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# *Interactive comment on* "Observations of high droplet number concentrations in Southern Ocean boundary layer clouds" *by* T. Chubb et al.

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# Response to comments from Anonymous Reviewer #2 for "Observations of high droplet number concentrations in Southern Ocean boundary layer clouds"

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

> Thank you for taking the time to provide a thoughtful review of our paper. We will address your comments point by point (our emphasized sections of the reviewer comment). There is a version of the revised manuscript with changes tracked since the original submission accompanying this response, and we indicate changes to the manuscript, with deletions indicated by red and additions indicated by blue.

### 1.1 Direct evidence for the conclusion is lacking

Direct evidence for our conclusionâĂŤthat 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 $360\mathrm{C}$. 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:

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

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

> While $N_\mathrm{C}$ 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)*

### 1.2 Missing CO and BC data

Firstly, there are no BC data for elevations below $1200\,\mathrm{m}$ 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 $4200\,\mathrm{m}$ 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 $N_\mathrm{U}$.

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 $3000–4000\,\mathrm{m}$ a.s.l., but slightly elevated CO concentrations at about $4000\,\mathrm{m}$ a.s.l. correspond to increased $N_\mathrm{U}$ of about $50\,\mathrm{cm}^{-3}$ from values of $10–20\,\mathrm{cm}^{-3}$ 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 $4000\,\mathrm{m}$ a.s.l. in profile 4, there was a slight … *(snip)* … to a maximum of $57\,\mathrm{ppbv}$ at $167\,\mathrm{m}$ 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 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, $N_\mathrm{U}$ varied much more in the boundary layer than it did near $4000\,\mathrm{m}$ a.s.l. for a similar variation in CO, so it is difficult to attribute the elevated $N_\mathrm{U}$ to long-range transport.

### 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 $D > 0.5$ µm. However, with the logarithmically spaced bins of the UHSAS (0.06 to $1$ µm) 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<$0.5$ µm. The actual values used were $0.391$, $0.359$, and $0.235$ µm 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:

1. New section (section 2.4 in revised manuscript) summarizing the methods used. Please see accompanying tracked changes document.
2. Figure 6 of revised manuscript now contains four panels: One for the combined CDP/2DC PSDs (as previously), and one panel for the UHSAS PSD for each profile.
3. We changed the averaging interval for the lowest leg to $60$ s to reduce noise. This had the effect of slightly changing some of the values quoted in section 3.2 for $N_\mathrm{U}$ and so on.
4. In addition, we aggregated the UHSAS bins into groups of 3 so that they remained logarithmically spaced. This did not affect the results of the analysis, but it makes the graphs somewhat easier to interpret.
5. Changes to discussion in section 3.2 to describe the new results. Please see accompanying tracked changes document.
6. Sentence added to section 6.1, last paragraph:

> While $N_\mathrm{C}$ values of . . . *(snip)* . . . was produced locally. We While alternative sources for the CCN cannot be completely ruled out without compositional analysis of the aerosol, we showed through modal decomposition of the UHSAS PSD for profile 4 that the mode representing primary marine aerosol, which is dominated by SSA, to be the main contributor to the aerosol population. We thus consider sea spray aerosol . . . *(snip)*

### 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. Bibliography item added for Kassianov et al. (2015).

### 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 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 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 flightis the most likely explanation for these observations.. . . *(snip)*

### 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 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.

> 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, $N_\mathrm{C}$ was in the range of 6–$10\,\mathrm{cm^{-3}}$ with particles of mean diameter 6–$7\,\mathrm{\mu m}$. Based on Figure 5, below cloud $N_\mathrm{C}$ for profile 4 was around $450\,\mathrm{cm^{-3}}$ with a mean diameter of $40\,\mathrm{\mu m}$. Am I reading the Figure incorrectly?

> Thank you for this comment. It appears some clarification is needed. Firstly, figures 4, 5, 9, and 10 have a profile-dependent offset (specified in the legend of each panel) so that the lines do not overlie one another. For the panel you refer to (Fig. 5 panel 2), there is an offset of $100\,\mathrm{cm^{-3}}$ 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 $\rho_\mathrm{L}$ 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 $1200\,\mathrm{m}$ (at least according to Figure 9). And CO is not significantly lower (65 and $70\,\mathrm{ppbv}$ for ~~flights~~ profiles 3 and 4 at altitudes $< 1000\,\mathrm{m}$) than the air masses tagged as anthropogenic during Flight 2 ($\sim 65\,\mathrm{ppbv}$ at 4000–$5000\,\mathrm{m}$).

> 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 $1000\,\mathrm{m}$ were about 55 and $57\,\mathrm{ppbv}$, 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 $55\,\mathrm{ppbv}$. 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–$4000\,\mathrm{m\,a.s.l.}$ in profiles 3 and 4 of RF06, but there is a layer between about 1250 and $2000\,\mathrm{m\,a.s.l}$ 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 $N_\mathrm{U}$ at 3000–$4000\,\mathrm{m\,a.s.l.}$ for a much larger increase in CO, anthropogenic sources could not account for the observed increase in $N_\mathrm{U}$ 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 $4000\,\mathrm{m\,a.s.l.}$ in profile 4, there was … *(snip)* … evidenced by $N_\mathrm{U}$ values strictly below $20\,\mathrm{cm^{-3}}$ and CO concentration

> of 52 ppbv between 1250 and 2000 m a.s.l. Within the boundary layer itself, CO concentrations decrease with height from 54 ppbv increase towards the surface to a maximum of 57 ppbvat 167 m a.s.l. to 50 ppbv at 1250 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 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, $N_\mathrm{U}$ 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 $N_\mathrm{U}$ to long-range transport.

2. Sentence added to section 4.3, paragraph 1:

> It is useful to provide some context ... *(snip)* ... CO concentrations ($R = 0.87$ and $R = 0.95$ respectively). On the other hand, the free troposphere was extremely clean based on the CO values of about 55 ppbv, BC of virtually zero, and $N_\mathrm{U}$ of 20–40 cm$^{-3}$.

3. Changes to section 6.1, paragraph 3:

> 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, $N_\mathrm{U}$ 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 ... *(snip)* ... 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 ... *(snip)*

## References

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