola, H.: Aerosol climate feedback due to decadal increases in Southern Hemisphere
 wind speeds, Geophys. Res. Lett., 37, L02805, 2010.

Lance, S., Brock, C. A., Rogers, D., and Gordon, J. A.: Water droplet calibration of the
 Cloud Droplet Probe (CDP) and in-flight performance in liquid, ice and mixed-phase
 clouds during ARCPAC, Atmos. Meas. Tech., 3, 1683–1706, 2010.

Modini, R., Frossard, A., Ahlm, L., Russell, L., Corrigan, C., Roberts, G., Hawkins, L.,
 Schroder, J., Bertram, A., Zhao, R., et al.: Primary marine aerosol-cloud interactions
 off the coast of California, Journal of Geophysical Research: Atmospheres, 2015.

Romashkin, P.: NCAR GV (HIAPER) Low Rate (LRT - 1 sps) Navigation, State
 Parameter, and Microphysics Flight-Level Data, Tech. rep., UCAR/NCAR - Earth

Observing Laboratory, Boulder, CO 80307, USA, available at: http://www.eol.ucar.

edu/system/files/PM_notes_HIPPO-4_-5.doc, (last access: 31 August 2015), 2012.

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Observations of high droplet number concentrations in Southern Ocean boundary layer clouds

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Discussion Paper

Discussion Paper

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Data from the standard cloud physics payload Cloud physics data collected during the NSF/NCAR High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO) campaigns provide a snapshot of unusual wintertime microphysical conditions in the boundary layer over the Southern Ocean. On 29 June 2011, the HIAPER sampled the boundary layer in a region of pre-frontal warm air advection between 58 and 48° S to the south of Tasmania. Cloud droplet number concentrations were consistent with climatological values in the northernmost profiles but were exceptionally high for wintertime in the Southern Ocean at 100–200 cm⁻³ in the southern profiles were up to 400 cm^{-3} .

Analysis of back trajectories and atmospheric chemistry observations revealed that while conditions in the troposphere were more typical of a clean remote ocean airmass, there was some evidence of continental or anthropogenic influence. However, the hypothesis of long range transport of continental aerosol fails to explain the magnitude of the aerosol and cloud droplet concentration in the boundary layer. Instead, the gale force surface winds in this case (wind speed at 167 m above sea level was $> 25 \text{ m s}^{-1}$) were most likely responsible for production of sea spray aerosol which influenced the microphysical properties of the boundary layer clouds. The smaller size and higher number concentration of cloud droplets

is inferred to increase the albedo of these clouds, and these conditions occur regularly, and are expected to increase in frequency, over windy parts of the Southern Ocean.

1 Introduction

The remote Southern Ocean (SO; poleward of 45° S) has received recent attention due to substantial biases in both reanalysis and climate simulations associated with clouds (Meehl et al., 2007; Trenberth and Fasullo, 2010). In this pristine environment, cloud properties may be sensitive to relatively small changes in aerosol concentrations, whether from an-

thropogenic or natural sources, but there has been a distinct lack of in-situ microphysical observational campaigns in this region in recent years. High latitude ocean-atmosphere interactions and processes have been identified as a key research frontier by the NSF Advisory Committee for Geosciences (2014).

- ⁵ The pristine environment of the SO raised questions about cloud droplet number concentration (N_c) and droplet sizes observed there in the early 1990s. Boers et al. (1996); Boers and Krummel (1998), and Boers et al. (1998) considered N_c within, and cloud condensation nuclei (CCN) concentrations below, SO boundary layer clouds in "baseline" conditions (with an airmass history far from continental Australia) during the First Aerosol Characterization
- ¹⁰ Experiment (ACE-I, November to December 1995) and the two phases of the Southern Ocean Cloud Experiment (SOCEX-I, July 1993; SOCEX-II, February 1995). In wintertime, $N_{\rm C}$ was found to be low, at typically 10–40 cm⁻³ for clouds of up to 300 m deep, compared to summertime values of 50–180 cm⁻³. A caveat with the lowest wintertime $N_{\rm C}$ values of that study is that they were highly correlated with the cloud liquid water content, suggesting that clear air may have been mixed into those samples.
- Seasonal differences in N_C during the SOCEX experiments were attributed to oxidation products of oceanic dimethylsulphide (DMS) acting as CCN, due to seasonal variation in the productivity of the ocean (Boers et al., 1998). At the time it was widely hypothesized that DMS-derived particulates made up the bulk of all sub-micrometer particles (Charlson et al., 1987), which linked ocean productivity to cloud albedo and thus global climate through the so-called "CLAW" hypothesis. However, a review of two decades of subsequent research suggested that the evidence for each of the stages in this mechanism was rather weak (Quinn and Bates, 2011), and that sea spray aerosol (SSA) comprises a substantial fraction of the marine boundary layer CCN concentration.
- ²⁵ The HIAPER Pole-to-Pole Observations flights (HIPPO; Wofsy, 2011) were not dedicated cloud physics experiments but have nevertheless provide provided some new data at high latitudes over the SO. Chubb et al. (2013) examined two SO flights that encountered low-altitude cloud (i.e. below 2 km) across a broad latitude rangeas far south as 67°S. Direct *N*_C observations were only available on one of those flights, which took place in the month of

April, and ranged from $30-50 \text{ cm}^{-3}$ in weakly convective stratocumulus cloud in the cold air sector of an extratropical cyclone at latitudes around 59° S, to $80-120 \text{ cm}^{-3}$ in a region of homogeneous stratiform cloud in moderate south-westerly flow between $62-67^{\circ}$ S. Broadly speaking, these values were in line with those from ACE-I and the SOCEX experiments.

- ⁵ Cloud particle effective radius r_e and optical thickness τ are standard retrievals (e.g. Nakajima and King, 1990) that may be performed with radiance data from the Moderate Resolution Imaging Spectroradiometer (MODIS; Salomonson et al., 1989). Bennartz (2007) used two and a half years of Aqua MODIS retrievals to calculate N_c over remote oceanic regions. N_c values for the Southern Hemisphere oceans (equatorward of 60° S) were 40– 67 cm⁻³, compared to 64–89 cm⁻³ for the Northern Hemisphere. The estimated frequency
- ¹⁰ 67 cm^{-3} , compared to $64-89 \text{ cm}^{-3}$ for the Northern Hemisphere. The estimated frequency of drizzle (based on empirical relationships with N_{C} and cloud geometric thickness) was substantially higher in the Southern Hemisphere oceans. A limitation in the use of these retrievals at high latitudes, which was not considered by Bennartz (2007), is the solar angle, which must be greater than about 65° to be reliable (Grosvenor and Wood, 2014). In ¹⁵ wintertime, it is virtually impossible to perform robust r_{e} retrievals over the SO.

The primary mechanism of SSA production is the bursting of small bubbles at the sea surface within breaking wave crests, or whitecaps (Day, 1964). The "film drop" particles produced are typically in the radius range of 0.01–1µm, remain suspended for long periods, and form the dominant contribution to marine SSA number concentration (Lewis and Schwartz, 2004, ch. 4). Larger particles can be formed in lower concentrations by "jet" and "spume" mechanisms, but these tend to fall to the sea surface on time scales of seconds to hours and may not contribute substantially to CCN number concentrations.

In spite of the intuitive link between wind speed and SSA concentration, which has long been recognized (Woodcock, 1953), the case for a formulation based on wind speed alone is mixed. SSA production flux per whitecap area is typically assumed to be independent of wind speed, permitting estimates based on fractional whitecap area (W). However, Wcan vary by an order of magnitude for the same wind speed (Lewis and Schwartz, 2004, ch. 3), and the underlying uncertainties in the production fluxes are large (de Leeuw et al., 2011). In spite of this, a relationship between the logarithm of SSA concentration (N_{SSA}) and the local wind speed is typically assumed. Several studies (e.g. Marks, 1990; O'Dowd and Smith, 1993; Nilsson et al., 2001) report very good agreement with this formulation, but during ACE-I, other investigators have reported little or no correlation of N_{SSA} with wind speed (Bates et al., 1998; Covert et al., 1998; Berg et al., 1998). More recently, (Blot et al., 2013) presented observations of N_{SSA} made in the unpolluted south-eastern Pacific during the VOCALS (Variability of the American Monsoon Systems (VAMOS) Ocean-Cloud-

during the VOCALS (Variability of the American Monsoon Systems (VAMOS) Ocean-Cloud-Atmosphere-Land Study) campaign. These data, recorded over <u>a range of 1000 km</u> along the 20° S meridian, showed only a weak relationship to wind speeds up to about 12 m s^{-1} . The authors concluded that other factors, especially local precipitation history, may play an important role in determining SSA concentrations.

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High aerosol concentrations over the SO—an important topic for this paper—could also be caused by long range transport of aerosol from the Australian continent. Using back trajectory analysis coupled with radon and condensation nuclei (CN) concentration observations, Downey et al. (1990) found that long range transport could explain up to 25 % of the variance of radon concentration, which is a good proxy for "land contact", at Mac-

of the variance of radon concentration, which is a good proxy for "land contact", at Macquarie Island (54.62° S, 158.85° E). CN concentrations reached values above 1000 cm⁻³ for short intervals while trajectories were of continental origin, but the trajectory statistics used ("hours of land contact" and "time since land contact") showed very poor, and even negative, correlation to CN concentrations. This was attributed to non-uniformity of CN sources
 on the continent and processes acting as sinks and sources over the ocean (neither of which affect radon concentration).

This paper focuses on some observations from a single flight over the SO in winter during the 4th HIPPO campaign (HIPPO-4), which we selected because cloud. Cloud droplet and aerosol concentrations were considerably higher than expected in a region that was more than 1500 km from the nearest potential pollution sources. The main hypothesis addressed by this paper is that these observations can be attributed to high SSA production due to very strong low-level winds associated with the approach of a strong cold front. Our Direct evidence for this hypothesis—in the form of observations of aerosol composition—is not available, so our objectives are firstly to verify and analyze the available in-flight mi-

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crophysics observations, which were not intensive due to their secondary importance for the HIPPO missions, and secondly to investigate the alternative hypothesis that long-range transport of continental/anthropogenic aerosols influenced microphysical conditions.

2 Methodology and data

⁵ With the primary objective of conducting a global survey of climatically important aerosols and trace gases, the NSF/NCAR HIAPER (a high-performance research aircraft based on a Gulfstream-V jet), conducted five global transects in different seasons between 2009 and 2011 for the HIPPO campaigns (Wofsy, 2011).

The primary dataset used to perform the analyses in this paper was the "Low Rate (1 Hz)
Navigation, State Parameter and Microphysics Flight-Level Data" product (Romashkin, 2012) prepared by the NCAR Research Aviation Facility (RAF). In addition, we used one-second data from additional instrumentation, which was processed by various HIPPO investigators separately from the flight-level data (see below). These data formed the basis for the median-filtered "Merged 10 s Meteorology, Atmospheric Chemistry, and Aerosol Data"
product (Wofsy et al., 2012), which has been used in many of the publications resulting from the HIPPO campaigns.

2.1 Flight overview

During daylight hours of 28–29 June 2011 (solar time), the HIAPER flew from Christchurch (New Zealand), to Hobart (Australia) via a way-point at 58° S, due south of Hobart. The
Mean Sea Level Pressure (MSLP) analysis for 00:00 UTC 29 June 2011 (Fig. 1) shows a remarkably strong blocking anticyclone with a high-pressure center of 1042 hPa over the Tasman Sea. A mature, decaying frontal system was approaching from the west, with a secondary wave anomaly located at about 48° S, 130° E. This synoptic pattern generated a strong south-westerly pressure gradient in the pre-frontal airmass, with ERA-Interim wind speeds at 950 hPa in excess of 20 m s⁻¹ associated with strong poleward warm air advection.

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The Aqua satellite passed overhead at 03:45 UTC, while the HIAPER was mid-flight. MODIS retrievals show widespread boundary layer clouds with cloud top temperature (CTT) of 270 to 280 K underneath the blocking high (Fig. 2). A complex of multilayer cloudclouds, with CTT in the range of 220–240 K, resided in the pre-frontal stream overlying the boundary layer cloud. In the vicinity of the secondary wave anomaly, the high level cloud band was broken, permitting retrievals of the boundary layer cloud beneath, which appeared to be consistent with the cloud well ahead of the frontal band.

The aircraft performed four descent/ascent profiles between 9000 and 160 m above sea level (a.s.l.) while in transit from the southernmost point to Hobart, which we discuss in reverse order (north to south) below. The locations of the short leg-low-level legs (at about 160 m a.s.l.) between the descent and ascent profiles are shown in Fig. 2, and imagery from the forward facing camera showing cloud top conditions are provided in Fig. 3. The mean true air speed varied during the profiles, but it was consistently about 130 m s⁻¹ at altitudes below 1500 m a.s.l. (well above the boundary layer). The vertical speed of the HIAPER was

about 7.5 m s⁻¹ for altitudes above 600 m a.s.l., and 2.5 m s⁻¹ below this. A total distance of about 38 km was covered between the lowest level and 1500 m a.s.l. for each profile. Conditions were quite varied between the profiles, with profiles 3 and 4 occurring close to the location of the synoptic front, and profiles 1 and 2 in pre-frontal conditions. In all but profile 1, there was some cirrus cloud well above the maximum altitude reached in the profiles.

2.2 HIPPO basic cloud physics instrumentation

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Basic cloud microphysics instruments were operated in addition to the primary payload instrumentation, including:

- Particle Measurement Systems (PMS) 2-D Cloud Imaging Probe (2DC). Precipitation particles larger than about 50 µm can be imaged by optical array probes such as the 2DC. The instrument returns particle statistics in the form of a size distribution histogram with 64 bins between 12.5 and 1600 µm as well as individual particle images. Here, as for most applications of the 2DC, we only use This type of probe is susceptible to uncertainties in depth-of-field for particles with diameters less than 200 μ m (Baumgardner and Korolev, 1997), but we made no specific correction for this other than only using particles with diameters larger than 62.5 μ m to determine drizzle drop number concentrations and rain rates (no ice was observed in the boundary layer clouds).

- Droplet Measurement Technologies (DMT) Cloud Droplet Probe (CDP). The CDP operates by illuminating individual droplets with a laser beam and measuring the intensity of the forward-scattered light over angles between 4 and 12° (Lance et al., 2010), and sizes them with a multi-channel analyzer. The instrument returns a particle size distribution over 30 bins between 2.0 and 50 µm at 1 Hz. Particle Droplet number concentration (N_C) and liquid water content (ρ_L ; g m⁻³) are subsequently derived from the size distribution. The CDP sizing was calibrated using glass beads of known sizes in Boulder, CO, prior to the commencement of HIPPO-4. Subsequently to the HIPPO missions, the CDP had its true sample area evaluated through a laboratory beam mapping by the manufacturer in June 2015.

We recalculated $\rho_{\rm L}$ and $N_{\rm C}$ using the updated sample area.

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- DMT Ultra High Sensitivity Aerosol Spectrometer (UHSAS). The UHSAS measures sizes of aerosol particles between 60 and 1000 nm based on light scattering (Cai et al., 2008). Due to the combined effect of electrical anti-ice and internal heating, and adiabatic heating of decelerated inlet air, we assumed that the measured particle diameters were close to their dry diameters (e.g. Blot et al., 2013; Kassianov et al., 2015). We designate the total particle concentration measured by the UHSAS as N_U in this paper. The instrument was calibrated using polystyrene latex beads of known sizes prior to HIPPO-4. We designate the total particle concentration measured by the UHSAS as N_U in this paper.
- PMS-King "hot-wire" probe. Total cloud liquid water content can be directly measured by exposing a temperature-controlled element to the flow outside the

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aircraft (King et al., 1978) . Within cloud, the power required to maintain a constant temperature is compared to that required in clear air to derive $\rho_{L,King}$.

Additionally, there are a number of basic thermodynamic and inertial observations used in this paper, and real-time forward digital camera imagery was available for all flights.

⁵ We partitioned the data by liquid water content, using $\rho_L < 0.01 \text{ gm}^{-3}$ for "probably clear" samples, needed for ensuring that the UHSAS observations were robust (see section 5.2); and $\rho_L > 0.05 \text{ gm}^{-3}$ for "confident cloudy" samples, for calculating N_C from the CDP data. Similar thresholds are commonly used to discriminate between clear and cloudy samples (e.g. Wood and Field, 2011; Boutle et al., 2014) when high-rate data is unavailable (as in this case). Our study differs by using two thresholds to more selectively discriminate between

cloudy and clear air.

2.3 HIPPO trace gas and aerosol instrumentation

To address the possibility that the high aerosol concentrations observed in the boundary layer are due to long range transport of pollution from the Australian continent, we used atmospheric chemistry collected during the flight:

- DMT Single Particle Soot Photometer (SP2). The presence of black carbon (BC), or soot, indicates combustion, and is an excellent tracer for anthropogenic aerosol sources. The SP2 measures the incandescence temperature of particles illuminated by a laser beam (Schwarz et al., 2006). BC data acquired in clouds were removed from the HIPPO dataset based on SP2 internal diagnostics, the 2DC and CDP, and the hot-wire liquid water sensor.
- AeroLaser Vacuum Ultra Violet (VUV) resonance fluorescence instrument. Carbon monoxide (CO) is another useful indicator of combustion, but there are also natural marine sources. The VUV operates on the principle of CO fluorescence in the 160–190 nm wavelength range upon excitation with ultra violet light at 150 nm. The technology is relatively mature and has been employed on aircraft platforms for over a decade (Gerbig et al., 1999).

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Unfortunately, there was no compositional analysis of aerosols performed apart from the presence of BC. In principle, this leaves open the possibility of elevated N_U values due to continental (mineral) dust in the absence of CO or BC, as anthropogenic aerosol emissions are almost exclusively produced in conjunction with combustion. We explore this possibility further in Sect. 4.4.

2.4 Modal decomposition of UHSAS particle size distribution

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Even without information about aerosol composition, it is still possible to estimate the contribution of SSA to the observed UHSAS concentration. Modini et al. (2015) fitted lognormal modes to observed aerosol size distributions off the Californian coast. The primary marine aerosol (dominated by SSA) was isolated by fitting a lognormal mode, constrained to having μ (mean diameter) of $0.2\mu m \pm 30\%$ (i.e. $0.14-0.26\mu m$) and σ (geometric standard deviation) of between 2.5 and 3. This mode was fitted to the upper portion of the PSD only ($D > 0.5\mu m$), and a second unconstrained mode, corresponding to a cloud-processed accumulation mode, was fitted to the residual of the PSD following subtraction of the SSA mode. Modini et al. (2015) fitted a third mode with mean diameter of

subtraction of the SSA mode. Modini et al. (2015) fitted a third mode with mean diameter of 25–80 nm, but we have no observations of particles in that size range.

We used this approach on 60 second PSDs for the UHSAS at the lowest level of flight for each profile, with the size bins aggregated into groups of three to reduce noise. The only difference with our analysis was that we optimized the cut-off for the SSA mode-fitting.

We chose a diameter in the range 0.2–0.5µm that minimized the root mean square error of the combined lognormal fits relative to the original PSD. This was necessary because the upper size of the UHSAS was considerably lower than the instruments used by Modini et al. (2015), and the sensitivity of the UHSAS in the larger bins is limited. The sensitivity of the results to the use of a variable threshold was indeed quite low.

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2.5 Calculation of back trajectories

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We used the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT; Draxler and Hess, 1998) to calculate back trajectories via the Air Resources Laboratory (ARL) portal (http://www.arl.noaa.gov/HYSPLIT.php). The meteorological data selected to
⁵ run the calculations was based on output from the U.S. National Weather Service's National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS). The ARL processes and archives this output as a 3 hourly, global, one degree latitude-longitude dataset on mandatory pressure levels (21 levels between 1000 and 1 hPa), and makes it available on. This dataset is sufficient to resolve the synoptic scale features which are dominant, at least in the free troposphere, and is available through the HYSPLIT portal. Back trajectories Draxler and Hess (1998) provide a discussion of factors influencing the accuracy of meteorological trajectories, which is worth summarizing here. The way that vertical motion is treated is especially important because horizontal wind components

generally vary with height, so any vertical displacement errors will contribute to horizontal displacement errors as well. Vertical winds are generally deduced from the divergence of modeled horizontal components and can be noisy. It is possible to calculate isentropic trajectories, which follow surfaces of constant θ , which is a good approximation to the motion of dry air in the free troposphere. However, this assumption is not useful in the boundary layer (which is important for this paper) where θ is well-mixed, so we used modeled vertical velocities in spite of these caveats.

Other factors that could influence the accuracy of our trajectories are that the reanalysis products over the SO are known to be questionable (e.g. Hande et al., 2012b; Huang et al., 2015), and that our region of interest was experiencing a rapid dynamical change with the approaching cold front. In order to test the sensitivity of the trajectory calculations these uncertainties, we used an ensemble approach in which 26 additional trajectories were calculated in addition to the "deterministic" one. These were initialized at horizontal perturbations of Δx and Δy of one grid point (one degree), and $\Delta \sigma$ of 0.01 (about 250 m), the standard configuration recommended by the ARL portal, and translated back to the central point.

Back trajectories ensembles were initialized along the flight path where the HIAPER was within the boundary layer cloud in profiles 1-4, at levels of 500 (within the boundary layer), 1500 (just above the boundary layer top) and at a third height between 3000 5 and 4500 m a.s.l., selected based on features of the atmospheric chemistry data. The initialization time for each location was the closest hour to the time that the HIAPER was in cloud, and the total duration of the trajectories calculated was 72 h. To account for synoptic-scale vertical motion, we used modeled vertical velocities instead of assuming isobaric or isentropic motion.

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In order to test the sensitivity of the trajectory calculations to some of the uncertainties identified above, we used an ensemble approach, where 26 additional trajectories were calculated in addition to the "deterministic" one. These were initialized at horizontal perturbations of Δx and Δy of one grid point (one degree), and $\Delta \sigma$ of 0.01 (about 250 m), which is the standard configuration recommended by the ARL portal.

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3 Analysis of flight data

3.1 **Basic thermodynamic observations**

Figure 4 shows thermodynamic observations from each of the descending profiles from the 1Hz dataset. The slantwise nature of aircraft profiles leaves open the possibility of horizontal inhomogeneity limiting the analysis, but the values for the ascending profiles 20 were not substantially different, except that the HIAPER ascended through a cloud-free patch during profile 2. Profile 1, the northernmost and furthest ahead of the synoptic front. is a classic example of a well-mixed marine boundary layer capped with stratocumulus cloud. There was a strong virtual potential temperature (θ_v) inversion at cloud top of about 6° C. The height of the inversion was about 1150 m, and the temperature at cloud top (CTT), 25

just below the inversion, was about 2°C, so there was no supercooled liquid cloud anywhere

in the layer. Immediately above cloud top the air was very dry, but below cloud base water vapor was well-mixed with a specific humidity (q_v) of about 6 g kg^{-1} . The cloud layer itself was about 400 m deep, and in cloud, ρ_L was near-adiabatic with peak values of around 0.60 g m^{-3} at cloud top. This environment appears to be typical for stratocumulus conditions. Horizontal winds were from the northwest and decreased from 20 to 15 m s^{-1} through the boundary layer, with little directional change at lower levels.

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Profile 2 shares a number of features with the classic example of profile 1, but most notably the θ_v profile is more complex. The boundary layer top (main θ_v increase) was at about 1130 m a.s.l. and was coincident with the cloud top, with a temperature of 2°C. Above

- ¹⁰ this was a cloud-free intermediate layer of about 200 m, with in which the specific humidity (q_v) remained similar to the in-cloud values. There was a weaker θ_v increase at 1320 m a.s.l., which was coincident with a sharp drop in q_v to nearly zero. At cloud top ρ_L was as high as $0.6 \text{ g m}^{-3}0.60 \text{ g m}^{-3}$, and was approximately adiabatic in the upper 300 m of the cloudy layer. Below thisthere was a layer about 250 m deep with relatively consistent values of
- ¹⁵ $p_{L} \simeq 0.25 \text{ g m}^{-3}$, which we interpret, the 10-second smoothed ρ_{L} (not shown) reached a minimum of 0.25 g m^{-3} before increasing briefly to 0.35 g m^{-3} and then dropping rapidly as the aircraft continued to descend. We interpret this feature as a cumulus cell rising into the stratocumulus deck above. Winds below cloud displayed a more or less typical Ekman spiral with a directional shift of about 15° and decrease of 5 m s^{-1} between the cloud base and the lowest flight level.

The intermediate layer between cloud top and the free troposphere had characteristics similar to the "buffer layer" described by Russell et al. (1998), with q_v of similar value to within the boundary layer, but decreasing sharply above the buffer layer. The wind speed was about 5 m s^{-1} lower within this layer than within the boundary layer below or the free troposphere above, but there was no significant directional change. Such intermediate layers were typically identified between the boundary layer and free troposphere in ACE-I. Hande et al. (2012b) identified buffer layers in about 33 % of all Macquarie Island (54.62° S, 158.85° E) soundings, so while profile 2 may differ from the classic structure of profile 1, it is considered to be common for the SO.

Profiles 3 and 4 are quite different in nature to the more typical profiles discussed above. Both profiles had a boundary layer depth of about 1250 m, with a 0.5°C inversion in profile 3 and about 4°C in profile 4.

The cloud that was observed in these profiles occurred in the lower levels where the temperature was exclusively above 0°C. Conditions were considerably more strongly stably stratified than for profiles 1 and 2 (θ_v increased with height but was still conditionally unstable), implying that the boundary layer was less well-mixed. The cloud was co-incident with high wind shear magnitude, in association with an Ekman spiral below 600 m a.s.l., especially in profile 4. While the cloud fields visually resembled stratocumulus layers (see Fig. 3), peak ρ_L values were not located near cloud top as they were in profiles 1 and 2. The cloud field in profile 3 was fairly continuous and flat-topped, but some gaps could be identified during the descent. Cloud top was less well defined in profile 4, with larger broken regions, and highly variable ρ_L values suggesting that clear air was sampled between cloud patches. Patchy mid-level cloud layers were also sampled between 2500 and 4500 m a.s.l. for both of these profiles, but some of these were beyond the altitude range plotted in Fig. 4.

The boundary layer wind speeds for profiles 3 and 4 were very high. Winds of 29 m s^{-1} in profile 3 were observed at around 500 m a.s.l. Above this altitude, winds receded slightly to about 25 m s^{-1} at 1000 m a.s.l. (the boundary layer top), then increased with height to a maximum of about 33 m s^{-1} in the lower free troposphere. The winds at the lowest level of flight were at least 23 m s^{-1} , which is likely in the range for spume production at the ocean surface. Profile 4 was windier still, with peak wind speeds in the boundary layer of nearly 35 m s^{-1} , and the wind speed was consistently greater than 30 m s^{-1} for altitudes above about 250 m a.s.l. At the lowest level of flight, the wind speed was greater than 25 m s^{-1} . Using a log scaling law to translate this to surface conditions, the ten meter winds would have

²⁵ been in the range of 17 to 20 m s⁻¹. Gale force winds speed gale force (≥ 17 m s⁻¹)occur regularly over the SO; weather station data from Macquarie Island, which is nearby in the storm track region, had halfhourly average surface wind speeds greater than this on about 15of days between 2008 and 2011.

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3.2 Microphysics variables

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Profiles of CDP cloud droplet number concentration and mean diameter, as well as UHSAS aerosol number concentration and 2DC-derived rain rate, calculated from the 2DC observations using droplet fall speeds from Pruppacher et al. (1998), are provided in Fig. 5. Where

 $_{5}$ ρ_{L} did not meet the criteria discussed in Sect. 2, data from the CDP and UHSAS are shown with dashed lines. More information about particle size within cloud and below cloud base is given by the particle size distributions (PSDs) in Fig. 6, which we discuss in detail in this section.

The cloud droplet number concentration $N_{\rm C}$ in profile 1 was relatively uniform throughout the cloud with a mean value of $45 \,{\rm cm}^{-3}$, which is perfectly consistent with the established literature (e.g. Boers and Krummel, 1998; Boers et al., 1998; Yum and Hudson, 2004, etc.) on wintertime cloud microphysical conditions over the pristine SO. Cloud droplet mean diameter $\overline{D_{\rm C}}$ increased from about 10 µm near cloud base to about 27 µm near cloud top. The cloud droplet effective radius ($r_{\rm e}$; Hansen and Travis, 1974), calculated from the PSD was 15 14.4 µm. This is just above the threshold suggested by Rosenfeld and Gutman (1994) for

precipitation, and indeed instantaneous rain rates near cloud base of up to $0.2 \,\text{mm}\,\text{h}^{-1}$ were calculated, with drizzle drops of diameter up to $400\,\mu\text{m}$ observed.

The picture was similar for profile 2, where peak $\rho_{\rm L}$ values were comparable, but the mean droplet number concentration was higher ($N_{\rm C} = 77 \,{\rm cm}^{-3}$) and the diameters smaller ($\overline{D_{\rm C}} = 23 \,\mu{\rm m}$ and $r_{\rm e} = 13 \,\mu{\rm m}$ near cloud top). The 2DC-derived rain rate for this cloud was much lower, with maximum values around 0.05 mm h⁻¹.

In profile 3, the HIAPER encountered some broken cloud at 900–1000 m a.s.l., and contiguous cloud between 167 m a.s.l. (the minimum altitude reached) and 700 m a.s.l. In the deeper cloud $\rho_{\rm L}$ was quite variable, but $N_{\rm C}$ was uniformly about 100 cm⁻³ in the top 300 m, and increased to about 150 cm⁻³ between 167 and 400 m a.s.l., with peak values above $200 {\rm cm}^{-3}$. $\overline{D_{\rm C}}$ tended to vary with $\rho_{\rm L}$, and had a average value of 14 µm, and $r_{\rm e}$ near cloud top was about 8.6 µm. Virtually no drops larger than 100 µm were imaged by the 2DC. In profile 4 the HIAPER appears to have flown through patchy or broken cloud, with $\rho_{\rm L}$ falling below both the 0.05 and the $0.01 \,{\rm gm}^{-3}$ thresholds at several points during the profile. Within the patchy cloud it is difficult to establish a representative $N_{\rm C}$ value, because over any given averaging interval there may have been a mixture of clear air and cloud. In a tensecond interval near cloud top, the mean value was $144 \,{\rm cm}^{-3}$, but where $\rho_{\rm L} \ge 0.05 \,{\rm gm}^{-3}$ at the altitude of maximum $\rho_{\rm L}$ the mean value was $188 \,{\rm cm}^{-3}203 \,{\rm cm}^{-3}$. The 1 Hz peak values, which are possibly the best estimate of the "adiabatic" cloud droplet concentration (Yum and Hudson, 2004), were up to $300 \,{\rm cm}^{-3}$. $\overline{D_{\rm C}}$ varied very little from $10-12 \,{\rm \mu}$ m, and $r_{\rm e}$ near cloud top was 7.0 $\,{\rm \mu}$ m. Very few drizzle drops greater than 100 $\,{\rm \mu}$ m diameter were observed in this profile.

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UHSAS aerosol concentration ($N_{\rm U}$; fourth panel of Fig. 5) observations are not a direct measurement of cloud condensation nuclei (CCN) concentrations, but they are all that was available for sub-micron airborne particles during HIPPO-4. One of the first things that we noted was the particularly low concentration of particles immediately above the boundary layer in each of the four profiles. Values of $N_{\rm U} \sim 10-20 \,{\rm cm}^{-3}$ were typical (except for profile 2, see Sect. 4.1). To put these values in context, at similar latitudes in HIPPO-4 RF10, a flight from Midway to Anchorage, Alaska on 7 July 2011, tropospheric $N_{\rm U}$ values (not shown) were typically above $100 \,{\rm cm}^{-3}$ and values above $500 \,{\rm cm}^{-3}$ were observed in two profiles.

²⁰ In profile 1, there is was a large spike in $N_{\rm U}$ at cloud top, with values reaching well above 600 cm⁻³, which is likely an artifact of splashing droplets. We discuss this further in Sect. 5.2. In clear air below the cloud, 60-second averages $N_{\rm U}$ of 74 cm⁻³ 82 cm⁻³ (which corresponded to 1.6 times 1.8 times the in-cloud $N_{\rm C}$), were observed. The median diameter of the observed particles was 0.143 µm.

²⁵ There was a narrow, well-established accumulation mode (usually associated with cloud processing) centered on 0.13 µm, and the broad SSA mode was centered on 0.18 µm. Profile 2 shows a similar but smaller spike in $N_{\rm U}$ at cloud top which we again attribute to droplet breakup. Below-cloud values were on average $\frac{113 \,{\rm cm}^{-3} 110 \,{\rm cm}^{-3}}{10 \,{\rm cm}^{-3}}$, or about $\frac{1.5}{1.4}$ times $N_{\rm C}$, with a broader accumulation mode , with median diameter of $0.174 \,{\rm \mu}m$. The

accumulation mode was centered on 0.15μ m, and the SSA mode was centered on 0.2μ m. Just above the cloud, at about 1500 m a.s.l., N_U reached 100 cm^{-3} in a layer about 300 m deep; an interesting feature which we discuss in Sect. 4.2.

The HIAPER did not descend below cloud base during profile 3, so a size distribution is not shownfor this profile. However, relatively clear air between clouds at 700 and 850 m a.s.l. was sampled. In this gap $N_{\rm U}$ was about $150 \,{\rm cm}^{-3}$, but this might not be representative of below-cloud values.

The The below cloud 60-second average value of $N_{\rm U}$ in the lowest leg of profile 4 was $383 \,{\rm cm}^{-3}$ below cloud base. This is more then double the mean $N_{\rm C}$ value, but as

- ¹⁰ discussed above, 374 cm^{-3} (1.8 times the in-cloud N_{C} might be better represented by values of 200–300 cm⁻³, which would mean that $N_{\text{U}} \simeq 1.5 N_{\text{C}}$. The size of these aerosol particles). The accumulation mode was centered on $0.12 \mu \text{m}$, which is somewhat smaller than for profiles 1 and 2. The SSA mode was centered on $0.15 \mu \text{m}$, which was similar to that in the accumulation mode for this profile.
- ¹⁵ For profiles 1 and 2, with median diameter 0.143μ m the modal decomposition suggested that the SSA mode contributed about 55% of the total $N_{\rm U}$. The contribution of the SSA mode was much greater for profile 4, where it accounted for 70%. In an absolute sense, the SSA mode was more than four times larger in profile 4 than in profiles 1 and 2. This suggests that SSA had a much stronger contribution to the total $N_{\rm U}$ for this profile than the those taken to the north.

3.3 Summary of flight data observations

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During a single flight the HIAPER sampled boundary layer cloud, all at temperatures above 0° C, in a range of different meteorological conditions. In the northernmost profiles (1 and 2), the boundary layer structure was "typical" for the SO: fairly well-mixed (in particular for profile 1) and neutrally stable, and capped with stratocumulus cloud. The microphysical conditions were within the envelope of expected values for the SO. To the south, conditions were much more thermodynamically stable (although still conditionally unstable) and poorly-mixed, and were characterized by high shear in gale-force winds. The values of $N_{\rm C}$

and $N_{\rm U}$ here were well outside the envelope that we would expect for pristine maritime conditions over the SO based on previous in-situ studies. In both the northern and southern profiles, $N_{\rm C}$ in cloud and $N_{\rm U}$ below cloud were related by a factor of about 1.5. Modal decomposition analysis of the UHSAS PSD suggests that SSA dominated the aerosol population for profile 4.

4 Airmass identification

In this section we present an analysis of back trajectories calculated as per Sect. 2.5, at points along the flight path of the HIAPER during HIPPO-4 RF06, to provide further context for our assessment of the microphysical and atmospheric chemistry observations below.

10 4.1 Back trajectories

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Figure 7 shows ensembles of back trajectories for profiles 1 and 2. For profile 1, the most "classic" of the profiles, the westerly motion two to three days before arriving along the flight track occurred while the subtropical ridge was confined to the continent some 48 h earlier (not shown), resulting in strong westerly winds along 40° S. As the blocking anticyclone moved and intensified over Tasmania, the trajectories stagnated and turned southwards with the approach of the cold front. The vertical motion was weakly descending and there was very little spread between the trajectories, as expected in the weak subsidence beneath the anticyclone. None of the ensemble members appear to pass over the mainland, but some cross over the coastline of remote western Tasmania.

- In profile 2, it is evident from the spacing of the 3 hourly markers in the "deterministic" trajectories that the winds were much stronger than for profile 1. Although far displaced from the cold front itself, these trajectories were more clearly driven by the pre-frontal motion, which is shown especially by the gradual ascent (about 1.5 cm s^{-1}) in the 3000 m ensemble during the 20 h prior to arrival, and in the 1500 m ensemble during the 10 h prior to arrival.
- ²⁵ All of the ensembles include trajectories which appear to have spent time over the coastal mainland immediately before strong advection from the north. The 1500 m a.s.l. "determin-

istic" trajectory and a large number of the ensemble members passed in the general vicinity of Port Pirie, a heavy industry center in South Australia, some 24 to 36 h before arriving at the location of profile 2. The same was true for an ensemble arriving at 3000 m, but this is not shown. None of the members for the 4500 m ensemble passed over land.

- The trajectories for profile 3 (Fig. 8) are considerably more complex. Our estimate for the distance ahead of the cold front, accounting for frontal motion between the ERA-Interim analysis at 00:00 UTC (Fig. 1) and the time on location, is about 200 km. It appears that some of the trajectory ensemble members were initialized to the west of (i.e. behind) the cold front, and others to the east, because there was considerable divergence in the air-
- ¹⁰ mass history. For each arrival height, some of the ensemble members originated from around the Nullarbor Plain, an unpopulated and sparsely vegetated coastal region in Western and South Australia; and some members originated over the remote Indian/Southern Ocean and did not pass over any land. The bifurcation is apparent in the trajectory altitude as well: those that originated near the continent (around 130° E, 30° S) ahead of the front ¹⁵ generally had ascending trajectories in the 20 h prior to arrival at profile 3, and those that originated over the remote oceans had descending trajectories.

Profile 4 was performed even closer to the cold front, so there is again substantial divergence in the airmass history. Based on the location provided in the ERA-Interim analysis, the aircraft was about 160 km ahead of the cold front, but the "deterministic" trajectories appear to have been initialized behind the front in the GDAS analysis. Although none of the ensemble members arriving at either 500 or 1500 m a.s.l. passed over directly land, a group arriving at 500 m a.s.l. originated from near the South Australian coastline some 48 h earlier. On the other hand, the group of trajectories with pre-frontal characteristics arriving at 4000 m a.s.l. were over south Western Australia at low altitudes about 24 h earlier.

In summary, the profile with the strongest case for continental/anthropogenic influence is profile 2, which had ensemble members at all levels passing nearby known areas of industrial activity. The likelihood of interaction with continental/anthropogenic aerosol sources decreased for profiles further to the south, where there was some evidence for continental contribution at around 4000 m a.s.l., but not within the boundary layer.

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4.2 Analysis of chemistry data with respect to back trajectories

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Figure 9 shows CO, BC, and N_U for the entire vertical extent of the four profiles. In profile 1, there was a weak increase in CO (up to 60 ppbv) and N_U (up to 75 cm^{-3}) between 2000 and 4000 m, but no signal in BC and minimal interaction with the continental airmass. CO

⁵ also increased with height above 5500m, but there was no signal in either BC or N_U at these levels. The air in profile 1 could be described as very clean, with no clear indication of continental influence.

The strongest chemical signal in profile 2, and indeed all of the profiles, is in a layer between 4000 and 5000 m a.s.l., where elevated, highly correlated CO and BC concentrations were observed. This is characteristic of combustion, but we found that trajectories arriving at 4500 m a.s.l. did not have the clear terrestrial interaction that 1500 and 3000 m (not shown) a.s.l. trajectories had, and aerosol concentrations were quite low ($N_U \simeq 30 \text{ cm}^{-3}$) at these levels. Just above cloud top at 1500 m a.s.l., N_{II} was slightly elevated at 100 cm⁻³, which

corresponded to a small increase in both CO and BC, and the back trajectories at this level
 had a clear terrestrial interaction. In any case, if these observations are indeed evidence of long range transport of anthropogenic pollution, the impact on the aerosol loading was small.

If there was terrestrial interaction for the air sample in profile 3, the signals in the observations were weak. Due to the presence of cloud and precipitation, BC observations were unfortunately missing unavailable at 3000–4000 m a.s.l., but slightly elevated CO concentrations at about 4000 m a.s.l. correspond to increased $N_{\rm U}$ of about 50 cm⁻³ from values of $10-20 \,{\rm cm}^{-3}$ just above the boundary layer.

At 4000 m a.s.l. in profile 4, there was a slight increase in the tropospheric CO concentration, with values up to 60 ppbv, or 10 ppbv higher than the values at the top of the boundary layer. This corresponded to a small peak in the $N_{\rm U}$ concentration of about $80 \,{\rm cm}^{-3}$, but BC observations, which were trending upwards with altitude below this, were missing here as well. Given that some of the trajectory ensemble members originated over land at low altitude, this could be evidence of diluted continental or anthropogenic influence. This feature is clearly decoupled from the boundary layer, as evidenced by $N_{\rm U}$ values strictly below $20 \,{\rm cm}^{-3}$ and CO concentration of 52 ppbv between 1250 and 2000 m a.s.l. Within the boundary layer itself, CO concentrations decrease with height from 54 ppbv increase towards the surface to a maximum of 57 ppbv at 167 m a.s.l. to 50 ppbv at 1250 m a.s.l. This negative GO gradient could be argued to correspond to the group of trajectories that passed near land, but a marine source below a poorly-mixed boundary layer could also account for this . In any Without BC observations at these levels, it is difficult to attribute the source of this CO to human activity (combustion) or natural marine sources. In either case, $N_{\rm U}$ varied much more in the boundary layer than it did near 4000 m a.s.l. for a similar variation in CO, so it is

¹⁰ difficult to attribute the elevated $N_{\rm U}$ to long-range transport.

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4.3 Comparison to clearly polluted/pristine cases

It is useful to provide some context to our discussion of a possible anthropogenic pollution plume by considering some other flights. In HIPPO-4 RF07 (the subsequent flight, two days later), the HIAPER flew from Hobart to Darwin and performed a descending and an ascending profile over the Bass Straight, almost directly south of Melbourne. Conditions (not shown) were somewhat different to profile 1 of RF06, with a deeper boundary layer (capping inversion at 1900 m a.s.l.); lower wind speed (about 10 m s⁻¹); and there was no cloud. The wind direction between 600 and 1500 m a.s.l. was directly from the north, and in this layer the pollution plume from the Melbourne urban area can be unequivocally identified in

Fig. 10, which shows observations for the entire vertical extent of the profiles of θ , CO, BC, and $N_{\rm U}$. Concentrations of CO were about 20–30 ppmv higher within the plume than in the free troposphere. $N_{\rm U}$ of up to 2000 cm⁻³ was observed, and the values were very highly correlated with both BC and CO concentrations (R = 0.87 and R = 0.95 respectively). On the other hand, the free troposphere was extremely clean based on the CO values of about 55 ppbv, BC of virtually zero, and $N_{\rm U}$ of 20–40 cm⁻³.

In HIPPO-3 RF06 (not shown), a return flight from Christchurch to 67°S in the previous campaign (April 2010) and the subject of Chubb et al. (2013), undoubtedly pristine maritime conditions were encountered. Low-level CO concentrations were about 41 ppbv (this may

be a seasonal difference compared to HIPPO-4 RF06), but still varied by about 5 ppbv between 5000 and 7000 m a.s.l. BC concentrations were practically zero in all profiles, and $N_{\rm U}$ concentrations were less than $20 \,{\rm cm}^{-3}$ in the free troposphere but rose to around $100 \,{\rm cm}^{-3}$ in the lowest levels sampled.

With these two comparison flights in mind, HIPPO-4 RF06 (the present flight) is neither an example of a pristine SO environment nor a heavily modified one. As discussed above, there is mixed evidence for anthropogenic influence in each of the profiles. However, the weak signals that can be identified occur well above the boundary layer, where back trajectories can in some cases be used link the history of the air to anthropogenic sources. The air within the boundary layer, on the other hand, does not display an anthropogenic signature that could explain the elevated N_{II} values.

4.4 What about mineral dust?

So far our analysis has concentrated on sources of aerosol associated with combustion, and therefore associated with CO and BC. However, there is the possibility that naturally occurring continental dust could have been the cause of the elevated $N_{\rm U}$ and $N_{\rm C}$ values in profiles 3 and 4. Indeed, dust from the Australian continent has been hypothesized to be an important fertilizing agent for SO phytoplankton (Martin, 1990), and dust samples from Antarctica have been geochemically linked to Australian sources (Revel-Rolland et al., 2006).

²⁰ While the principal sources of Australian dust are further to the east in the Murray–Darling Basin (De Deckker et al., 2010), the Nullarbor Plain is a known secondary source of dust. However, the month of June 2011 was relatively wet in the Nullarbor Plain, temperatures were about average for winter, and wind speeds in the days before HIPPO-4 RF06 were unremarkable. Furthermore, observations of suspended dust are routinely reported at Aus-

tralian Bureau of Meteorology from a number of sites in the Nullarbor (O'Loingsigh et al., 2014), and there were no reports of any suspended dust in the week before the flight (T. O'Loingsigh, personal communication, 2015).

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Another argument comes directly from the trajectories (and as such applies also to the hypothesis of anthropogenic aerosols): the trajectories with continental interaction arriving at 500 and 1500 m a.s.l. in profile 3 are very similar, both in the horizontal and vertical. If the elevated $N_{\rm U}$ values in the boundary layer were due to dust, we should expect to find similar $N_{\rm U}$ values at 1500 m as well, but they were an order of magnitude lower. The same argument applies to profile 4, where trajectories arriving at 4000 m a.s.l. may have been near the surface of south Western Australia about 36 h before, but $N_{\rm U}$ values at these levels were small in comparison to the boundary layer values.

4.5 Can we explain elevated droplet and aerosol concentrations by considering potential anthropogenic or continental sources?

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To summarize the results of this section, we used the combination of back trajectory ensembles with in-situ observations as a tool to identify continental/anthropogenic aerosol influences. In profiles 3 and 4 there was evidence of weak anthropogenic influence between 3000 and 5000 m a.s.l. through the increase of N_U in association with BC and CO. ¹⁵ When compared to profiles through a clear pollution plume in another flight, it is evident that any influence in HIPPO-4 RF06 was highly diluted. Furthermore, given similar CO signals in the boundary layer as in the upper levels, the N_U values were far too high to be attributed to anthropogenic pollution. In addition, we were unable to identify any dust storm activity around the Nullarbor in the week before the flight, and surface observations suggest that ²⁰ dust activity was unlikely. The trajectory analysis suggested that dust, if present, should have resulted in similarly increased N_U values in both the troposphere and the boundary layer, but there was an order of magnitude difference between the two.

The conclusion that we draw from this analysis is that the elevated $N_{\rm C}$ and $N_{\rm U}$ values within the boundary layer can not be predominantly attributed to long-range transport of anthropogenic pollution or continental dust.

5 Evaluation of observational uncertainties

5.1 CDP observations

The accuracy of the CDP is typically stated as $\pm 10\,\%$ due to uncertainties in the true sample volume and in the sizing of small particles through Mie scattering. Using an However, the

- ⁵ <u>PMS-King probe consistently showed about 0.68 of ρ_L from the CDP. Using a parcel ascent</u> model (which conserved θ_e) initialized with conditions near cloud base for profile 1, we calculated the theoretical adiabatic ρ_L for this cloud. This calculation suggested that the value at cloud top would be about 0.64 gm⁻³, very close to the observed CDP value of 0.60 gm⁻³. When all observations within this cloud were compared to the theoretical adiabatic values
- through linear regression, excluding a small region where entrained air was apparent, the observations were found to agree to within 1. This 1%. This analysis, combined with the beam mapping performed by the manufacturer, suggests very strongly that the CDP observations were robust.

Even if the CDP did significantly overestimate ρ_L —which we believe to be unlikely—it may have also overestimated N_C by the same fraction, depending on the cause of the error. However, this would ultimately have little impact on our conclusions, because it would still be necessary to explain a factor of five increase in N_C between profiles 1 and 4.

5.2 UHSAS observations in ambiguous conditions

Vidaurre and Hallett (2009) established droplet breakup criteria upon impact with a cylin drical surface based on the Weber number, or the ratio of particle impact kinetic energy to surface energy. This depended primarily on particle diameter and speed of impact (which is in turn dependent on the inlet geometry). For a representative airspeed of the HIAPER and geometry of the UHSAS inlet, their criteria predict that droplet breakup should be minimal for droplets with diameters under about 8 µm, but severe for droplets with diameters over about 20 µm.

In profiles 1 and 2, the air beneath the cloud was unequivocally clear, with zero $N_{\rm C}$ observed by the CDP, so we have no concerns about droplet splashing affecting these data.

In profile 3, the cloud-free $\rho_L < 0.01 \,\text{gm}^{-3}$ threshold was never met in the lowest leg and droplet diameters of 10–12 µm were observed. These droplet sizes could have caused splashing on the UHSAS inlet so we do not consider these data. However there was some suitably clear air for a short interval between clouds during descent, and we consider these data to be usable, although perhaps not representative of below-cloud values.

The case for the robustness of the UHSAS data in profile 4 is much better. The air sampled at during the lowest leg met our "probably clear" criterion of $\rho_L < 0.05 \,\mathrm{gm^{-3}}$. However, there was a non-zero cloud droplet concentration in most one-second intervals, and N_C was in the range of 6–10 cm⁻³ (ρ_L was $0.001-0.005 \,\mathrm{gm^{-3}}$), with particles of mean diameter 6–7 µm. We interpret these conditions as hazy sub-cloud air, and according to the work of Vidaurre and Hallett (2009), we expect that droplet splashing should not affect the 15 observations. Indeed, setting the "probably clear" ρ_L threshold as low as $0.002 \,\mathrm{gm^{-3}}$ revealed very little sensitivity in N_C . We are thus highly confident that the average values of $N_U \simeq 383 \,\mathrm{cm^{-3}}$ below cloud base were indeed reliable for profile 4.

5.3 Uncertainties in back trajectories

In general, the accuracy of back trajectories depends on the accuracy of the wind fields in the gridded data, but is also influenced by temporal and spatial resolution of the product used (Rolph and Draxler, 1990). For trajectories over the open ocean, the 3hourly one degree dataset used is sufficient to resolve the synoptic scale features which are dominant, at least in the free troposphere.

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The way that vertical motion is handled can be important: horizontal wind components generally vary with height, so any vertical displacement errors will contribute to horizontal displacement errors as well. Vertical winds are generally deduced from the divergence of the horizontal components and can be noisy. It is possible to calculate isentropic trajectories, which follow surfaces of constant θ , which is a good approximation to the motion

- We are interested in boundary layer conditions for the purposes of this paper, but acknowledge that boundary layer trajectories are especially prone to the uncertainties mentioned above, especially in highly sheared environments. In addition, the representation of boundary layer structure over the SO in global reanalyses is known to be questionable (e.g. Hande et al., 2012b; Huang et al., 2015). We chose to analyze trajectories arriving at multiple levels, with two of the three ensembles for each profile initialized above the boundary layer, with this concern in mind.
- The task of simulating back trajectories for HIPPO-4 RF06 presents an additional challenge in that the region is experiencing a rapid dynamical change in the form of an approaching cold front. The heterogeneity of the wind field in this situation compounds the uncertainties in the back trajectory due to inaccuracies in the meteorological analysis. We used an ensemble approach to represent the uncertainty in the airmass history. For profiles 1 and 2, the location was sufficiently far from the cold front that the wind field heterogeneity did not overly influence the back trajectories. There was still some variability amongst the ensemble members, but the "deterministic" trajectories should be accurate. For profiles 3 and 4, the perturbation in the initialization points was sufficient to straddle the cold front, resulting in two "clusters" of trajectories for each ensemble. The spread in each of these clusters was comparable to the spread of the entire ensembles for profiles 1 and 2. The in-situ observations suggest that the profiles were performed ahead of the cold front, which suggests that the more northerly clusters were the most representative.

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6 **Discussion**

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6.1 Summary of arguments presented in this paper

Downey et al. (1990) argued that long range transport of aerosol from the Australian continent was responsible for cases of high CN concentrations at Macquarie Island, but it seems that a direct hit on an urban center was required, and they reported high correlation with radon (a continental tracer) in these instances. We used CO and BC as continental/anthropogenic markers, and showed that they were very effective in identifying the pollution plume from Melbourne in HIPPO-4 RF07.

Of the four vertical profiles performed in HIPPO-4 RF06, the one with the most convincing signature of anthropogenic/continental influence was profile 2, where a clear correlation between CO and BC was observed, possibly in several layers. The strongest signature was found between 4000 and 5000 m a.s.l., but even here there was no correlation to the submicron particle number concentration $N_{\rm U}$. Better correlation between the three values was identified at 1500 m a.s.l., where peak $N_{\rm U}$ values were over $100 \,\mathrm{cm}^{-3}$, and back trajectories clearly suggest a terrestrial pollution source. In the boundary layer clouds sampled in profile 2, mean cloud droplet number concentrations were about 77 cm⁻³, which is not particularly unusual for the remote SO.

For profiles 3 and 4 we have argued that the most likely signature of anthropogenic/continental influence was at 3000-4000 m a.s.l., which was well above and decou-

- ²⁰ pled from the boundary layer , and that by a layer of clean air. Furthermore, $N_{\rm U}$ at those levels (about 80 cm⁻³) was insignificant in comparison to values within the boundary layer (nearly 400 cm⁻³). The trajectories that arrived in the boundary layer for profiles 3 and 4 show much weaker evidence for anthropogenic/continental influence, and were not coincidental with a trajectory from any industrial/urban centers. Yet the $N_{\rm C}$ and $N_{\rm U}$ values in
- ²⁵ profile 4 were about twice those of profile 2, and up to four times the values for profile 1. The very small increase in CO in the boundary layer may <u>equally</u> suggest a highly diluted anthropogenic signature , but or a natural maritime source, but in any case it is not nearly of sufficient magnitude to explain the $N_{\rm U}$ values. The microphysical results for profile 3 were

similar, but we have somewhat less confidence in the UHSAS concentrations due to the potential for artifacts due to splashing droplets.

While $N_{\rm C}$ values of 150–300 cm⁻³ are by no means exceptional in a global context, they are unexpected for the pristine maritime environment of the SO, especially during wintertime when ocean productivity is lowest. If the hypothesis that such values were predominantly 5 caused by long range transport of continental pollutants can be rejected, as we argue in this paper, then we are left with the conclusion that the elevated particle concentration observed by the UHSAS, which probably includes most of the CCN, was produced locally. We While alternative sources for the CCN cannot be completely ruled out without compositional

analysis of the aerosol, we showed through modal decomposition of the UHSAS PSD for 10 profile 4 that the mode representing primary marine aerosol, which is dominated by SSA, to be the main contributor to the aerosol population. We thus consider sea spray aerosol to be the best candidate to explain the elevated aerosol concentrations, and indeed there are many studies that suggest that SSA can dominate the marine boundary layer CCN population (e.g. Clarke et al., 2006; Murphy et al., 1998). 15

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6.2 General discussion of results

Our conclusions contrast with the findings of Blot et al. (2013), which suggest that wind speed was not a factor in controlling SSA concentrations in the VOCALS campaign, and other authors (Bates et al., 1998; Covert et al., 1998; Berg et al., 1998) have also reported poor correlations of SSA concentrations with wind speed. However, the low level wind speeds of $25-35 \text{ m s}^{-1}$ encountered during profiles 3 and 4 was extreme, and well outside the ranges reported by Blot et al. (2013). Because the background aerosol concentrations were so low in this region, the additional SSA production would have had a significant impact on overall CCN as well as $N_{\rm C}$.

This result is of interest in discussions of the cloud structure and radiation bias over 25 the SO. Strong boundary layer Gale force winds, such as those encountered in HIPPO-4 RF06, are a regular occurrence occur regularly over the SO-; weather station data from Macquarie Island, which is nearby in the storm track region, had half hourly average surface

wind speeds greater than this on about 15% of days between 2008 and 2011. Moreover, Korhonen et al. (2010) showed an increase in wind speed in the latitude band $50-65^{\circ}$ S from 1980 to 2002 in reanalysis data, which has been verified observationally by Hande et al. (2012a). Over the same period, modeled CCN concentrations increased by 19% on average, and they found that wind speed accounted for 48% of the variance and was the most important cause of the changes. The resultant negative radiative forcing in this latitudinal band was on the same order as the positive forcing due to greenhouse gases.

The CCN concentrations for the study of Korhonen et al. (2010) were derived from a global aerosol model which includes a wind speed dependent SSA parameterization, but the . The basis for such parameterizations has been questioned by several authors, as discussed in this paper. Nevertheless, the hypothesis that SSA could be a factor in such a climate feedback mechanism is supported by the observations of HIPPO-4 RF06. Targeted observations are clearly needed to more convincingly address this hypothesis, which fits squarely within the stated priorities of the NSF Advisory Committee for Geosciences (2014).

7 Conclusions

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In this paper, we have presented thermodynamic, microphysical and atmospheric chemistry observations from vertical profiles performed during HIPPO-4 RF06. Large variation in microphysical characteristics of the boundary layer clouds and the aerosol concentration were found, and in particular the CDP cloud droplet number concentration $N_{\rm C}$ and UHSAS aerosol concentration $N_{\rm U}$ were substantially higher (by a factor of two to five) than expected for the southernmost profiles. At these latitudes the wind speeds were the most extreme, at 25–35 m s⁻¹ at very low altitudes (about 167 m a.s.l.). We were unable to attribute these observations to continental/anthropogenic sources through the analysis of the atmospheric chemistry data and back trajectories, although there were indications of weak impacts at much higher altitudes in the profiles. We conclude that these observations are consistent with the local production of sea spray aerosol through the due to high winds in the southernmost regions of the flightis the most likely explanation for these observations. In order to reduce ambiguities such as those discussed in this paper, we strongly recommend the inclusion of aerosol chemical composition measurements for future cloud physics observational missions over the Southern Ocean.

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References

15

Bates, T. S., Kapustin, V. N., Quinn, P. K., Covert, D. S., Coffman, D. J., Mari, C., Durkee, P. A., De Bruyn, W. J., and Saltzman, E. S.: Processes controlling the distribution of aerosol particles in the lower marine boundary layer during the First Aerosol Characterization Experiment (ACE 1), J. Geophys. Res.-Atmos., 103, 16369–16383, 1998.

Baumgardner, D. and Korolev, A.: Airspeed corrections for optical array probe sample volumes, Journal of Atmospheric and Oceanic Technology, 14, 1224–1229, 1997.

- Bennartz, R.: Global assessment of marine boundary layer cloud droplet number concentration from satellite, J. Geophys. Res.-Atmos., 112, D02201, doi:10.1029/2006JD007547, 2007.
 - Berg, O. H., Swietlicki, E., and Krejci, R.: Hygroscopic growth of aerosol particles in the marine boundary layer over the Pacific and Southern Oceans during the First Aerosol Characterization
- Experiment (ACE 1), J. Geophys. Res.-Atmos., 103, 16535–16545, 1998.
 Blot, R., Clarke, A. D., Freitag, S., Kapustin, V., Howell, S. G., Jensen, J. B., Shank, L. M., Mc-Naughton, C. S., and Brekhovskikh, V.: Ultrafine sea spray aerosol over the southeastern Pacific: open-ocean contributions to marine boundary layer CCN, Atmos. Chem. Phys., 13, 7263–7278, doi:10.5194/acp-13-7263-2013, 2013.
- Boers, R. and Krummel, P. B.: Microphysical properties of boundary layer clouds over the Southern Ocean during ACE 1, J. Geophys. Res.-Atmos., 103, 16651–16663, 1998.
 - Boers, R., Jensen, J., Krummel, P., and Gerber, H.: Microphysical and short-wave radiative structure of wintertime stratocumulus clouds over the Southern Ocean, Q. J. Roy. Meteor. Soc., 122, 1307–1339, 1996.

- Boers, R., Jensen, J., and Krummel, P.: Microphysical and short-wave radiative structure of stratocumulus clouds over the Southern Ocean: summer results and seasonal differences, Q. J. Roy. Meteor. Soc., 124, 151–168, 1998.
- Boutle, I. A., Abel, S. J., Hill, P. G., and Morcrette, C. J.: Spatial variability of liquid cloud and rain: ob-
- servations and microphysical effects, Q. J. Roy. Meteor. Soc., 140, 583–594, doi:10.1002/qj.2140, 2014.
 - Cai, Y., Montague, D. C., Mooiweer-Bryan, W., and Deshler, T.: Performance characteristics of the ultra high sensitivity aerosol spectrometer for particles between 55 and 800 nm: laboratory and field studies, J. Aerosol Sci., 39, 759–769, 2008.
- ¹⁰ Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, Nature, 326, 655–661, 1987.
 - Chubb, T. H., Jensen, J. B., Siems, S. T., and Manton, M. J.: In situ observations of supercooled liquid clouds over the Southern Ocean during the HIAPER Pole-to-Pole Observation campaigns, Geophys. Res. Lett., 40, 5280–5285, 2013.
- ¹⁵ Clarke, A. D., Owens, S. R., and Zhou, J.: An ultrafine sea-salt flux from breaking waves: implications for cloud condensation nuclei in the remote marine atmosphere, J. Geophys. Res.-Atmos., 111, D06202, doi:10.1029/2005JD006565, 2006.
 - Covert, D. S., Gras, J. L., Wiedensohler, A., and Stratmann, F.: Comparison of directly measured CCN with CCN modeled from the number-size distribution in the marine boundary layer during
- ACE 1 at Cape Grim, Tasmania, J. Geophys. Res.-Atmos., 103, 16597–16608, 1998.
 - Day, J.: Production of droplets and salt nuclei by the bursting of air-bubble films, Q. J. Roy. Meteor. Soc., 90, 72–78, 1964.
 - De Deckker, P., Norman, M., Goodwin, I. D., Wain, A., and Gingele, F. X.: Lead isotopic evidence for an Australian source of aeolian dust to Antarctica at times over the last 170,000 years, Palaeo-
- ²⁵ geography, Palaeoclimatology, Palaeoecology, 285, 205–223, doi:10.1016/j.palaeo.2009.11.013, 2010.
 - de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.: Production flux of sea spray aerosol, Rev. Geophys., 49, RG2001, doi:10.1029/2010RG000349, 2011.
- ³⁰ Downey, A., Jasper, J., Gras, J., and Whittlestone, S.: Lower tropospheric transport over the Southern Ocean, J. Atmos. Chem., 11, 43–68, 1990.
 - Draxler, R. and Hess, G.: An overview of the HYSPLIT_4 modelling system for trajectories, dispersion and deposition, Aust. Meteorol. Mag., 47, 295–308, 1998.

Discussion Paper

- Gerbig, C., Schmitgen, S., Kley, D., Volz-Thomas, A., Dewey, K., and Haaks, D.: An improved fastresponse vacuum-UV resonance fluorescence CO instrument, J. Geophys. Res.-Atmos., 104, 1699–1704, 1999.
- Grosvenor, D. P. and Wood, R.: The effect of solar zenith angle on MODIS cloud optical and mi-
- ⁵ crophysical retrievals within marine liquid water clouds, Atmos. Chem. Phys., 14, 7291–7321, doi:10.5194/acp-14-7291-2014, 2014.
 - Hande, L., Siems, S., and Manton, M.: Observed trends in wind speed over the Southern Ocean, Geophys. Res. Lett., 39, L11802, doi:10.1029/2012GL051734, 2012a.
 - Hande, L., Siems, S., Manton, M., and Belusic, D.: Observations of wind shear over the Southern Ocean, J. Geophys. Res.-Atmos., 117, doi:10.1029/2012JD017488, 2012b.

10

30

- Hansen, J. E. and Travis, L. D.: Light scattering in planetary atmospheres, Space Sci. Rev., 16, 527–610, 1974.
 - Huang, Y., Franklin, C. N., Siems, S. T., Manton, M. J., Chubb, T., Lock, A., Alexander, S., and Klekociuk, A.: Evaluation of boundary-layer cloud forecasts over the Southern Ocean in a limited-area
- numerical weather prediction system using in situ, space-borne and ground-based observations,
 Q. J. Roy. Meteor. Soc., 141, 2259–2276, doi:10.1002/qj.2519, 2015.
 - Kassianov, E., Berg, L. K., Pekour, M., Barnard, J., Chand, D., Flynn, C., Ovchinnikov, M., Sedlacek, A., Schmid, B., Shilling, J., et al.: Airborne aerosol in situ measurements during TCAP: A closure study of total scattering, Atmosphere, 6, 1069–1101, doi:10.3390/atmos6081069, 2015.
- ²⁰ King, W., Parkin, D., and Handsworth, R.: A hot-wire liquid water device having fully calculable response characteristics, Journal of Applied Meteorology, 17, 1809–1813, 1978.
 - Korhonen, H., Carslaw, K. S., Forster, P. M., Mikkonen, S., Gordon, N. D., and Kokkola, H.: Aerosol climate feedback due to decadal increases in Southern Hemisphere wind speeds, Geophys. Res. Lett., 37, L02805, doi:10.1029/2009GL041320, 2010.
- Lance, S., Brock, C. A., Rogers, D., and Gordon, J. A.: Water droplet calibration of the Cloud Droplet Probe (CDP) and in-flight performance in liquid, ice and mixed-phase clouds during ARCPAC, Atmos. Meas. Tech., 3, 1683–1706, doi:10.5194/amt-3-1683-2010, 2010.
 - Lewis, E. R. and Schwartz, S. E.: Sea salt aerosol production: mechanisms, methods, measurements, and models – a critical review, EOS T. Am. Geophys. Union, doi:10.1002/9781118666050, 2004.
 - Marks, R.: Preliminary investigations on the influence of rain on the production, concentration, and vertical distribution of sea salt aerosol, J. Geophys. Res.-Oceans, 95, 22299–22304, 1990.

- Martin, J. H.: Glacial-interglacial CO₂ change: the Iron Hypothesis, Paleoceanography, 5, 1–13, 1990.
- Meehl, G. A., Covey, C., Taylor, K. E., Delworth, T., Stouffer, R. J., Latif, M., McAvaney, B., and Mitchell, J. F.: The WCRP CMIP3 multimodel dataset: a new era in climate change research, B.
- Am. Meteorol. Soc., 88, 1383–1394, 2007. 5

15

20

30

Modini, R., Frossard, A., Ahlm, L., Russell, L., Corrigan, C., Roberts, G., Hawkins, L., Schroder, J., Bertram, A., Zhao, R., et al.: Primary marine aerosol-cloud interactions off the coast of California, Journal of Geophysical Research: Atmospheres, 2015.

Murphy, D. M., Anderson, J. R., Quinn, P. K., McInnes, L. M., Brechtel, F. J., Kreidenweis, S. M.,

- Middlebrook, A. M., Pósfai, M., Thomson, D. S., and Buseck, P. R.: Influence of sea-salt on 10 aerosol radiative properties in the Southern Ocean marine boundary layer, Nature, 392, 62-65, doi:10.1038/32138, 1998.
 - Nakajima, T. and King, M. D.: Determination of the optical thickness and effective particle radius of clouds from reflected solar radiation measurements. Part I: Theory, J. Atmos. Sci., 47, 1878–1893, 1990.
 - Nilsson, E., Rannik, Ü., Swietlicki, E., Leck, C., Aalto, P. P., Zhou, J., and Norman, M.: Turbulent aerosol fluxes over the Arctic Ocean: 2. Wind-driven sources from the sea. J. Geophys. Res.-Atmos., 106, 32139-32154, 2001.

NSF Advisory Committee for Geosciences: Dynamic Earth: GEO Imperatives and Frontiers 2015-2020, Tech. rep., National Science Foundation, Arlington, VA 22230, USA, 2014.

- O'Dowd, C. D. and Smith, M. H.: Physicochemical properties of aerosols over the northeast Atlantic: evidence for wind-speed-related submicron sea-salt aerosol production, J. Geophys. Res.-Atmos., 98, 1137–1149, 1993.
 - O'Loingsigh, T., McTainsh, G., Tews, E., Strong, C., Leys, J., Shinkfield, P., and Tapper, N.: The
- Dust Storm Index (DSI): a method for monitoring broadscale wind erosion using meteorological 25 records, Aeolian Res., 12, 29-40, 2014.
 - Pruppacher, H. R., Klett, J. D., and Wang, P. K.: Microphysics of clouds and precipitation, Kluwer Academic Publishers, P.O. Box 17, 3300 AA Dordrecht, the Netherlands, 1998.
 - Quinn, P. and Bates, T.: The case against climate regulation via oceanic phytoplankton sulphur emissions, Nature, 480, 51-56, 2011.
- Revel-Rolland, M., De Deckker, P., Delmonte, B., Hesse, P., Magee, J., Basile-Doelsch, I., Grousset, F., and Bosch, D.: Eastern Australia: a possible source of dust in East Antarctica interglacial ice, Earth Planet. Sc. Lett., 249, 1-13, 2006.

Discussion Paper

Microphysics Flight-Level Data, Tech. rep., UCAR/NCAR – Earth Observing Laboratory, Boulder, CO 80307, USA, available at: http://www.eol.ucar.edu/system/files/PM_notes_HIPPO-4_-5.doc, (last access: 31 August 2015), 2012.

Rosenfeld, D. and Gutman, G.: Retrieving microphysical properties near the tops of potential rain clouds by multispectral analysis of AVHRR data, Atmos. Res., 34, 259–283, 1994.

Russell, L. M., Lenschow, D. H., Laursen, K. K., Krummel, P. B., Siems, S. T., Bandy, A. R., Thorn-

- ton, D. C., and Bates, T. S.: Bidirectional mixing in an ACE 1 marine boundary layer overlain by a second turbulent layer, J. Geophys. Res.-Atmos., 103, 16411–16432, 1998.
 - Salomonson, V. V., Barnes, W., Maymon, P. W., Montgomery, H. E., and Ostrow, H.: MODIS: advanced facility instrument for studies of the Earth as a system, IEEE T. Geosci. Remote, 27, 145–153, 1989.
- Schwarz, J. P., Gao, R. S., Fahey, D. W., Thomson, D. S., Watts, L. A., Wilson, J. C., Reeves, J. M., Darbeheshti, M., Baumgardner, D. G., Kok, G. L., Chung, S. H., Schulz, M., Hendricks, J., Lauer, A., Kärcher, B., Slowik, J. G., Rosenlof, K. H., Thompson, T. L., Langford, A. O., Loewenstein, M., and Aikin, K. C.: Single-particle measurements of midlatitude black carbon and light-scattering aerosols from the boundary layer to the lower stratosphere, J. Geophys. Res.-Atmos., 414, D10007, doi:10.1000/000010007070_00000
- ²⁰ 111, D16207, doi:10.1029/2006JD007076, 2006.

5

30

- Trenberth, K. E. and Fasullo, J. T.: Simulation of present-day and twenty-first-century energy budgets of the southern oceans, J. Climate, 23, 440–454, 2010.
 - Vidaurre, G. and Hallett, J.: Particle impact and breakup in aircraft measurement, J. Atmos. Ocean. Tech., 26, 972–983, 2009.
- ²⁵ Wofsy, S.: HIAPER Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically important atmospheric gases and aerosols, Philos. T. R. Soc. A, 369, 2073–2086, 2011.
 - Wofsy, S. C., Daube, B. C., Jimenez, R., Kort, E., Pittman, J. V., Park, S., Commane, R., Xiang, B., Santoni, G., Jacob, D., Fisher, J., Pickett-Heaps, C., Wang, H., Wecht, K., Wang, Q.-Q., Stephens,
 - B. B., Shertz, S., Watt, A. S., Romashkin, P., Campos, T., Haggerty, J., Cooper, W. A., Rogers, D., Beaton, S., Hendershot, R., Elkins, J. W., Fahey, D. W., Gao, R. S., Moore, F., Montzka, S. A., Schwarz, J. P., Perring, A. E., Hurst, D., Miller, B. R., Sweeney, C., Oltmans, S., Nance, D., Hintsa, E., Dutton, G., Watts, L. A., Spackman, J. R., Rosenlof, K. H., Ray, E. A., Hall, B., Zondlo,

M. A., Diao, M., Keeling, R., Bent, J., Atlas, E. L., Lueb, R., and Mahoney, M. J.: HIPPO Merged 10-second Meteorology, Atmospheric Chemistry, and Aerosol Data (R 20121129), Tech. rep., Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, doi:10.3334/CDIAC/hippo 010, 2012.

- Wood, R. and Field, P. R.: The distribution of cloud horizontal sizes, J. Climate, 24, 4800-4816, 5 2011.
 - Woodcock, A. H.: Salt nuclei in marine air as a function of altitude and wind force, J. Meteorol., 10, 362-371, 1953.

Yum, S. S. and Hudson, J. G.: Wintertime/summertime contrasts of cloud condensation nuclei

and cloud microphysics over the Southern Ocean, J. Geophys. Res.-Atmos., 109, D06204, 10 doi:10.1029/2003JD003864, 2004.



Figure 1. ERA-Interim mean sea level pressure at 00:00 UTC on 29 June 2011, with synoptic features as analyzed by the Australian Bureau of Meteorology, 950 hPa wind speed (colors; $m s^{-1}$), and 950 hPa temperatures (dashed red contours at 5, 10 and 15° C). The flight track for HIPPO-4 RF06 is shown in magenta.



Figure 2. Flight track for HIPPO-4 RF06, with MODIS cloud top temperature overlay. The locations of the lowest level of the four profiles are labeled as per the text.



Figure 3. Forward camera imagery of boundary layer cloud top conditions for the four profiles as labeled.



20 30 40

WSC ($m s^{-1}$)

280 290 300 310 320 330

WDC (deg)

Figure 4. Profiles of thermodynamic variables within and above the boundary layer for RF06. From left: virtual potential temperature (θ_v); specific humidity (q_v); CDP liquid water content (ρ_L); wind speed (WSC); and wind direction (WDC). The colors indicate the profile number and location (see top panel). Note that for display purposes, the values for some profiles have been offset by the amount indicated in the legend in each panel.

0.0 0.2 0.4 0.6 0.8 1.0 10

 ho_L (g m⁻³)

+0

HIAPER Altitude (m)

280 290

300 310 320

 θ_{v} (K)

0 5 10 15 20 25 30 35 40

 q_v (g kg⁻¹)

+10



Figure 5. Profiles of microphysics variables within and above the boundary layer for RF06, with offsets for each profile indicated in the legend. From left: virtual potential temperature (θ_v , repeated for reference to other variables); CDP number concentration (N_c , masked dashed where $\rho_L < 0.05 \text{ g m}^{-3}$); CDP mean diameter ($\overline{D_c}$, masked dashed where $\rho_L < 0.05 \text{ g m}^{-3}$); UHSAS number concentration (N_U ; masked dashed where $\rho_L > 0.01 \text{ g m}^{-3}$); and 2DC-derived rain rate (RR).

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Figure 6. Left: Particle size distribution distributions (10averagesPSDs) for aerosol observed below cloud base by the UHSAS during HIPPO-4. Right Upper left panel: 10 second average cloud droplets droplet PSDs observed by the CDP at near cloud top, and drizzle drops drop PSDs observed by the 2DC near cloud base(also 10averages; note). Note the logarithmic y-y scale for this panel. Upper right, and lower panels: 60 second average PSDs for UHSAS data for profiles 1, 2 and 4 as labelled, with fitted modes shown by dashed lines. The contribution to the observed $N_{\rm U}$ for each mode is shown in the legend, and the cutoff size used for identifying the SSA mode marked by a black dashed line (see text). Note the different scales on the y-axes.

Figure 7. 72 h HYSPLIT Back trajectory ensembles for profiles 1 and 2, with arrival heigths indicated in the legends. Back trajectories were calculated with 1° horizontal resolution Global Data Assimilation System meteorological data, with the different ensemble members representing perturbations from the aircraft location of 1 grid space in the horizontal and 0.01σ (about 250m) in the vertical. The "deterministic" trajectories are heavier weighted lines with three hourly circle markers for each ensemble.

Figure 8. As for Fig. 7, but for profiles 3 and 4. In this figure some additional ensemble members, which represent substantially different airmass histories from the "deterministic" trajectories, have been highlighted with 12 hourly diamond markers.

Figure 9. Trace gas and aerosol profiles for comparison flight RF06. From left: potential temperature (θ , note offset of 10K between profiles); Aerolaser VUV resonance fluorescence carbon monoxide concentration (CO); SP2 black carbon (BC) mass concentration; and UHSAS number concentration (N_U ; missing for in-cloud conditions). Note that the y scale is different to Figs. 4 and 5, and now shows the entire vertical extent of the profiles.

Figure 10. As for Fig. 9, but for RF07, showing a clear example of a polluted plume over the Bass Straight to the south of Melbourne. Note different x scales for CO, BC, and N_U compared to Fig. 9.