

Interactive
Comment

Interactive comment on “In-situ measurements of tropical cloud properties in the West African monsoon: upper tropospheric ice clouds, mesoscale convective system outflow, and subvisual cirrus” by W. Frey et al.

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The authors would like to thank Anonymous Referee #3 for her/his careful reading and the resulting helpful comments. The issues raised by the referee are discussed below and most of these have lead to changes in the revised version of the manuscript. (In this reply we quote additional references which are listed at the end.) Since the other reviewers raised similar concerns, the particular issues related to shattering and the performance of the cloud particle probes are discussed in detail in a separate reply titled “On the issues of instrument performance and shattering artefacts for the FSSP

C3235

Full Screen / Esc

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Discussion Paper



and CIP”. We suggested to the editor to include this document in a properly modified form as online-supporting material to the paper, if accepted.

(A.) Major comments:

Comment: Outflow is not well defined, nor the nuances involved in the concept appreciated. Outflow from thunderstorms involves a great deal of surface air, but also entrainment from mid and higher levels. All this air is mixed into the air exiting a thunderstorm at the upper levels. Does outflow by some convention only imply air from the surface? How do the authors here identify outflow? What exactly are the tracers that will be used, and what will be concluded from them? We need to know how the authors plan to identify outflow. Can NO/NO_y produced by lightning be used to identify outflow? If so outflow from what level? Yes it identifies air from lightning, but is it indicative of surface air, or just air in the cloud? How does air in the anvil of a storm differ from the outflow? Over what altitude range may we expect outflow? How representative of outflow are the measurements presented here? In the contrasting cloud samples from non outflow regions where is the air from?

Reply 1: It is clear, that in an outflow scenario not ALL possible origins of the outflowing air (like from the boundary layer, or from the lightning zones in the cloud centre) can be discerned by means of tracer measurements. Also, when flying in the vicinity of a large cloud body one may sometimes not recognise an outflow, because the sampled air does not contain any measurable “outflow fingerprints”, or these were diluted beyond “recognition”. Due to different chemical compositions of air masses in the boundary layer and in the UT/TTL region air transported by convection may however carry specific signatures. For example, CO₂ can be used as tracer over continental regions because plants remove CO₂ from the air in the boundary layer. When transported upwards through the convective updraughts air with reduced CO₂ concentrations (i.e. a deviation with respect to the average value) probed in the upper troposphere is an indicator for convective influence from a ground “source”. A quantification of such effects

C3236

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



can be found in the model study by Fierli et al., 2011.

NO mixing ratios also may be enhanced due to uplift of soil emissions. For example, Stewart et al. (2008) found mixing ratios of 1 nmol/mol for soil emissions. However, lightning is the dominating source of NO_x in the upper troposphere (Schumann and Huntrieser, 2007; Höller et al., 2009; Huntrieser et al., 2009) and lightning is (mostly) present in the deep convective parts of MCSs. Thus, cloudy or cloud free air encountered in the vicinity of MCS with enhanced NO and NO_y must have passed through the “lightning activity region” of the cloud cores. Otherwise, it must have originated from the ground. In any case this air has spent some of its time in the convective regions. Therefore, NO, NO_y , and CO_2 , all measured aboard the M55 Geophysica, are the main tracer species used for the identification of outflow air masses in this study.

Another consideration is of relevance for identification of the “outflow zone” inside an anvil, for example. Cloud particles may sediment out of the outflow air stream, extending the anvil downwards to regions without outflow current. Furthermore, as reported by e.g. Garrett et al. 2006, pileus clouds may form directly above the anvil. Thus, clouds may be present just above or below the outflow without belonging to the direct convective outflow. In both cases tracers do help for differentiation. Therefore, our definition is kept rather conservative by only looking at the tracer distributions. This consideration is now added to the revised version of the manuscript.

Finally, the identification of outflow zones from large cloud systems by means of particular trace gases is a well established concept as can be seen from many publications, so for example in Huntrieser et al., 1998, 2007, and 2009.

Comment: The authors are too ready to identify any increase in particle concentration as a new particle formation (NPF) event. All measurements are seen as evidence supporting this point of view, while competing possibilities for the source of these high particle concentration measurements, are not discussed seriously. Particle concentrations below the NPF events are missing with no

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explanations. Surface sources of particles from biomass burning, or other activities, are not discussed, nor measured. No source of fresh SO₂, assumed required for the NPF events, is identified. The measurements of high particle concentrations should be included, and pointed out. The suggestion that they may be indicative of new particle formation can be made, but with the caveat that other possibilities cannot be ruled out by the analysis presented in this paper. Thus further work will be required to establish the source of these particles, and leave it at that. Much better than jumping to unproven conclusions.

Reply 2 on NPF issue: Here, we used a concept introduced (for tropical high altitude environments) by Lee et al., 2003, and Lee et al., 2004, for NPF in clear air and in clouds. Under background conditions the N_6 and N_{15} are almost equal either because no freshly nucleated particles are present with sizes below 15 nm, or if the NPF was “long ago”, the below-6-nm-sized particles have coagulated to above 15 nm. Thus, under no-nucleation event conditions the COPAS channels with cut-offs of 6 nm, 10 nm, 15 nm all measure the same concentrations within the experimental errors. Considering a measurement uncertainty of 15% for each channel (see the Weigel et al., 2009, COPAS characterisation) an NPF in principle could be identified, if the N_6 concentration multiplied by 0.85 exceeds the N_{15} concentration multiplied by 1.15. The 0.85 and 1.15 are for the most part determined by counting statistics. However, in most NPF cases the difference between N_6 and N_{15} is much more than 100 particles per cm³ so that NPF is unambiguously identified. The NPF identification is described in more detail in the new paper by Weigel et al., 2011, which currently is on the ACPD discussion page. Particles in the 6 nm to 15 nm which occur in large number densities must have been created by gas to particle conversion. The fact that under no-NPF conditions the difference ($N_6 - N_{15}$) = 0 can be seen from Weigel et al., 2011, Figure 2. Data of “missing” particle concentrations from lower altitudes under no-NPF conditions (below the NPF events) are provided in the same paper and also in Borrmann et al., 2010 from measurements aboard the DLR Falcon. In no-NPF conditions the background number densities of N_6 and N_{15} were equal (within counting statistics) and of the order of tens

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

to hundreds per cm^3 . Furthermore, in Weigel et al., 2011, FLEXPART and FLEXTRA trajectory calculations are provided for the West African flights using the EDGAR inventory. The result of these calculations is that there indeed were SO_2 ground sources providing high enough emissions for NPF at high altitudes. Of course, it is impossible for the individual cloud NPF event to provide trajectory calculations linking a specific ground source along a particular “path” of the air mass through the cloud with the points at which the Geophysica sampled. Thus we believe these are not unproven conclusions.

Comment: On the positive side the ice particle size distribution measurements near African convection and sub visible cirrus (SVC) form a nice data set, and are well presented, except for one serious problem. The FSSP-100 is sensitive to particles between $2.7 \mu\text{m}$ and $31 \mu\text{m}$, which means it is sensitive to cloud droplets as well as small ice. This point is not discussed, and all size distributions and cloud particle number concentrations are presented, and discussed, as if they represent only ice, except in one case on 16 August. What is the evidence for this conclusion of ice in the FSSP data? Were all clouds sampled glaciated? If so how is this determined? The authors discuss one observation of ice particle number concentrations of 8.3 cm^{-3} , which is > 8000 per liter of ice, which is a very high ice crystal concentration. How do the authors know that liquid droplets were not part of this measurement?

Reply: All cloud particle data that are discussed in the manuscript from the 7 and 8 August were measured below -40°C which is below the threshold for homogeneous freezing. In Figure 1 one size distribution from the 11 August was measured at -39°C and seven size distributions from the 16 August were measured at ambient temperatures between -33°C and -40°C , thus, there might be a slight chance that some particles were liquid and not yet frozen. The temperatures during the second cloud part on 16 August varied from -7°C to -41°C , thus, the presence of cloud droplets cannot be ruled out. The CIP images, however, clearly show aspherical ice particles. Furthermore, a

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

large presence of droplets would show up in the aerosol depolarisation which should be reduced when compared to regions of unambiguous ice presence, since droplets contribute to the backscattering but not to the depolarisation. As far as one can tell from the MAS measurements, this was not the case, apart maybe for a few points on a single layer around 344 K.

In the revised version we added a few sentences for clarification of this point on page 764.5 and on page 768.20 reading as:

(1.) "... Ambient temperatures during this cloud crossing were below -47°C , thus, the observed cloud particles are ice particles."

(2.) "...the CIP imaged very large ice cloud particles as snow flakes and aggregates. A few examples are shown in Fig. 15. Since the ambient temperatures in this cloud part varied from -7°C to -41°C , the presence of cloud droplets, detected by the FSSP-100, cannot be ruled out completely. However, the aerosol depolarisation measurements do give no indication for a large presence of cloud droplets. ..."

(B.) Specific and minor comments:

Comment: More specific comments about these points and others follow here, along with some minor suggestions, given manuscriptototically.

747.13 – comma after images.

755.23 – comma after concentrations

762.3 ... are compiled in Table 2, ...

We are grateful for these comments and changed accordingly.

Comment: 762.26: Calling this a mesoscale convective system (MCS) is a stretch of the language. This may become an MCS, but the evidence in Fig. 2 for an MCS is slim. Did these cells in fact develop into an MCS after the measurements, which, however, are characterized as young outflow from an MCS?

Reply: From the satellite images it can be seen that this particular convective system

is in a developing state and grows to MCS size after the probing. The terminology is changed to “developing MCS”.

Comment: 763.5-10 and Fig. 3, 4: Either add altitude to Fig. 3, or present Fig. 4 using potential temperature not altitude. “Apparently, the cloud layer . . .” Why apparently. Do you not trust the Ncloud/IWC/RH measurements? Are not these pretty clear evidence of cloud between 347 and 353 K? Or is it subvisual above 350 K?

Reply: The respective altitudes are added as vertical coordinate on the right axis to figures 3, 8, and 12. “Apparently” changed to “obviously”.

Comment: 763.5-10: Provide a justification for the labels “above outflow” and “outflow”. I presume the latter is from the NO, NO_y mixing ratios, but the discussion is not clear about this. Label Fig 3 with at least the 3 sub layers if not also the AOF/OF 1-2 layers.

For justification of above/in outflow see the first statement of this reply (labelled as “Reply 1” above). Since the sub-layers stretch only over very narrow altitude bands (sub-layer 2 was even mainly probed on level flight) adding these altitude bands to figure 3 would lead to confusion. Therefore, we would like to not label the figure with the particular altitude bands. However, we added a statement in the text specifying altitudes and thicknesses of all three layers.

Comment: 763.11: Lower compared to what, the layers below?

Reply: Yes, we changed this sentence to:

“The uppermost sub-layer (denoted as “Sub-layer 1” in Fig. 4) contained lower cloud particle concentrations, compared to the sub-layers below, in coincidence with a strong in-cloud New Particle Formation event (NPF).”

Comments: 763.15-20: Several comments. . . considered as MCS anvil part . . . What does this mean? Do you intend, “. . . considered as part of an MCS anvil”?

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

If so where is the MCS? See comment above on MCS classification. What about the “ice particle data” identifies this as an MCS anvil? Then, but the low NO/NO_y indicates it is not outflow from an MCS. This is also confusing. How does a thunderstorm produce an anvil without the anvil being part of the outflow from the storm? Does outflow have some special definition that I am missing?

Reply: We changed to “considered as part of the developing MCS anvil”. As stated above in the outflow identification, ice particles itself do not define an outflow since they might have formed otherwise (like a pileus cloud) or they might have sedimented from an outflow above.

Comment: . . . The labeling of the region with high particle number concentration as a new particle formation event is not justified. The Weigel et al. (2011) reference is “in preparation”, and even if submitted to acpd is not a refereed source, yet this reference is repeatedly used. The “evidence” is only for an abrupt increase in particle concentration, which could result from transport, which has not been ruled out. In Fig. 3 the particle concentrations are not supplied below 348 K, which would help identify how unique the layer is. Why are these particle data not shown? Then finally talking about “quenching” a nucleation event, which has not been demonstrated, is artificial and speculative, and should be avoided.

Reply: This comment has been addressed in our reply labelled as “Reply “2” on NPF issue” above. The reference Weigel et al. (2011) is now available and citeable from ACPD and, indeed, it is almost a “companion paper”. As the timing for both manuscripts now is, we anticipate them both through the review process fairly simultaneously. If our MCS paper here is accepted and if the editor likes, we can procrastinate the publication of this manuscript until the Weigel et al., 2011, also is accepted. However we believe this is not necessary.

COPAS operates reliably at pressures roughly between 400 hPa and 50 hPa (see the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Weigel et al., 2009, instrument characterisation paper), thus, measurements below 348K were not available for this flight.

Comment: ... “Below, from 56 385–56 557 sUTC, the third cloud sub-layer (“Sub-layer 3” in Fig. 4) extended between 11.9km and 11.0km ...” Why now all this detail and switching to time to the second, when this wasn’t necessary before? Also “the third cloud sublayer (“Sub-layer 3” in Fig. 4)” is rather redundant. Could you not just say here, and elsewhere, ...Cloud sub-layer 3 (Fig. 4) extended ...

Reply: The times have been removed and replaced by references to the respective sub-layers.

Comment: ... The elevated NO and NO_y is indicative of lightning activity or surface air, or? Please clarify.

Reply: To clarify, an additional sentence is included, reading:

“... involving high particle number concentrations and very high values for NO and NO_y. Since soil emissions may contribute up to 1 nmol/mol (Stewart et al., 2008), the mixing ratios observed here give a clear indication for production in lightning. Thus, this is the MCS outflow where the detrainment must have occurred very recently, since the elevated NO and NO_y had not been diluted yet.”

Comment: 763.25: How is the source region of the NO_x identified?

Reply: The convective core was termed here as source region of the NO_x (separation of charge occurring in the convective cores). We changed the wording to:

“From the enlarged satellite image and the flight track of the aircraft one can estimate that the sampling occurred at a maximum distance of roughly 30 km from the convective core.”

Comment on Fig. 4: I presume the widths of the blue bars differ because the ice particle concentrations are different enough that larger sampling times are re-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



quired to reduce the counting uncertainties? Still why are the very thin sampling regions picked where ice particle number concentration is slightly disturbed rather than where it is quite stagnant?

Reply: Yes, the width depended on the counting statistics, which were excellent during the outflow event (c.f. small error bars). Furthermore, as stated in Section 3.3 in the separate reply on instrumental performance and shattering, a problem in the data acquisition software occurred from time to time (not continuously) during the outflow event. Therefore, the averaging time periods needed to be adapted to those times where we are sure the CIP data are of good quality. However, the CIP concentrations had only a minor contribution to total concentrations (mean N_{CIP} were around 0.02 cm^{-3} , maximum 0.2 cm^{-3}).

Comment 765.9-16: Same complaint as above about classifying the regions of high particle formation as particle nucleation events. This may be the case, but the evidence shown here is equivocal, although better here with the volatility measurements. In addition from the data shown, the reader cannot discern when the high particle concentrations are in or out of the cloud, and similarly for the volatile fraction, in our out of cloud. Finally why no particle concentrations below 353 K, yet volatility below this point?

Reply: As stated in the manuscript: The profiles "include measurements from ascent, descent, and one dive. Thus, spreads in the single parameters might result from the profiling at different locations." Therefore, the in and out of cloud parts can indeed not be discerned. However, we set a reference to Section 4.3.3 "New particle formation event" and Figure 9 where this particular event is discussed and can clearly be seen. There were two disjoint COPAS systems aboard, meaning two separate boxes with separate inlets, electronics, controls, at two different locations aboard the aircraft. We are very conservative when deciding which data to accept. Both COPAS units (one of them providing the volatility measurements) need time to stabilise after start-up until they commence delivering acceptable data. The COPAS unit with the volatility channel

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

provided reliable data somewhat earlier than the other system, possibly due to the shorter inlet line, which led to the different data contribution.

Comment: 765.26: “The other coloured dotted lines of additional COPAS data show mostly nonzero differences ...” What is intended with this statement? Does it not just indicate there are particles in these size ranges? What are the authors concluding from this fact?

Reply: This is one of the few situations where all instruments and all channels concurrently delivered high quality data, and one almost could derive “two bin size distributions” in the nanometre size range. The conclusion from the non-zero differences is that there are N_{6-15} particles larger than 6 nm and smaller than 15 nm, then in addition N_{6-10} particles larger than 6 nm and smaller than 10 nm. Thus, freshly nucleated particles exist, some of which already coagulated forming (slightly) larger sizes this way. The University of Denver group uses the same approach for the WB-57 and ER-2, although they have a CPC capable of delivering size distributions with 5 bins. We appreciate this comment and include an explanatory statement.

Comments: 765.28-766.1: The CO₂ concentration is changing rapidly through the region of high particle concentrations, suggesting a region of strong mixing? This complicates the picture of the conditions which produced the high particle concentrations. 766.1-9: This is one explanation of the observations, but not the only one, and one could imagine other scenarios not requiring new particle formation. Even if there were new particle formation, the argument that a significant fraction would have already grown above 10 nm needs more support. Could coagulation do that in the time available with the particle concentrations observed? Thus the claim here of another “NPF” event has to be tempered with other possibilities.

Reply: Due to the large amount of N_{6-15} of 5000 cm^{-3} gas to particle conversion must have been the underlying process, similar as in the observations of Lee et al., 2004. We

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

do not have evidence from the literature showing other realistic possibilities. In order to answer the question whether the time was sufficient for coagulation to proceed, one needs detailed cloud microphysical modelling. On one hand we do not know how long the NPF proceeded and the initial number concentration of the smallest particles. On the other hand we cannot readily specify at what rate molecules and smallest particles are lost to the surfaces of the pre-existing cloud particle surfaces. Due to the length of the manuscript we abbreviated this discussion and included a reference to the Weigel et al., 2011, paper, which is more detailed (including model calculations) with respect to the nucleation events.

Comment: 766.15-28: This section should be re-titled to entrainment and mixing as possible source of the high particle concentrations, and then rewritten to address that possibility. The present title assumes the “NPF” has been proven, and the paragraph and Table 3 add nothing new to Figure. 9, except the relative humidity, which could be added to Fig. 9. In short this section should either be deleted, or really focused on what mixing would mean to possible sources for observations of high particle concentrations. The present effort here provides nothing new.

Reply: We removed the headline “entrainment and isobaric mixing . . .” of the submitted version and merged the remainder into the previous section “New particle formation event”. However, we would like to retain Table 3 with the detailed numbers especially because we hope someone from the cloud modelling community picks up these open questions. We added a remark highlighting that the “mixing hypothesis” remains speculative without detailed model calculations.

Comment on Fig. 10: Which measurements are above, inside, below the outflow? What identified the outflow? Why include CO₂ on two panels, when it is exactly the same, and then missing on all other panels? What is the CO₂ used for?

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Reply: CO₂ was taken for these cases to identify whether the cloud was located in an outflow region or not. The altitude profile shows a very distinct minimum, thus, clouds in this altitude band are considered as outflow clouds. The CO₂ mixing ratios were the same by chance; the third event was found in the same altitude band as the CO₂ minimum and is therefore also regarded as outflow. Furthermore, the trajectory analysis indicates a convective influence on these clouds. From other flights during SCOUT-AMMA on convectively less active days we know that the CO₂ mixing ratios at similar altitudes as the outflow events were 4 to 6 μmol/mol (i.e. significantly) higher. The lower right corner summarises the three events and additionally shows cloud size distributions from above and below the outflow (identified by higher CO₂ mixing ratios).

Comment: 767.1-11: Here is a good example of my confusion about outflow. Here are multiple cloud passes with ice crystal size distributions, but some are classified as outflow and some not. What is the difference? How is the outflow identified? Where is the air in the other samples from?

Reply: The outflow air masses on this particular flight are identified by low CO₂ mixing ratios (as visible in Figure 8 between 353 K and 360 K). This is stated on page 765.3-6. Thus, clouds located above/below this CO₂ minimum band are classified as above/below outflow. As stated in Reply 1, these clouds could be reminders of pileus clouds, sedimented out from an outflow or formed in situ. Thus, these air masses do not necessarily need to be processed in convection but could be residing at those altitude levels a longer time. In any case the air above and below the outflow is distinctively different from the “low CO₂ portion”.

Comment: 767.27: What does an outflow age represent? The time since the air left the cloud, or since it was in the updraft core, or since it was at the surface, or ???, and how is it measured?

Reply: The age represents the time between the measurement and the time when the trajectories “intersected” with the convective region in the past. To clarify this, an addi-

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

tion is made to the sentence on page 762, line 9:

“Trajectory analysis (Fierli et al., 2011) indicate an age of less than three hours, i.e. the time between the measurement and the time when the when the trajectories intersected with the convective core region in the past.”

Comment: 768.6: Start a new paragraph with, “A closer ...”, then explain what criteria are used for this separation, and that they differ from the boxes – BOF1... in Figure 13. Here is a good example, in the discussion of the first layer, of the difficulty of using the FSSP-100 for ice particle measurements. Here for the first time the authors point out that the FSSP-100 could be sensitive to particles other than ice, e.g. here aerosol particles, precipitation, and haze particles. But if it is sensitive to these particles it also sensitive to cloud droplets.

Reply: The separation is visualised in the figure (similar to the sub-layers in Figure 4). The criteria for the separation are as follows: Part1 – cloud part with small particles, detected by the FSSP only. Part 2 – cloud with ice particles (as imaged by the CIP) without outflow signatures, part 3 – cloud part with outflow signatures. We commented on the possibilities of frozen vs. liquid cloud droplets as function of temperature above. We also supplied an extra document on the instrument related issues and shattering in response to the other reviewer’s comments. In this document we detail the specific procedures of the size binning. In particular, we artificially decreased the size resolution and this way considerably increased the bin-widths for the FSSP sizing. In the end the size bins are as wide, and the binning as result of the T-matrix method for ice particles and from the Mie theory calculations for spherical objects as similar, that the small aspherical ice particles and possible spherical droplets should be sorted correctly into the corresponding size bins. For the particles smaller than $10\ \mu\text{m}$ (a guess) the reviewer has a point here. There may be large aerosol particles or haze droplets (of unknown refractive index) counted into these size bins. These cannot be differentiated. We added a remark on this in the instrument section describing the FSSP. In addition we note in the text on Figure 14 that this may be the case here.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



Comment: 769.24-26: ... events did the CIP detect shattering. ... SVC are noticeably affected ...

Reply: Unfortunately, we do not understand what the reviewer would like us to do or comment on. Hopefully, this is clarified by the separate reply.

Comment: 769.28: How did Lawson do it, if the particles are too small to be imaged?

Reply: Paul Lawson used measurements by the CPI to determine particle shapes. The CPI has a lower detection limit and a higher resolution than the CIP used in this study and therefore, particle shapes could be deduced. This is clarified in the revised manuscript.

Comment: 775.21: $8.3 \text{ cm}^{-3} = 8300$ per liter. Are you confident of this measurement? It is very high for ice, but not for cloud droplets.

Reply: The ambient temperatures during this cloud crossing were below -47°C , thus, below the homogeneous freezing threshold and consequently we are confident that the cloud particles were not liquid but ice particles. Similarly high ice particle concentrations have been observed before (see, e.g. Pruppacher and Klett, 1997; Reprint 2000, Chapter 2; Fig. 2-42).

Comment: 776.29: I do not understand the purpose of this contrast of particle shapes in outflow during AMMA with former SVC particle observations. Is this to suggest that the SVC shapes in AMMA, which were too small to be observed, were similar to other SVC measurements?

Reply: The contrast in depolarisation and colour index derived from MAS measurements shows that ice particles in the MCS outflows and SVCs have a different morphology, i.e. different shape structures. Former SVC observations have shown different shapes than those observed in the MCS clouds. Thus, it remains possible, that shapes in the SVCs observed during AMMA were similar to those observed in the other

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

campaigns.

Comment: 777:3-10: These conclusions on NPF are premature based on the measurements presented and discussed.

Reply: In the section on the NPF we rephrased more cautiously, and highlighted the need for detailed cloud microphysical modelling. Furthermore we referenced the Weigel et al., 2011, paper again remarking that some modelling is provided there. In the conclusions it is also said “possibly due to entrainment and mixing”. With this we think we have done enough to emphasise the preliminary nature of the interpretation of the observations.

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Full Screen / Esc

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Interactive Discussion

Discussion Paper



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