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

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1 Anonymous Referee #1

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Review of Observations of high droplet number concentrations in Southern Ocean boundary layer clouds by Chubb et al.
Recommendation: Requires minor revision before acceptance in ACP.

1.1 General Comments

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

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

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

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

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 manuscript, namely we conclude that local production of sea spray aerosol through the high winds in the southernmost regions of the flight is the most likely explanation for these observations. I think it would be better to state that the observations are consistent with the high winds causing the production of the sea salt aerosol, because this is really inferred from the data rather than establishing a relationship between these variables. I think this change in language is needed because the authors do admit that there is some uncertainty in the trajectory analysis.

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 these observations are consistent with the local production of sea spray aerosol through the due to high winds in the southernmost regions of the flight, which is 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 (wind force ≥ 17 m s\(^{-1}\)) occur regularly over the SO. A weather station data from Macquarie Island, which is near the storm track region, had half-hourly average surface wind speeds greater than this on about 15% of days between 2008 and 2011.

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

   This result is of interest ... (snip) ... . Strong boundary layer Gale force winds, such as those encountered in HIPPO-4 RF06, are a regular occurrence. A weather station data from Macquarie Island, which is near the storm track region, had half-hourly average surface wind speeds greater than this on about 15% of days between 2008 and 2011. Moreover, ... (snip)

In terms of the microphysics data and the uncertainties, I was surprised that there were no statements about how the bulk water content derived from the size distributions compared to that measured by a bulk water probe. I am assuming there must have been some sort of King or hotwire probe on the G-V. This would be a basic test that could help verify that the CDP is well calibrated (especially since some of the channel boundaries 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 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 approximately twice that of the King probe. This was in spite of the standard calibration using glass beads during the HIPPO-4 campaign and subsequent post-processing (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 mapping is a relatively new technique which evaluates the true sample area of the specific instrument (as opposed to the “theoretical” sample area which had been used previously).
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 mm$^2$, but the subsequent beam mapping of CDP #16 showed that the sample area was 0.309 mm$^2$. We recalculated the droplet concentration and liquid water content using the measured beam area for this paper.

Data from profile 1 plotted against altitude (m a.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 $\theta_e$) 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:

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,\(^a\) 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:

1. 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
could affect \(\rho_{L,\text{CDP}}\) without affecting the CDNC. Neither of these possibilities
change our conclusions in any way, because in the case of (1.), we would still need
to explain the factor of five (or more) difference in the CDNC for profiles 1 and 4, and in the case of (2.), the CDNC data are unaffected. Following our intensive
quality control of the CDP data we elected to present these in order that our results
could be fully reproducible. However, we agree that it is worth mentioning that
these analyses have been performed in the manuscript, in such a way that it does
not distract from our message.

We have made the following changes to the manuscript:

1. Item added to section 2.2: to describe bulk water measurements:
   - \textit{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 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 \(\rho_{L,\text{King}}\).}

2. Sentences added to section 5.1:
   - 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 \(\rho_L\) \textit{from the CDP. Using a parcel ascent ... (snap)}
   - Even if the CDP did significantly overestimate \(\rho_L\)—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.

3. New bibliography item added for King et al. (1978).

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:

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 ... (snip)

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

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.

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

1. Changes to section 2.2:

   - Particle Measurement Systems (PMS) 2-D Cloud Imaging Probe (2DC). Precipitation particles larger than about ... (snip) ... 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 µm (Baumgardner and Korolev [1997], but we made no specific correction for this other than only using particles with diameters ... (snip)

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 \( \rho_L \) and \( N_C \) 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 \( \rho_L \) and \( N_C \) 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 m s\(^{-1}\) at altitudes above 600 m, and 2.5 m s\(^{-1}\) below this, at a true air speed of 130 m s\(^{-1}\). 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 profile data, except in profile 2 where there was no cloud sampled in the ascending profile. We do not anticipate major effects from this factor, but have highlighted these details in the revised manuscript. Changes:

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

   The aircraft performed four descent/ascent profiles ... (snip) ... 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 ... (snip)

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

   Figure 4 shows thermodynamic observations from each of the descending profiles from the 1 Hz dataset. The slantwise nature of aircraft profiles leaves open the possibility of horizontal inhomogeneity limiting the analysis, but the values for the ascending profiles were not ... (snip)

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

Author Response: The maximum values for \( N_C \) were included for profiles 3 and 4 but not for profiles 1 and 2 simply because there was much more variability in profiles 3 and 4. The maximum values are particularly important for profile 4 because \( N_C \) was correlated with \( \rho_L \) for this profile only, indicating that entrainment was important. We are reluctant to introduce more values in to the descriptions for profiles 1 and 2 because they could be distracting, and the values could be read from the graphs if readers are interested.

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

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

Author Response: If you are referring to the cloud layer above 2400 m a.s.l. in profile 3, we think that this is highly unlikely. There was no evidence of any precipitation particles above the boundary layer cloud top, and there was a vertical displacement of nearly 1500 m between the cloud layers in this instance.

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

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

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

   • **DMT Ultra High Sensitivity Aerosol Spectrometer (UHSAS).** The UHSAS measures sizes of aerosol ... (snip). 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). ... (snip)


### 2 Anonymous Referee #2

Received and published: 29 October 2015

#### 2.1 General comments

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

2.1.1 Direct evidence for the conclusion is lacking

Direct evidence for our conclusion that 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:

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

   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)

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

   While NC values of ... (snip) ... 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 ... (snip)

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 $N_U$.

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.

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 $N_U$ of about 50 cm$^{-3}$ from values of 10–20 cm$^{-3}$ just above the boundary layer.

2. 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 of 57 ppbv at 167 m a.s.l. This negative CO 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 case, 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, $N_U$ 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 difficult to attribute the elevated $N_U$ to long-range transport.

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

The changes to the manuscript as a result of the new analysis are as follows:
2.1.4 Shift of UHSAS data to dry diameters

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

1. Added sentence to UHSAS item in section 2.2:

   • DMT Ultra High Sensitivity Aerosol Spectrometer (UHSAS). The UHSAS measures sizes of aerosol ...(snip). 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). ...(snip)

2.1.5 Softening of conclusions

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 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 in the conclusion, so we have made the following changes:

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 flight is the most likely explanation for these observations... (snip)

2.1.6 Strong recommendation for aerosol chemical composition measurements on future flights over the Southern Ocean

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

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.

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

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.

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

P. 25525, Lines 11–14: It is stated in the text that during profile 4, \( N_C \) was in the range of \( 6-10 \text{ cm}^{-3} \) with particles of mean diameter \( 6-7 \mu \text{m} \). Based on Figure 5, below cloud
$N_C$ for profile 4 was around 450 cm$^{-3}$ with a mean diameter of 40 µm. 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 overly 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 $\rho_L$ 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 the lines for each profile. Subtracting the offsets specified in the panel legend, the CO values for profiles 3 and 4 below 1000 m were about 55 and 57 ppbv, respectively. You are correct in pointing out that these are not too different from the values above the boundary layer in RF07 (“Flight 2”), which were about 55 ppbv. However, we would argue that the air above the boundary layer in RF07, in spite of having a recent history over the Australian continent, is very clean based on the absence of BC and extremely low NU. We present RF07 as an example of a clearly polluted boundary layer against the background of a pristine airmass.

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

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

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

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

I have reviewed the manuscript “Observations of high droplet number concentrations in Southern Ocean boundary layer clouds” by Chubb et al. The work presents results from a small subset of HIPPO flights and examines the microphysical properties of boundary-layer clouds from a small set of observations made near Tasmania. The work highlights that a wide range of cloud droplet number concentrations were observed during these flights. The authors hypothesize that the large number of drops could be related to either anthropogenic emissions or sea-spray aerosol. Based on their analysis of model back trajectories the authors argue that the most likely cause of the large numbers of particles is the generation of sea-spray aerosol associated with very strong winds. It is important to note that this finding is not based on direct observations, but rather on the elimination of a number of other potential sources of the particles. Overall, the manuscript provides a clear and concise description of the results, and I feel that the manuscript would likely be acceptable for publication in Atmospheric Chemistry and Physics with some minor changes. That said, the study would be much more convincing...
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 have had the potential to make our conclusions much stronger, which we of course completely agree with. This point was also raised by Reviewer #2, who additionally requested that we soften our conclusions as a result. Reviewer #1 also requested a softening of our conclusions based on the ambiguities relating to aerosol composition. We have made two several changes to the manuscript as a result of those requests which should address your comment, including a recommendation for aerosol chemical composition measurements on future flights over the Southern Ocean. Please note that there is a version of the revised manuscript with changes tracked since the original submission accompanying this response.

1. 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.

   **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)**

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

   While Nc values of ... (snip) ... observed by the UHSAS, which probably includes most of the CCN, was produced locally. **While alternative sources for the CCN cannot be completely ruled out without compositional analysis of the aerosol, we showed through ... (snip)**

3. Changes to section 7 to soften final conclusion:

   In this paper, ... (snip) ... 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 flight is the most likely explanation for these observations. ... (snip)

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

   In this paper, ... (snip) ... 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.

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

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 \( N_C \) and \( N_U \) 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.

Page 25505, line 29. “Provide” should be “provided”.

Author Response: Accepted; please see tracked changes document.

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

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:

1. 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 isn’t 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?

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 there was a relatively consistent $\rho_L$ in a layer about 250 m, which is about right based on our Figure 4. When viewed as a time series and smoothed, as in Figure 2 of this document, there is a local maximum near $t = 18865$. The slope of this feature drops sharply after about $t = 18870$, even though the descent rate decreased dramatically at about this time, suggesting that the aircraft is indeed exiting a convective feature through the side (nearly horizontally) rather than through the base. A reasonable estimate for the traversal time of the convective core might be 10 s (1300 m), which is not unreasonable for this type of feature.

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.

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

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

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

Profile 2 shares a number of ... (snip) ... cloudy layer. Below this was a layer about 250 m deep with relatively consistent values of $\rho_L = 0.25 \text{ g m}^{-3}$, which we interpret as the 10-second smoothed $\rho_L$ (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 ... (snip)
Page 25517, line 3. The text states that there is a peak in $N_U$ 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”.

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

References


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

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Abstract

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 southernmost profiles. Sub-micron (0.06 < \(D\) < \(1\) μm) aerosol concentrations for 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 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-
thorogenic 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).

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 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, \(N_C\) 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 \(N_C\) 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.

The HIAPER Pole-to-Pole Observations flights (HIPPO; Wofsy, 2011) were not dedicated cloud physics experiments but have nevertheless 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 range as far south as 67\(^\circ\)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 $59^\circ \text{S}$, to $80–120 \text{ cm}^{-3}$ in a region of homogeneous stratiform cloud in moderate south-westerly flow between $62–67^\circ \text{S}$. Broadly speaking, these values were in line with those from ACE-I and the SOCEX experiments.

Cloud particle effective radius $r_e$ and optical thickness $\tau$ 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 $60^\circ \text{S}$) were $40–67 \text{ cm}^{-3}$, 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 $65^\circ$ to be reliable (Grosvenor and Wood, 2014). In 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 $0.01–1 \mu\text{m}$, 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, $W$ can 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 $N_{\text{SSA}}$ with wind speed (Bates et al., 1998; Covert et al., 1998; Berg et al., 1998). More recently, (Blot et al., 2013) presented observations of $N_{\text{SSA}}$ made in the unpolluted south-eastern Pacific during the VOCALS (Variability of the American Monsoon Systems (VAMOS) Ocean-Cloud-Atmosphere-Land Study) campaign. These data, recorded over a range of 1000 km 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.

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 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 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-
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

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.
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 clouds, 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 legs 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

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 µm (Baumgardner and Korolev [1997]), but we made no specific correction for this other than only using particles with diameters larger than 62.5 µm 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 ($\rho_L$; 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 $\rho_L$ and $N_C$ using the updated sample area.

- **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
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 $\rho L_{\text{King}}$.

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.

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).
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

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 $\mu$ (mean diameter) of $0.2\mu m \pm 30\%$ (i.e. $0.14–0.26 \mu m$) and $\sigma$ (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 $25–80 nm$, 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 $0.2–0.5 \mu m$ 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.
2.5 Calculation of back trajectories

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 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 \( \theta \), 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 \( \theta \) 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 \( \Delta x \) and \( \Delta y \) 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 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.

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 $\Delta x$ and $\Delta y$ 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.

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 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 ($\theta_v$) inversion at cloud top of about 6 °C. The height of the inversion was about 1150 m, and the temperature at cloud top (CTT), 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 \(6 \text{ g kg}^{-1}\). The cloud layer itself was about \(400 \text{ m}\) deep, and in cloud, \(\rho_L\) was near-adiabatic with peak values of around \(0.60 \text{ g m}^{-3}\) at cloud top. This environment appears to be typical for stratocumulus conditions. Horizontal winds were from the northwest and decreased from \(20\) to \(15 \text{ m s}^{-1}\) through the boundary layer, with little directional change at lower levels.

Profile 2 shares a number of features with the classic example of profile 1, but most notably the \(\theta_v\) profile is more complex. The boundary layer top (main \(\theta_v\) increase) was at about \(1130 \text{ m a.s.l.}\) and was coincident with the cloud top, with a temperature of \(2{^\circ}\text{C}\). Above this was a cloud-free intermediate layer of about \(200 \text{ m}\), with \(q_v\) remained similar to the in-cloud values. There was a weaker \(\theta_v\) increase at \(1320 \text{ m a.s.l.}\), which was coincident with a sharp drop in \(q_v\) to nearly zero. At cloud top \(\rho_L\) was as high as \(0.6 \text{ g m}^{-3}\), and was approximately adiabatic in the upper \(300 \text{ m}\) of the cloudy layer. Below this there was a layer about \(250 \text{ m}\) deep with relatively consistent values of \(\rho_L \simeq 0.25 \text{ g m}^{-3}\), which we interpret, the 10-second smoothed \(\rho_L\) (not shown) reached a minimum of \(0.25 \text{ g m}^{-3}\) before increasing briefly to \(0.35 \text{ g m}^{-3}\) 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 \(5 \text{ m s}^{-1}\) 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 \(5 \text{ m s}^{-1}\) 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 \((54.62^\circ \text{S}, 158.85^\circ \text{E})\)- 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 $0.5^\circ$C inversion in profile 3 and about $4^\circ$C in profile 4.

The cloud that was observed in these profiles occurred in the lower levels where the temperature was exclusively above $0^\circ$C. Conditions were considerably more strongly stably stratified than for profiles 1 and 2 ($\theta_v$ 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 $\rho_L$ 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 $\rho_L$ 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 been in the range of 17 to 20 m s$^{-1}$. Gale force winds speed $\text{gale force (≥ 17 m s}^{-1})$ 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 15 of days between 2008 and 2011.
3.2 Microphysics variables

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 $\rho_L$ 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 $N_C$ in profile 1 was relatively uniform throughout the cloud with a mean value of 45 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 $\overline{D_C}$ increased from about 10 $\mu$m near cloud base to about 27 $\mu$m near cloud top. The cloud droplet effective radius ($r_e$; Hansen and Travis 1974), calculated from the PSD was 14.4 $\mu$m. 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 mm h$^{-1}$ were calculated, with drizzle drops of diameter up to 400 $\mu$m observed.

The picture was similar for profile 2, where peak $\rho_L$ values were comparable, but the mean droplet number concentration was higher ($N_C = 77$ cm$^{-3}$) and the diameters smaller ($\overline{D_C} = 23$ $\mu$m and $r_e = 13$ $\mu$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 $\rho_L$ was quite variable, but $N_C$ 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 cm$^{-3}$. $\overline{D_C}$ tended to vary with $\rho_L$, and had a average value of 14 $\mu$m, and $r_e$ near cloud top was about 8.6 $\mu$m. Virtually no drops larger than 100 $\mu$m were imaged by the 2DC.
In profile 4 the HIAPER appears to have flown through patchy or broken cloud, with \( \rho_L \) falling below both the 0.05 and the 0.01 \( \text{gm}^{-3} \) thresholds at several points during the profile. Within the patchy cloud it is difficult to establish a representative \( N_C \) value, because over any given averaging interval there may have been a mixture of clear air and cloud. In a ten-second interval near cloud top, the mean value was \( 144 \text{cm}^{-3} \), but \( \rho_L \geq 0.05 \text{gm}^{-3} \) at the altitude of maximum \( \rho_L \) the mean value was \( 188 \text{cm}^{-3} \). The 1 \( \text{Hz} \) peak values, which are possibly the best estimate of the “adiabatic” cloud droplet concentration (Yum and Hudson, 2004), were up to \( 300 \text{cm}^{-3} \). \( D_C \) varied very little from 10–12 \( \mu \text{m} \), and \( r_e \) near cloud top was 7.0 \( \mu \text{m} \). Very few drizzle drops greater than 100 \( \mu \text{m} \) diameter were observed in this profile.

UHSAS aerosol concentration \( (N_U; \text{fourth panel of Fig. 5}) \) 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 \( N_U \sim 10–20 \text{cm}^{-3} \) 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 \( N_U \) values (not shown) were typically above \( 100 \text{cm}^{-3} \) and values above \( 500 \text{cm}^{-3} \) were observed in two profiles.

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

There was a narrow, well-established accumulation mode (usually associated with cloud processing) centered on 0.13 \( \mu \text{m} \), and the broad SSA mode was centered on 0.18 \( \mu \text{m} \). Profile 2 shows a similar but smaller spike in \( N_U \) at cloud top which we again attribute to droplet breakup. Below-cloud values were on average \( 113 \text{cm}^{-3} \), \( 110 \text{cm}^{-3} \), or about \( 1.5–1.4 \) times \( N_C \), with a broader accumulation mode, with median diameter of 0.174 \( \mu \text{m} \). The
accumulation mode was centered on 0.15μm, and the SSA mode was centered on 0.2μm. 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 shown for this profile. However, relatively clear air between clouds at 700 and 850 m a.s.l. was sampled. In this gap $N_U$ was about 150 cm$^{-3}$, but this might not be representative of below-cloud values.

The below cloud 60-second average value of $N_U$ in the lowest leg of profile 4 was 383 cm$^{-3}$ below cloud base. This is more than double the mean $N_C$ value, but as 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 $N_U \approx 1.5N_C$. The size of these aerosol particles). The accumulation mode was centered on 0.12μm, which is somewhat smaller than for profiles 1 and 2. The SSA mode was centered on 0.15μm, which was similar to that in the accumulation mode for this profile.

For profiles 1 and 2, with median diameter 0.143μm the modal decomposition suggested that the SSA mode contributed about 55% of the total $N_U$. 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 $N_U$ for this profile than the those taken to the north.

### 3.3 Summary of flight data observations

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 $N_C$
and \( N_U \) 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, \( N_C \) in-cloud and \( N_U \) 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.

4.1 Back trajectories

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 \( 40^\circ S \). 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 \( 1.5 \text{ cm s}^{-1} \)) 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. 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 airmass 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 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.
4.2 Analysis of chemistry data with respect to back trajectories

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 also increased with height above 5500 m, 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 ($N_U \approx 30$ cm$^{-3}$) at these levels. Just above cloud top at 1500 m a.s.l., $N_U$ 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 $N_U$ of about 50 cm$^{-3}$ from values of 10–20 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 $N_U$ concentration of about 80 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 fea-
ture is clearly decoupled from the boundary layer, as evidenced by $N_U$ values strictly below $20 \text{ cm}^{-3}$ and CO concentration of $52 \text{ ppbv}$ between $1250$ and $2000 \text{ m a.s.l.}$ Within the boundary layer itself, CO concentrations decrease with height from $54 \text{ ppbv}$ increase towards the surface to a maximum of $57 \text{ ppbv}$ at $167 \text{ m a.s.l.}$ This negative CO 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 case, 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, $N_U$ varied much more in the boundary layer than it did near $4000 \text{ m a.s.l.}$ for a similar variation in CO, so it is difficult to attribute the elevated $N_U$ to long-range transport.

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 $1900 \text{ m a.s.l.}$); lower wind speed (about $10 \text{ m s}^{-1}$); and there was no cloud. The wind direction between $600$ and $1500 \text{ m a.s.l.}$ 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 $\theta$, CO, BC, and $N_U$. Concentrations of CO were about $20-30 \text{ ppmv}$ higher within the plume than in the free troposphere. $N_U$ of up to $2000 \text{ cm}^{-3}$ 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 $55 \text{ ppbv}$, BC of virtually zero, and $N_U$ of $20-40 \text{ cm}^{-3}$.

In HIPPO-3 RF06 (not shown), a return flight from Christchurch to $67^\circ \text{ S}$ 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 $41 \text{ ppbv}$ (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 $N_U$ concentrations were less than $20 \text{ cm}^{-3}$ in the free troposphere but rose to around $100 \text{ cm}^{-3}$ 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 $N_U$ 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 $N_U$ and $N_C$ 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).

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 Australian 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).
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 $N_U$ values in the boundary layer were due to dust, we should expect to find similar $N_U$ 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 $N_U$ 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?

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. 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 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 $N_C$ and $N_U$ 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 ±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 $\rho_L$ from the CDP. Using a parcel ascent model (which conserved $\theta_e$) initialized with conditions near cloud base for profile 1, we calculated the theoretical adiabatic $\rho_L$ 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 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 $\rho_L$—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 cylindrical 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 $N_C$ 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 during the lowest leg met our “probably clear” criterion of $\rho_L < 0.05 \, \text{gm}^{-3}$. 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$^{-3}$ ($\rho_L$ was 0.001–0.005\,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 observations. Indeed, setting the “probably clear” $\rho_L$ threshold as low as 0.002\,gm$^{-3}$ revealed very little sensitivity in $N_C$. We are thus highly confident that the average values of $N_U \approx 383 \, \text{cm}^{-3}$ 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 3-hourly one degree dataset used is sufficient to resolve the synoptic-scale features which are dominant, at least in the free troposphere.

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 $\theta$, 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 where $\theta$ is well-mixed, or in regions where vertical motion due to moist convection may be present, so using modeled vertical velocities was a better choice.

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.
6 Discussion

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 sub-micron particle number concentration $N_U$. Better correlation between the three values was identified at 1500 m a.s.l., where peak $N_U$ values were over 100 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 decoupled from the boundary layer, and that by a layer of clean air. Furthermore, $N_U$ 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 $N_C$ and $N_U$ values in 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 equally 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 $N_U$ 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 \( N_C \) 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 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 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).

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 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 \( N_C \).

This result is of interest in discussions of the cloud structure and radiation bias over 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° 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 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

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 $N_C$ and UHSAS aerosol concentration $N_U$ 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...
high winds in the southernmost regions of the flight is 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


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.
Figure 4. Profiles of thermodynamic variables within and above the boundary layer for RF06. From left: virtual potential temperature ($\theta_v$); specific humidity ($q_v$); CDP liquid water content ($\rho_L$); 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.
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 ($\theta_v$, 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 ($\bar{D}_C$, 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 ($RR$).
Figure 6. **Left:** Particle size distribution distributions (10 averages PSDs) 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 10 averages; note). Note the logarithmic 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 $N_\text{U}$ 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 heights 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 0.01 σ (about 250 m) 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 ($\theta$, note offset of 10 K 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$_x$, and $N_U$ compared to Fig. 9.