

We thank the referees for their useful comments and suggestions which have helped us to improve the manuscript. Comments from reviewer 1 are in red, reviewer 2 in blue. Our responses are in black and bullet-pointed. The main changes to the manuscript are summarised as follows:

Reviewers comments

Reviewer 1, Reviewer 2

Main Changes (see discussion below):

1. Title updated
 2. Figure 5 removed (*original figure numbers have been used for reference in this document*).
 3. Using “Case 1-7” instead of flight numbers.
 4. Size distributions (Fig. 4) updated to show individual probe data.
 5. Text in Sections 2.3, 3.2, 3.3 cut significantly, Section 4 text made more concise
 6. Classification scheme included as Table S1.
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General Comments

“Exploring the variability of aerosol particle composition in the Arctic: a study from the springtime ACCACIA campaign” by Young et al. focus on the chemical composition observed during six flights conducted in spring 2013.

Reading the manuscript it is clear that the authors worked with a limited set of data. Nevertheless, any airborne observations of aerosol properties should be shared with the community, as we still lack some fundamental understanding on this topic. As pointed out, models are not yet able to fully reproduce details in the variation of the Arctic aerosol properties. Unfortunately, the authors chose to focus on low-level data, and in my opinion the most interesting conclusion was based on comparing two levels in the vertical.

- Low-altitude data was the focus of this study as particles collected were thought to be the primary contributions of INPs and CCN to the clouds in the region. It was also the aim to compare samples between flights from similar locations with respect to cloud. Below cloud filters were available for each of the 6 flights shown in this study (Fig. 1), whilst above cloud filters were only available in flights B764, B765 and B768. The only feasible below/above cloud comparison (B764) was analysed and included in this study.

The conclusion about the origin of the air in FL764 and the ubiquitous presence of minerals in all flights, are sufficient to warrant the publication of these data.

Before any publication I have a few comments that might improve the manuscript. To begin, I think the title must be revised. The “variability of aerosol particle composition”

implies some statistical representation of a larger data set than is actually presented here. In total I calculated about 1.5 hours of sampling time and less than 10 cubic meters of air. Besides some mean values, there are few statistical measures on the variability presented in the manuscript. I propose a more direct approach, where the title reads something like: Observed size dependent chemical compositions of aerosols in the sub-Arctic during six measurement flights during the ACCACIA campaign. Far from perfect suggestion, but I hope the point is made.

- Title has been updated to read as follows: “Size-segregated compositional analysis of aerosol particles collected in the European Arctic during the ACCACIA campaign.”

On the topic of variability, may I suggest the reference by Tunved et al. 2013 (ACP) for a climatology of physical properties observed at the Zeppelin station to include in your introduction. Also pertinent to this study are references from the ASTAR 2007 campaign, i.e. Hara et al.

- Citations to these articles (Hara et al. 2003 (ASTAR 2000), Tunved et al. 2013) have been included in the introduction as suggested.

This study analyzed the size distributions and element compositions of samples collected by aircraft during the springtime segment of the Aerosol Cloud Coupling and Climate Interactions in the Arctic campaign (ACCACIA). The study is important because it leads toward a better understanding of the composition of particles directly below and above clouds in the Arctic. The types of particles studied here could have served as cloud condensation nuclei or ice nucleating particles in the vicinity of Svalbard, Norway, and therefore, may lead toward a better understanding of the effect of clouds on the Arctic climate. The paper cites existing literature extensively, which normally is most helpful. However, here the literature could have been presented more concisely, and thus, for this reason alone the paper could have been shortened.

- Portions of text throughout manuscript have been shortened, see below for details.

The paper presents a substantial amount of information that was reported previously in the literature, and thus, need not be reported here. For example, Section 2.3 discusses extensively the process of analyzing particle populations with energy-dispersive x-ray spectroscopy associated with SEM. Particle population analysis by SEM has been well established for decades, including normalizing the composition by weight percent, which is the only plausible way to report composition if particle standards are impractical.

- Information pertaining to the analysis set-up in Section 2.3 has been simplified in the manuscript.

In another example where the lack of a concise explanation causes some confusion, the authors indicate in one place that carbon and oxygen were measured, but later indicate that carbon and oxygen were not quantified. More about this below under Specific Comments.

- Addressed below.

In reporting of results and the follow-up discussion, some of the interpretations are questionable or lack the appropriate emphasis. For example, I do not see where one

can draw convincing conclusions from Figure 5 about differences in the complement of particles classes between smaller particles (<0.5 μm) and larger particles (>0.5 μm). As the authors admit, the compositional variability is great among the samples from the different aircraft flights. The between-sample variability in Fig. 5 overwhelms any size-dependent trend. Regarding appropriate interpretive emphases, the authors make a number of definitive claims of the data showing a “clear” effect where the data are more nuanced. I recommend that the authors be more careful in interpreting their data. Specific examples are presented below.

- This is a fair point, need to be careful with language. This required some general rewording throughout the manuscript, however a discussion usefulness of Figure 5 is mentioned below. The conclusions of the paper would be unchanged with adequate evidence if Fig. 5 was removed, therefore it has been.

Overall however, I find the paper informative because the authors do manage to return at the end of the paper to their main objective. That is, they discuss rather well (with appropriate caveats) how the size-segregated compositional analysis here relates to CCN, INPs and cloud microphysics.

1 Introduction

29407 Section 1.1 line 22

Nuclepore is a trade name. It would be good to indicate the source (i.e, manufacturer) of the filters.

- Filters used were Whatman Nuclepore track-etch membranes. The manuscript has been updated to include this information in the Methodology section (Sect. 2.2, line 134)

2 Methodology

29408 Section 2.1 paragraph 2

There is extensive particle-size overlap among the particle size distribution techniques used: PCASP, CAS-DPOL, and CDP. This suggests that for the overlap regions these probe techniques could have been compared. However, this was not done.

- Figure 4 has been updated in the manuscript to include the probe data individually, therefore allowing the regions of overlap to be viewed and compared.

2.2. Filter collection

Line 20-25

My understanding is that sub-isokinetic sampling is not the trick for removing large particles or droplets. It is the virtual impactor at the bend (where the inlet direct the sample flow through the fuselage) that gives this positive effect.

- Agreed, sub-isokinetic sampling in fact has an enhancement effect on large particles. It is the bend in the mechanism which provides inertial separation of cloud drops and rain into a bypass tube. This has been made clearer in the manuscript.

This design in itself generates a blunt cut-off in the sampling of particles, which is not addressed in this manuscript. When sampling sub-isokinetic with a forward facing probe,

this introduces a size dependent sampling efficiency which favors larger particles. How much, this effect influences depends on the ratio between the sampling flow and the volume swept through by the opening area of the inlet as the plane moves forward. How large is this ratio, and how does this potentially influence the size dependent sampling for the filter substrates?

- It is not known if the collection efficiency of the sampling mechanism has been quantified but, as stated by Formenti et al. 2008, a precise calculation would require the characterisation of the inlet in a wind tunnel. Despite this, Andreae 2000 compared ground-based and aircraft samples during the ACE-2 campaign and found the sampling efficiency of the aircraft inlet (MRF C-130 version) to be about 35% for coarse-mode aerosol. The same comparison showed good agreement for fine-mode aerosol particles. Formenti et al. 2003 use this information to set an approximate 50% cut-off diameter of 3micron for sea-salt particles collected during the SAFARI campaign (also using the MRF C-130 aircraft). Samples collected here were done so using the same mechanism investigated by these two studies, therefore this information should similarly apply in this study (with some deviation allowed for differing sampling conditions etc.). This would suggest that the coarse-mode is under-represented in these ACCACIA filter samples, whilst the collection efficiency of fine-mode aerosol is much better. This agrees with the conclusions discussed in Section 4.1, where it is speculated that it is an artefact of the SEM analysis which is causing the significant disagreement with the probe data in the accumulation mode. This conclusion was reached by Chou et al. 2008, who found good agreement between filter (analysed with TEM) and probe data on scales of a few tenths of a micron up to 0.5micron. Agreement deviated at sizes >0.5micron, where they observed significant large particle enhancement due to the sub-isokinetic sampling. Such enhancement was not observed in this study, and probe data always produced a greater concentration than the filters in the coarse mode. Given this information, the losses of coarse-mode particles appear to dominate over the enhancement effect introduced from sub-isokinetic sampling, and these efficiency issues have been emphasised further in the manuscript to make this clearer.

Even with a tapered tip (as I believe is used on the FAAM platform) there is a potential risk of drops shattering on the probe tip, or more so, inside the probe if the probe is not aligned with the streamlines of the surrounding air. That is, the probe is of axis during low speeds with high aircraft attack angles or in turns etc. These problems could cause spurious effects in the data during cloud traverses. Where any such observed, or was this problem considered in the analysis somehow?

- The filters used were exposed only during horizontal flight legs where potential angle-of-attack issues were not encountered. The aircraft speed was $\sim 100\text{ms}^{-1}$ in all sampling cases, therefore this issue was not evident in the data.

2.3 Environmental scanning electron microscopy

The paper describes the use of an ESEM with EDS for analyzing the various polycarbonate filter samples. The ESEM is used in the high-vacuum mode, so it functions here as a conventional SEM with EDS. Since the instrument is not used as an ESEM, i.e., at low vacuum with a water vapor atmosphere, it should be indicated in this section that the instrument is essentially a conventional SEM to avoid confusing readers unfamiliar with an ESEM. All later mentions of ESEM in the paper should be changed to SEM.

- This is a valid point and all references to ESEM were changed to SEM in the manuscript.

29410 lines 19-21

The paper explains at length how particles are analyzed by SEM, and as mentioned above, much of this is already in the literature. There is, however, one significant gap: the software procedure used to perform the particle population analysis in the SEM should be better explained. It is stated that the electron beam is controlled by the EDS system to provide automated analysis of the sample, i.e., the analysis of the particle population on a filter. This implies that the X-ray signal is used to detect the presence of a particle and determine where in the particle spectra are to be taken. I know of no commercial software, EDAX Genesis included, that does this. Rather, software uses the backscatter electron or secondary electron signal to detect a particle. Typically, software decides where to point the beam within the particle for X-ray collection by assessing the shape of the particle and then judging where the center of the particle is. The paper fails to indicate whether the backscatter or secondary electron signal was used and whether the beam was held stationary or rastered within the particle. These issues should be made explicit in the Methodology.

- The backscatter electron signal was used. As described by the reviewer, the software identifies the particle via greyscale thresholds and locates the centre. The analysis was done in core mode, using 70% of presented area, and the beam was rastered over that area to compute each spectra. This is what we meant by an averaged spectra, however we agree that this was not clear. The text in Section 2.3 has been updated to reflect this, in lines 170-178.

29411 lines 6-7

It is indicated that carbon and oxygen are included in the analysis. However, these elements would be highly problematic because X-rays from the polycarbonate substrate would certainly penetrate the particles. The authors do mention this problem, and two paragraphs later they state that carbon and oxygen were not included. It would be clearer and more concise if the authors simply stated that carbon and oxygen were not quantified. Later the authors indicate that the presence of carbon and oxygen in an X-ray spectrum containing no other elements provided evidence that the particles were carbonaceous. Here, we are to assume that carbon and oxygen in a spectrum were used qualitatively, not quantitatively.

- We have updated Section 2.3 to reflect this request. We have stated that elements C to Zn are scanned, but C and O are presented qualitatively due to the polycarbonate filter issue. This is difficult to address as C and O measurements were made and approximate thresholds were used to identify when there were no other elements detected (See Table S1 for classification criteria), therefore the data is used. We have made this clearer to the reader in Sections 2.3 and 2.4.1.

Page 29411 Line 11

What does 4 pixels correspond to in μm ?

- Minimum sizes are listed in Table 3. A reference to reflect this has been included on line 180.

29411 paragraph 2 (lines 12-20)

This paragraph focuses on an analysis of a blank. More information should be provided about the nature of the blanks. For example, how many blanks were used? Were they field blanks in that they were somehow exposed in the aircraft, or were they simply lab blanks taken out of a box prior to the SEM analysis? Field blanks are much preferred in this type of study.

- One field blank filter pair from flight B762 was analysed. These were treated similarly to the exposed filters (i.e. taken aboard the aircraft) without any exposure to the air flow. The manuscript has been updated to address this (lines 191-194).

Page 29412

Last line “The number: : :” what is the fraction?

- A bug was identified in the code which had been removing more particles than first thought. Approx 3-5% more particles were removed than should have been. All of these were classified as carbonaceous once they were included in the data. This has been rectified and Fig 6 and 8 have been re-done and updated in the manuscript. The figures have changed very little but in the interest of consistency they have been re-done. The fraction of particles removed is ~4-5%, though this is variable between different filters and different scans. This information has been updated in the manuscript.

2.4 Classifications

29413 Section 2.4 and Table 4

The omission of quantitative carbon and oxygen measurement and the identification of carbonaceous particles based on the x-ray spectrum exhibiting qualitatively only carbon and oxygen has a significant drawback. Certainly, the authors were correct to avoid quantifying these elements. However, mixed-phase aged particles of carbon (organic or soot) plus mineral dust could not be classified, not to mention carbonate minerals alone. The authors should discuss this drawback and possible solutions.

- More information about the limitations of the analysis (with respect to C and O measurements) has been included in Section 2.4.1 and 4.3. Information on coatings that was originally in Section 4.3 has been moved to the introduction to emphasise its importance in the study.

The classification scheme presented in Table 4 only indicates qualitatively how particles were classified. The actual scheme had consist of numeric boundaries, i.e., weight percent boundaries. What do the following mean: “significant Na and Cl”, “major fractions of Na, Cl, S”, “mixtures primarily containing Si and Al”, etc? It is important to report the quantitative scheme so other researchers can utilize the same scheme if desired.

- Classification scheme has been written up and included instead as a supplementary table (Table S1) due to its length.

2.4.5 Biomass tracers

Page 29417 Line 25 The word “present”, is this meant to be “originated”? If not I don't get the logic of the paragraph. Surely such aerosol may be present in the Arctic.

- Such particles may be present in the Arctic, but yes it is unlikely that they originated

there. Their presence could therefore be used as an indicator for long-range transport. The text in Section 2.4.5 has been updated to make this clearer.

2.4.6 Other

Why is it implied that particles are well mixed if they are classified as other, i.e., not within the classification scheme?

- Particles unclassified by the scheme are thought to be mixes of the classifications as they had not met the specific criteria for any category. Typically, many different elements were detected in quantities not substantial enough to allow for distinct classification. As many elements were measured, these were assumed to be mixes. However, as these were typically Si and Al deficient, they were not thought to be dusts. As agreed below, these should not be referred to as “well-mixed” and this has been updated in the manuscript.

Also, the following statement is problematic: “: : the automated scan will not catalogue the spatial dependencies and instead computes a mean spectrum for presented particle surface area.” It is important to use precise wording. Scanning in automated SEM allows for the detection of a particle from the backscattered electron or secondary electron signal. Typically, the acquired spectrum is not a mean but rather the result of the electron beam held stationary on the particle for a duration after the particle center is determined. To clarify, the authors should state that automated SEM for particle analysis does not acquire an element spatial map of each particle.

- Agreed, the wording of Section 2.4.6 has been updated to reflect the reviewers comments.

3.1 HYSPLIT back trajectories

Page 29418 Line 22 Strike “monotonically” as this is not always true and especially not for all trajectories.

- “Monotonically” has been removed from line 360.

3.2 Aerosol size and morphology

Figure 4. This figure is somewhat central to the manuscript as it is used both to corroborate different measurement technics as well as in the interpretation of the data. I understand that the probe data are means from each flights for the corresponding period of the filter samples. However, it is not clear if these are arithmetic or geometric means.

- All averages and values quoted as “averaged values” are arithmetic means of the data. These have been updated in manuscript.

Also, there are no indications of variability (some call it uncertainty) for each size bin or at least size range.

- Figure 4 has been updated in the manuscript to include the data from each individual probe with 1 standard deviation displayed. Similarly, an average of the SEM data has been overlaid on top of the scatter (x) points to make comparison easier. Only upwards error bars are included for clarity. Error bars have not been

included on the SEM data as the figure would become incoherent, however the spread is still indicated by the scatter (x) points.

The ESEM data is unclear to me. Can a similar line be produced as for the probes? What does each marker represent, fixed size range or number of particles? Presumably, these must represent some sort of histogram if they are to be $dN/d\log D_p$ values.

- The updated version of Figure 4 displays the $dN/d\log D$ data from the filters as an averaged line in addition to the scatter (x) points. The reviewer is correct in stating that each SEM data point represents a histogram of number concentration normalised by bin width ($dN/d\log D_p$). The data was originally presented this way to indicate the resolution of the SEM and the large number of particles detected in each case. This is preserved in the new version of the figure.

I think I got that both filters are used for each data point (if I'm wrong correct me). My main request is that the authors include meteorology and ambient conditions to the manuscript. It is not reported and could fit very well into a table with average T and RH for each sampling period for instance. The result of this could enter the discussion of figure 4. If the RH is high (i.e. >80%) this will significantly change the ambient vs dry aerosol apparent sizes. In some cases, the probe data could be shifted as much as a factor of 2, or more (towards smaller particles) comparing ambient and dry conditions.

- Mean temperature and RH data are now included in Tables 2 and 4. All RH values were found to be high (>90%). Temperature data does not give much to the interpretation of the data but it is helpful to contextualise the exposures. Calculated RH value for B762 (case 3) is not trusted. This may suggest condensation on the probe surface interfering with the measurements. Agreement in Figure 4 between the SEM and probe data is worst for B762 (case 3) and B764 (case 4), and these cases resulted in the greatest RH measurements. Derived RH values for B761 (case 2) and B765 (case 5) are similar even though some in-cloud sampling was definitely noted in B761 (case 2). Probe data from these cases appear to have reasonable, and arguably similar, agreement with the SEM data. Lastly, the lowest RH values were deduced in B760 (case 1) and B768 (Case 6) and, keeping with the trend, these cases provide the best agreement with the SEM data. It must be noted that the derived RH values are not dissimilar. However, these data do appear to help interpret the data, and have therefore been included in Tables 2 and 4 in the revised manuscript.

Later this figure is used in connection of "comparable" in section 5, it is important to note over what size ranges this is true and under what conditions (after the difference between ambient and dry conditions are considered).

- The text in Section 5 has been updated to refer to the comparable size ranges (~0.5micron to 10micron) and address the relevance of the derived RH values.

29420 lines 9-15

I don't think it is correct to say that the agreement between the SEM data and the probe data is clearly dependent on whether or not the cloud was sampled. There appears to be some dependence, but I would say that the out-of-cloud B765 case exhibits less agreement between the SEM and probe data than for the B761 case which had exposure to the cloud. Here, the authors should be more careful in their cause-and-effect assertion.

- This is a good point and was also raised by Reviewer 1. This has been addressed by both the updated figure, showing the variability of the probe data, and the included temperature and relative humidity data in Tables 2 and 4. The RH data helps to explain the agreement or lack thereof between the probe and SEM data, as described above. It also points out that B761 (case 2) and B765 (case 5) are not dissimilar in terms of ambient RH, so offering an explanation of the poorer agreement displayed.

What is more interesting and obvious is the lack of agreement between the SEM and probe data in most cases (the exception perhaps is B768) for particles < 1 μm . Presumably, the PCASP can assess sizes down to 0.1 μm while the SEM analysis can assess sizes down to 0.13 μm (Table 3). We are left hanging on this issue until the Discussion section. It would have been helpful if the authors acknowledged the obvious here in Results and said something like: “more about this in the Discussion.”

- The inlet issues are suggested here as a possible explanation of the disagreement, and a link to the Discussion section has been included in line 404.

3.3 Aerosol composition

29420 Section 3.3

I am not sure what is meant by an “element index” in the first paragraph. If one simply takes the composition data normalized to weight percent in the analysis software, then the effect from the filter is not a problem.

- We are unsure of the question, so will explain the method: The analysis software produces an elemental weight percent for each element (C to Zn). C and O were included in the spectral fit to avoid spurious errors in the fitting software, therefore a measure of C and O is included in the data for each particle. As stated, this measure of C and O is not used quantitatively in main classification groups (it is used approximately in carbonaceous and biogenic, see Table S1), as the filter substrate is influencing it. To do compositional analysis on each particle, the measured weight percent of each element (Na to Zn) is normalised by either the summed total (Na to Zn) measured or a specific ratio is calculated to create a new fraction, or index, for each element. This index is what was referred to distinguish from the raw weight percentages. However, this text has been removed in the manuscript due to the inclusion of the quantified classification criteria in Table S1.

29421 and Figures 5-8

The classification data presented in Figure 5 are far less informative than data presented in Figs. 6 and 8. The only reasonable conclusion drawn from Fig. 5 is that distribution of particle classes between flights is highly variable. I do not see how the mixed chlorides and metallic classes are independent of size. I cannot agree that sulphates, carbonaceous, and biomass tracers are strongly detected in the smaller particles. There is too much between-flight variation. I would caution the authors against reading too much into Fig. 5. In fact, the authors may not want to use Fig. 5 at all.

- Fig. 5 was included to show variability between flights and indicate the number of particles detected in each size range. As pointed out, there is a lot of inter-flight variability. This variability can be seen from Fig. 6, and there are few references to Fig. 5 in the text. Therefore, Fig. 5 has been removed and the total number of

particles scanned in each case is now indicated in Fig. 6. The conclusions of the paper and evidence for them are unaffected by the removal of Fig. 5.

Figure 6 shows a clearer distinction between particles sizes based on particle classes. However, this appears to be due to the authors selecting particles sizes where there was agreement between the SEM size data and the probe data.

- In Figure 6, only sizes which showed relatively good agreement with the wing-mounted probes (0.5micron to 10micron) were included as data obtained out with this range may not be representative of the aerosol population sampled. The lack of agreement is most likely due to the efficiency issues discussed in Section 4.1. Few particles >10micron were measured in most cases; however, case B761 did have a greater number of these. In addition, it has been noted in previous studies (Kandler et al 2011) that signal-to-noise issues arise for particles <0.5micron, and the SEM interaction volume of small particles is likely to be larger than the particle itself. This data was therefore averaged (~1st size bin of Figure 6, and panel A of Fig 8) to give some indication of the composition of the small particles. This was not taken further (e.g. size segregation) as we were not confident if this data was representative given these SNR issues and the inlet/sampling issues discussed in Sections 2.2 and 4.1.

Figure 7 shows only a few “clear” distinctions: in particular, the elevated K/Al, Ca/Al, and Fe/Si levels for the B768 case. Again, I would caution the authors against making strong statements such as there being a “clear” peak in the Si/Al data for the B764 case. Yes, it is the highest mean, but the B765 mean is very close.

- Agreed, the text in Sections 3.3 and 4.2 have been updated to reflect this.

Figure 8 is perhaps the most relevant of all in this paper because of the positional importance of bands CCN and INP (below vs. above) with respect to the cloud.

4 Discussion

4.1.2 Case B768

29425 Case B768

I do not see how the authors can make the claim that Fig. 4 shows a much higher particle loading for the B768 case. The probe data B768 data are about comparable to the B765 data and perhaps less overall than the B761 data.

- This is a very good point. The size distribution of B768 (case 6) is not as dissimilar to B761 (case 2) and B765 (case 5) as the absolute number concentrations would suggest. The total number of particles for each case is quoted in new version of Fig. 6. Though the total numbers collected for this filter pair are not dissimilar to B761 (case 2), the significantly shorter sample time (4mins vs 30mins) infers the greater particle loading. The manuscript has been updated to reflect these changes.

4.3 Internally-mixed aerosol particles

This section is too verbose and contain much background information which is more pertinent into the introduction. Cut what is not essential to the findings of this study. In my opinion there is information about minerals, but nothing about their IN potential,

which would potentially reduce this section a lot.

- Significant portion of Section 4.3 discussing mixed mineral dusts has been removed and inserted in the Introduction.

29428 Section 4.3

I do not see how one can assess with confidence the degree of internal mixing from the variability in the mean composition fractions in Fig. S1. Internal mixing suggests that particles have spatially-separated phases such as for the particle in Fig. 2. (I would disagree that this is a “well-mixed” particle!)

- Good point again, mixed yes but not well-mixed! The term “internal-mixing” has perhaps been used more loosely than it should, therefore the majority of these references have been changed to “mixed” in the manuscript.

In this paper, it seems that internal mixing is determined by the number of elements detected in the particles. As many minerals are compositionally complex, a mineral particle may be compositionally complex but with phases that are not spatially separated, and thus, the particle cannot be considered internally mixed.

- We can see the confusion, however those in the “other” category have passed through the variable criteria for minerals (Table S1), not met these, and so are unclassified. We agree minerals are compositionally complex and may contain numerous elements, yet those left in the “other” category are mostly Si and Al deficient, suggesting they are not dusts and are mixes of something else. We agree that the internally mixed hypothesis should be treated better: the text in Sections 2.4.6 and 4.3 has been updated to reflect this. The particles in the “other” category should be referred to as “mixed”, not necessarily “internally-mixed”. There is no way from this data to concretely determine whether these particles are mixed in the atmosphere or at the source; however, it could be suggested that those detected in the above-cloud case (case 7) had undergone mixing over transport due to their measurement location.

5 Conclusions

29431 lines 1-5

As stated above, conclusions drawn from Fig. 5 are not convincing. I do not see where one can say that carbonaceous particles and sulphates are prevalent in the smaller particles – more present, yes, but not prevalent.

- The text in Section 5 has been updated to refer to these categories as “mostly observed” in the sub-micron limit. As Figure 5 has been removed, the figure reference has been updated to refer to Fig. 6 instead.

Page 29431 Line 7-8

Ok until “;”, after that can be striked).

- Manuscript has been updated to reflect this change.

Page 29432 Line 2

To make my point, meteorology is used as an argument, but not reported in the manuscript. Please, include some info on T and RH at least.

- Mean temperature and RH values have been calculated and included in Tables 2 and 4 to address this.

In general it would be nice if dates for flights were used and flight numbers were referred to in the table and not vice versa. It makes much more sense to other people not specifically involved in the campaign.

- Table 1 updated to reference each below-cloud case as 1-6 rather than use flight numbers. The above cloud case discussed in Sect. 3.4 has been updated to case 7. This should make the number of samples analysed more obvious to the reader. References to each filter pair sampled throughout the text have been updated to reflect these changes.

Given the relatively short sampling times, the trajectory analysis could be greatly simplified by fewer trajectories over the actual sampling time. The 30s interval brings no additional information and only clutters the graphs. Trajectory at start and at end would probably be more than sufficient for the analysis and conclusions.

- The small temporal interval does clutter the graphs somewhat, making B764 (case 4) and B765 (case 5) especially quite difficult to see. A new version of the figure with the reviewers suggestion, i.e. trajectory at start and end of exposure, has been made and included in the manuscript. Some of the variability in B762 (case 3) and B768 (case 6) is lost, but this isn't important to the conclusions of the paper.

It took me some time to understand the samples available. First I thought it was six, then an extra showed up. It is also not clear how many particles were analyzed per filter. Can these things be made clearer in the text/tables perhaps?

- The text in Sect. 1.1 and Table 1 has been updated to make the number of cases considered more obvious to the reader (lines 97-100). Particle numbers are indicated in Table 3 (total) and Fig. 6 (per case). References have been included in Section 2.3 (line 189) and text updated to make this more obvious.
-