Supplement to:
In situ measurements of tropical cloud properties in the West African Monsoon: Upper tropospheric ice clouds, Mesoscale Convective System outflow, and subvisual cirrus

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On the issues of instrument performance and shattering artefacts for the FSSP-100 and CIP

For the measurements presented here (and any other measurements performed with *in situ* cloud particle probes) there are two main issues: the instrument performance and the question how to ensure that shattering does not affect the measurements significantly. Therefore, this supplement presents the general arguments related to those two issues and specifies how data have been treated in order to avoid shattering biased results from the combined FSSP-100 and CIP measurements.

1 General arguments

1.1 Experiments dedicated to shattering

The wind tunnel studies as well as the measurements underlying the publication by Korolev et al. (2011) were conducted under vastly different conditions when compared with the measurements from the West African MCS. The SCOUT-AMMA measurements were performed at much lower temperatures (i.e. below -40°C and down to -80°C) and at much lower humidities and number concentrations than (1.) the wind tunnel measurements and (2.) the measurements in mostly mixed phase clouds of the Airborne Icing Instrumentation Evaluation (AIIE) campaign. Thus, an extrapolation of the factor 100 to 1000 enhancements due to shattering from the reported conditions of AIIE campaign to the low temperature, low humidity, low concentration conditions during SCOUT-AMMA is not justified until solid evidence for this is provided for example by dedicated wind tunnel experiments or more airborne instrument intercomparisons (like e.g. Jensen et al., 2009) at such UT/LS conditions. This is also suggested by the study of Lawson (2011), who performed measurements in anvil cirrus at temperatures from -30°C to -63°C and concluded that the post processing interarrival time analysis is well suited for rejection of suspected shattered particles and that this analysis might even carry more weight than the application of modified tips. For example, the ice particles we measure have no quasi liquid layers and may even consist of a glassy physical structure (Murray et al., 2010) and thus their break-up and bounce behaviour will differ from the behaviour of the much warmer hydrometeors. For these reasons it is by far premature to conclude that all FSSP-100 data are obsolete because of the shattering influence, especially when other data from partly overlapping instruments -like the CIP- are available. One should still be able to provide useful data after careful analyses of the individual size distributions. This "careful analysis", which ultimately led to the rejection of data not included in the paper, is described below in Section 2 and 3. Of course, the influence of shattering cannot be exclude completely, but the presented data can be limited to cases where shattering influence is believed to be low, and the possibly contaminated data can be highlighted to the reader.

1.2 CIP and FSSP-100 performance

Jensen et al. (2009) state: "The agreement between size distributions derived from the CIP and 2D-S imaging instruments is excellent throughout their overlapping size range (>50 μm)." The authors refer to tropical measurements in Costa Rica at 11.4 km to 12 km altitude and probably these are similar to the "young outflow" conditions of 7 August 2006, in Section 4.2 of the paper.

Concerning the FSSP-100, Cairo et al. (2011) state in their conclusions: "A comparison of optical properties for tropical high altitude cirrus clouds, directly measured and inferred from particle size distribution observations, has been carried out. Results suggest that the fraction of the size spectrum available from FSSP-100 particle counter observation, i.e. particles with diameters from 2.7 μm to 31 μm, is effective in reproducing cirrus optical properties in the visible part of the spectrum. This result keeps validity for backscattering cross sections spanning over 5 orders of magnitude. Optical particle counters observations are thus a valid tool to assess the cloud particle density and to provide size distributions for modelling cloud microphysical processes and radiative effects in the visible region of the spectrum." To arrive at this result Cairo et al. (2011) used a careful selection of the SCOUT-O3 data from Darwin, Australia, the SCOUT-AMMA data from West Africa, and the TROCCINOX data from Brazil, all obtained with our instruments aboard the Geophysica. They applied the MAS backscatter sonde on Geophysica to measure directly backscatter and depolarisation in the vicinity of the aircraft within the tropical high altitude cirrus clouds and compared this data with the backscatter ratios derived from the *in situ* measured particle size distributions from the FSSP-100. The optical backscatter for the most part depends on the small particle concentrations reported by the FSSP-100 and much less on the larger sizes from the CIP. If shattering had enhanced the corresponding small particle number densities by factors of

100 to 1000, then this intercomparison would have severely failed. Of course, this only holds for the used data sets and may not be "extrapolated" to cirrus in general.

1.3 Gas phase derived IWC vs. CIP and FSSP-100

In the study by de Reus et al. (2009), the IWCs derived from Lyman-α hygrometer H_2O measurements are directly compared with the concurrently measured particle IWCs, as calculated by using the Baker and Lawson (2006) scheme, for the observations during the SCOUT-O3 campaign. They showed that the IWCs independently derived from these measurements did agree remarkably well within the measurement uncertainties. Measured IWCs ranged between $10^{-5}\,\mathrm{g\,m^{-3}}$ and $10^{-2}\,\mathrm{g\,m^{-3}}$. If shattering had significantly influenced the FSSP-100 results then discrepancies between the IWCs derived from the gas phase and the particle measurements could have been expected. Thus, shattering is believed to not significantly alter the IWCs and the FSSP-100 size distributions during the SCOUT-O3 measurements. For IWCs larger than roughly $0.001\,\mathrm{g\,m^{-3}}$ the underlying volumes are mostly influenced by the large particles. For the smaller IWCs the FSSP-100 size range contributes more than 50% of the IWC. At least here one can assume that additionally detected particles from shattering would enhance the IWC artificially for the CIP and FSSP-100 on the abscissa. However, in the graph the IWC from the particle instruments are even too low when compared with the gas phase instruments. The IWCs of the encountered outflow events as presented in this study range from $6\times10^{-6}\,\mathrm{g\,m^{-3}}$ to $6\times10^{-2}\,\mathrm{g\,m^{-3}}$ with many values below $0.001\,\mathrm{g\,m^{-3}}$. For the data presented from the subvisual cirrus and uppermost UT cirrus the IWC were much lower than $0.001\,\mathrm{g\,m^{-3}}$. (Note for clarity: A similar plot unfortunately cannot be prepared for the SCOUT-AMMA flights, because the four involved instruments were not often enough operational concurrently at the same time and while inside the clouds.) At least for these cases it is unlikely that shattering influence of factors between 100 and 1000 would have gone by unnoticed.

1.4 Comparison SCOUT-AMMA/SCOUT-O3 data with CEPEX

In Figure 1 a summary of the SCOUT-O3 data from Darwin and the SCOUT-AMMA data from West Africa is presented together with the parameterisation which McFarquhar and Heymsfield (1997) extracted from their CEPEX measurements. In the lowest potential temperature bin of Figure 1 IWCs larger than $0.001\,\mathrm{g\,m^{-3}}$ were found such that the argument from Section 1.3 is not applicable here. However, our measurements agree quite well with the CEPEX parameterisation particularly for the particle sizes below roughly $20\,\mu$m. The major differences between CEPEX and SCOUT-AMMA occur at the very large sizes. During CEPEX the particles were measured with a VIPS and a 2-DC probe. The VIPS has an entirely different "inlet"-geometry and measurement principle w.r.t. the FSSP-100 and shattering -presumably- is not an issue. If shattering had introduced artefacts to our particle number concentrations here on the factor 100 to 1000 levels like indicated by the study of Korolev et al. (2011), then this intercomparison would have turned out very differently. This point is better visible in Figure S1, taken from the PhD thesis of Wiebke Frey, 2011. Again the lognormal fits of the measurements compare well with the calculated CEPEX size distributions, especially for the small sizes in the lowest potential temperature bin. (For the higher potential temperatures the results lie well below the CEPEX data and/or particles were too small for significant shattering.) Here, the ordinate displays absolute concentrations and not normalised as in Figure 1 of the paper.

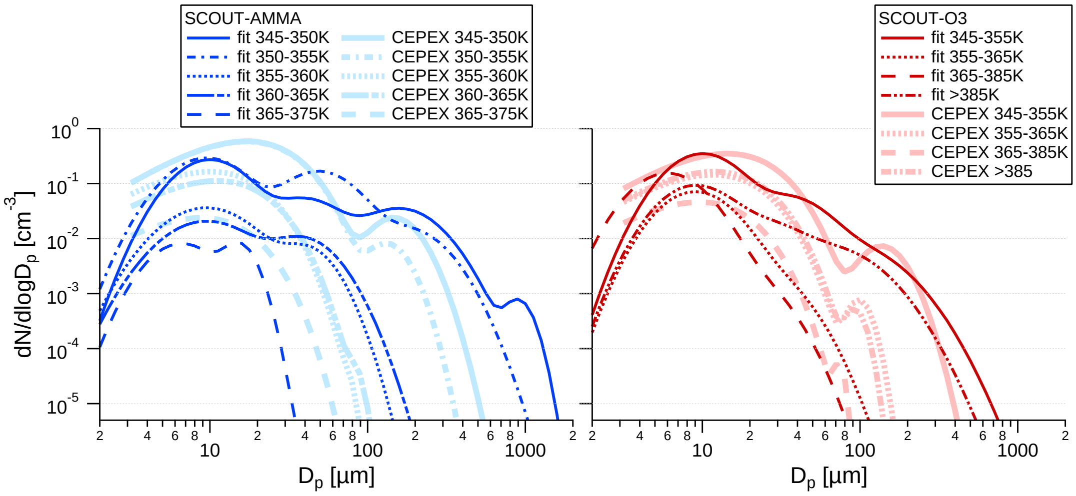

Figure S1: Lognormal fits of the size distributions and CEPEX parameterisations for SCOUT-AMMA (left panel) and SCOUT-O3 (right panel). Taken from Frey (2011).

Based on these general arguments we believe that careful inspection and selection of the data does allow us to retain the CIP data and some of the FSSP-100 measurements.

2 Shattering artefacts for the CIP and small particle detection

2.1 Small particle detection

It has been questioned whether the CIP has problems with the detection of particles with diameters less than 150 μm. The CIP used for this study has newer electronics with faster response times compared to the first CIP instruments and the 2DC probe. Baumgardner et al. (2001) described this improvement and determined that there is no more dependency of the depth of field on the aircraft's velocity (which was described by Baumgardner and Korolev (1997). Furthermore, Lawson (2011) stated, based on recent measurements, that the CIP reliably measures droplets of 50 μm at speeds below 150 $\mathrm{m\,s^{-1}}$. During our measurements the aircraft velocities were 135 $\mathrm{m\,s^{-1}}$ (young outflow case), 140 $\mathrm{m\,s^{-1}}$ (recent outflow case), and between 145 $\mathrm{m\,s^{-1}}$ and 157 $\mathrm{m\,s^{-1}}$ in the aged outflow. Thus, we assume that the CIP has reliably reported the particles over its entire detection range, keeping also in mind the statement from Jensen et al. (2009, see Section 1.2 above).

2.2 Interarrival time criterion for shattering of the CIP

If the interarrival time threshold is set to too high values, a significant fraction of legitimate particles may be removed. If it is set to too low values, a significant part of shattered particles may not be removed. Therefore, the interarrival time threshold was determined specifically for each particular flight. In the analyses of the SCOUT-AMMA data the interarrival time rejection has always been applied to the whole data set of each flight including the subvisual cirrus cases. The shortest interarrival time encountered by the CIP measurements during the four SVC cases was 180 μs which is much larger than the interarrival time thresholds of 2.6 μs to 5 μs which were adopted for the respective data sets. For this reason no particles were erroneously removed from the SVC cases. The shortest interarrival time encountered during the aged outflow events from 11 August was 1300 μs and thus, no particles have been rejected during these events.

2.3 Percentage of shattering for the CIP data

In order to gain a feeling of how many CIP images are rejected due to shattering, the fraction of shattered particles is calculated as percentage of the total number of particles detected by the CIP. The percentage itself does not give information about the particle sizes of the shattered artefacts. For the SCOUT-AMMA data over 85% of the shattered particles are found in the smallest three size bins (i.e. particles smaller 175 μm). In Figure S2 the CIP size distribution of outflow event 1 (7 August 2006) from the paper is shown, where 10% of the particles detected by the CIP were shattered fragments. The shattered fragment particles are displayed in red, while the size distribution of all particles including the shattered particles is shown in black. The blue curve gives

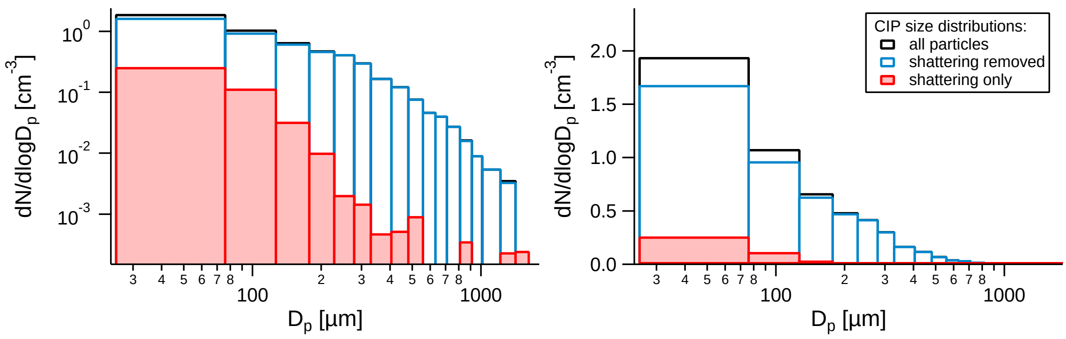

Figure S2: Comparison of size distributions including shattered fragments (black), shattered fragments only (red), and with shattering removed (blue) on a logarithmic and linear scale for better comparison.

the resulting size distribution with removed shattering. The right panel provided the results with linear ordinate for better clarity and one can see that the removal of the shattered particles does not affect the size distribution significantly. For the most part the difference probably is within the limits of uncertainty due to counting statistics and the sample volume.

3 General treatment of FSSP-100 data and relation to shattering artefacts

3.1 Sizing of FSSP-100 data into bins: Application of the T-matrix method

In principle the size bins from the T-matrix model of the FSSP (models 300 and 100) scattering geometries after Borrmann et al. (2000) can be used. However, the adopted T-matrix code does not converge anymore for particles with sizes above 16 μm diameter. The FSSP-100 data presented in the paper extend from 2.7 μm to 29.2 μm. Based on the T-matrix method this range could be subdivided into 15 bins including three bins from 16 μm to 29.2 μm. It needs to be assumed that the T-matrix results can be extrapolated to these last 3 bins. In practice the size range from 2.7 μm to 29.2 μm covered by the FSSP-100 is subdivided here into 7 bins only. This artificial reduction of the size resolution was done by carefully inspecting the corresponding scattering cross sections from the Mie- and the T-matrix curves and defining the bin limits "manually". The reason is, that in fact it is difficult to apply the FSSP-100 for ice particles and that the T-matrix method only can serve to demonstrate -within narrow limits- that it is not impossible to measure inside cirrus. (This was the original intent of the Borrmann et al., 2000, paper.) To be conservative and reduce potential cross-sensitivity, where particles are counted into bins they do not belong to, the size resolution is further decreased from 15 to 7 bins. For the subvisual cirrus data the counting statistics mostly is not good, such that a further reduction of the size resolution to only 6 bins is justified even more. In summary, the T-matrix scattering cross section curves are used but the size resolution was decreased to one half of the theoretically possible number of bins.

3.2 Combined FSSP-100 and CIP size distributions

As shown in the drawing in Figure S3 the size distributions from the FSSP-100 are simply overlaid onto the CIP size distributions. The first CIP size bin actually extends down to sizes of 25 μm but is only displayed down to 29.2 μm which is the upper size limit of the FSSP-100 size distribution. In the paper the presented size distributions consist of what is delineated by the green lines. No smoothing or running averages have been applied to the size distribution data and the unaltered measurement results are shown with error bars based on propagation of counting statistics and sample volume uncertainties. For us this constitutes the most transparent and honest approach of presenting the data. The size distributions from the FSSP-100 are considered as contaminated by shattering in all bins, if the two highest bins (shown in red above) do not well overlap with the lowest CIP bin (shown in blue).

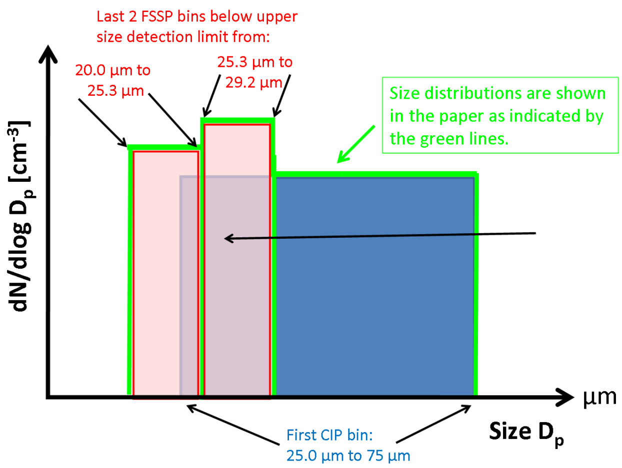

Figure S3: Schematic showing how the FSSP-100 and CIP size distributions are combined. No smoothing or running averages have been applied.

3.3 Rejection of FSSP-100 data based on poor overlap with CIP

Such cases of poor overlap always exhibit much higher FSSP-100 concentrations in the last two bins than the CIP in its first bin. The CIP is considered as reporting the correct concentrations (after application of the state-of-the-art corrections), because unlike for the FSSP-100 there are tools like interarrival time analyses available for identification of shattering events. If both instruments exhibit overlap within their error bars, the FSSP-100 data are considered not to be significantly influenced by shattering.
Nevertheless, the volume of data that had to be rejected based on poor overlap is rather small. All relevant measurements for this study were obtained at potential temperatures above 345 K and poor overlaps were mostly found below this level. Data from below

roughly 10 km are not shown in the paper except for the time series of 16 August in Figure 13 and size distributions in Figure 14. From the measurements above 345 K, only 3 size distributions needed to be discarded. Specifically, on the flight on 8 August one and on the flight on 11 August only two size distributions (accumulated from time periods of 10 and 20 seconds, respectively) were removed at potential temperatures between 345 K and 346 K. Thus, only the lowest potential temperature bin in Figure 1 (lowest panel) of the paper is affected from such rejection. Data obtained in the outflow regions on 7 August and 16 August and in the subvisual cirrus cases are not affected by data rejection due to poor overlap.
Furthermore, due to a problem in the data acquisition software (which was easily recognised a-posteriori) some data shortly before and during the MCS outflow crossing on 7 August was lost. This lost data should also have been measured at the lowest considered potential temperatures (345 K – 350 K). The measurements obtained in the outflow regions on 16 August and 11 August and in the subvisual cirrus cases are not affected by this problem.
"Poor overlap conditions" seem to occur preferably when the Geophysica performs particular manoeuvres like narrow turns. It has to be borne in mind that good overlaps between aircraft borne aerosol instruments of such different nature are all but common occurrence. For example the agreement in the overlapping size ranges of the FCAS and FSSP-300 instruments deployed on the ER-2 in conditions, which are simple by comparison, namely within the stratospheric Pinatubo aerosol, often enough was much worse (Wilson et al., 1993; Jonsson et al., 1995).

3.4 Correlation analysis of small crystal concentration - large crystal mass

In order to identify measurements potentially affected by shattering Jensen et al. (2009) suggest to perform an analysis using correlations between large particle IWC and small particle number concentrations. Such analyses have been performed in order to address the reviews and the results are reported in this section.
Figure S4 shows the concentration of FSSP-100 particles vs. IWC from the CIP particles above 125 μm for the flights from 11 and 16 August. The data from 11 August are from inside and the vicinity of aged outflow events, and show no correlation. This follows the

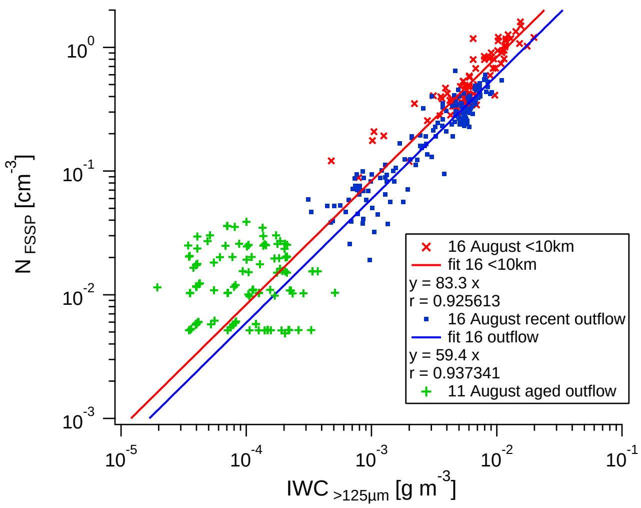

Figure S4: Correlation analysis of small crystal number concentrations versus large crystal IWC following Jensen et al. (2009). See text for further explanation. Taken from Frey (2011).

expectation of Jensen et al. (2009) for aged clouds in case shattering is not significant. By contrast, Jensen et al. (2009) do expect a correlation for young ice clouds based on microphysical arguments. Furthermore, they anticipate such a correlation in case shattering introduces significant amounts of artefacts. Indeed, the measurements from 16 August in young clouds and recent outflow also exhibit such a correlation. We see more shattering for the cloud measurements below 10 km from the CIP data, and the red data points (plus line) indicate a stronger slope of the correlation compared to the recent outflow case in blue data points and line (Note the log scales and the absolute difference in the coefficients for the slopes). The young cloud data of 7 August show a similar figure as those from the 16 August but exhibit a somewhat worse correlation. Thus, this analysis confirms the expectations of Jensen et al. (2009).
In order to quantify possible shattering Jensen et al. (2009) suggested a further step for analysis. They applied 5 different filter criteria to their CAS data to find enhanced, spurious concentrations due to shattering. As can be seen from the figure above there

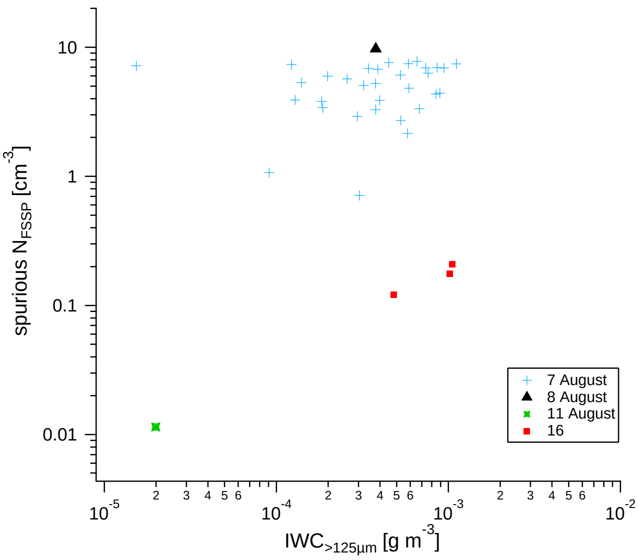

Figure S5: Correlation analysis of spurious small crystal number concentrations (filtered) versus large crystal IWC following Jensen et al. (2009). See text for further explanation. Taken from Frey (2011).

are only very few measurements left in the SCOUT-AMMA data after this filtering is applied. For 7 August, only, enough data points (light blue) remain after the filtering to perform a correlation analysis. However, only a poor correlation results with a Pearson coefficient of 0.05, which indicates that the few spurious particles do not depend on IWC, i.e. "plus-minus" the presence of the largest particles. Thus, shattering seems to have not impacted our measurements to a significant amount.
In summary the SCOUT-AMMA data confirm the assumption of Jensen et al. (2009) according to which (1.) in young outflow scenarios a correlation between the IWC from particles with sizes above 125 μm and the number densities of small particles is present, (2.) no correlation in SVC and aged outflows can be found, and (3.) not as much "alarming" data points survive the filtering as was the case for the CAS.

4 Summary of the effects of shattering in the data of the paper

This section summarises which figures of the paper possibly are affected by shattering artefacts and what has been done in order to highlight potential problems to the reader.

Figure 1 The lowest two panels for the potential temperature bins from 345K to 355K may be influenced by shattering. However, we consider this as insignificant based on the arguments from Section 1.2, 1.4, 3.2, and 3.3. The panels for the higher potential temperature bins are not affected because there the CIP did not detect particles which are large enough to cause significant shattering, or simply, the CIP did not detect any shattering according to the interarrival time method.

Figure 5 The size distributions "AOF1" and "AOF2" contained less than 10% shattered particles in the CIP size range (1.5% and 7.4%, respectively), furthermore, the overlap of FSSP-100 and CIP size range is excellent. "OF1" and "OF2" both contained 10% shattered particles. Therefore, the size range of the FSSP-100 in these size distributions is highlighted.

Figures 10 and 16 These figures are not affected by shattering because the largest reported particles are too small and the interarrival times for the CIP were by far above the threshold value.

Figure 14 The mean percentage of shattered particles in the CIP size range for the cases "BOF1", "BOF2", "OF3", and "OF4" is above 10% (12.8%, 14.5%, 12.2%, and 13.5%, respectively). Therefore, the FSSP-100 part of these size distributions is highlighted as in Figure 5. "OF5" contains less than 10% shattered particles (8.4%), furthermore, the number densities of particles larger than 500 μm is very low and the analyses shown in Section 3.4 indicates that this event is not affected significantly by shattering.

Figure 19 Here, only the upper two red curves might be influenced from shattering in the size range below 30 μm, although we believe this is not significant based on the arguments in Section 1.2, 1.4, 3.2, and 3.3. The black curves are most likely not affected because of the low number concentrations of the large particles and the interarrival times of particles detected by the CIP were by far above the threshold value. The green curves may be contaminated by shattering in the FSSP-100 size range (although we believe this is insignificant) while the data for the CIP have been corrected for shattering using the interarrival times.

We believe with these measures we have performed state of the art analyses as far as these are possible for the adopted instruments and we provided best possible transparency on the use of the data to enable readers to arrive at their own opinions. We also believe that the data should be presented -provided all these caveats highlighted properly- because such measurements from West Africa are practically non-existent, and further *in situ* experiments like this will be next to impossible in the near to mid range future.

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