## **Authors' Final Response**

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We are grateful to both the reviewers for the insightful comments and suggestions on our manuscript. We respond to them in detail below. Reviewers' comments are given in black, our anwers in blue. The responses mention specific corrections which were applied as the result of reviewers' suggestions. Next, we explain minor corrections introduced by ourselves. We also attach the full revised manuscript with all the changes marked (generated with latexdiff).

## **Response to the Anonymous Referee #1**

## **General comments**

It is challenging to characterize the sample volume of the shadowgraph system (Sect. 3) without accurate information regarding the properties of the plume (e.g. uniformity, size distribution of poly-dispersed droplets). Uncertainties in the calibration method can have large uncertainties on the droplet number concentration and size distribution. The assumptions and speculations in Sect. 3 are all justified, but some uncertainties remain. For example, the different lens systems show different dependencies of the number concentration on the horizontal, vertical and axial position (Fig. 8), some of which are attributed to the non-uniformity of the plume (x2, x4), whereas others are explained by instrumental flaws (x1). A better characterization of the plume would strengthen the interpretation.

We agree with the referee that the better characterization of the plume would strengthen the interpretation, i.e. allow to distinguish the effect of its non-uniformity from instrumental flaws. However, we were not able to do this with the available instrumentation. Therefore, we point out in the text that some particular conclusions cannot be delivered with high confidence based on our experiment.

The most important correction we developed involves limitation of the depth of field  $z_{95}$  in Eq. (5). It was first derived analytically and the experiment proved to confirm its validity. It did not serve to derive the calibration, i.e. none of the coefficients used in data processing was fitted to the collected data. The calibration constants  $a_1, \ldots, a_6$  were prior provided by the manufacturer and resulted from the calibration performed with the Patterson globe targets (Kashdan et al., 2003).

Concerning dependency of the number concentration on the position illustrated in Fig. 8, we do not claim that in case of lens setting x2 and x4 the effect is entirely due to the non-uniformity of the plume. What we mean is that such dependency might have been caused by the plume properties, hence we are not entitled to blame instrumental issues on the ground of our results. Lens setting x1 features such a large difference that, in our opinion, it is hardly possible to explain it with the properties of the plume only.

Regarding the particle sizing, a FMAG was used to produce mono-dispersed droplets (in the range of 15 to 72 µm in diameter). For the size calibration experiments, droplet diameters of 20.13, 39.35 and 57.55 µm were chosen. Would it be possible to produce size calibration experiments with smaller sizes (e.g. by using a Vibrating Orifice AerosolGenerator (VOAG); or PMMA/PSL spheres with atomizer or fluidized bed)? Previous studies have shown that sizing uncertainties are largest for the smallest particles, so I would recommend performing sizing experiments for smaller sizes and if possible include additional instruments for better validation. Furthermore, cloud droplets are generally smaller than 20 µm. For example, the cloud droplet size observed in Sect. 5 all lie below the smallest calibration size applied in this study (20.13 µm).

We are aware of this limitation. Naturally, it would be of advantage to verify the sizing of smaller droplets as we also expect largest uncertainties for the smallest particles. However, the instrument which was at our disposal (FMAG) can reach down to 17  $\mu$ m in diameter using ultra purified water (UPW) as the fluid (Duan et al., 2016). In the course of the experiment, we tried to generate droplets smaller than 20  $\mu$ m but this resulted in quite a wide spread of the diameters. In FMAG, the size is controlled by the liquid flow rate and the vibration frequency. To obtain small sizes, high vibration frequency is necessary (e.g. 200 Hz for the diameter of 17  $\mu$ m, see Table 2 in Duan et al. (2016), which probably results in less-accurate breakage of the fluid stream into droplets and more likely collisions. The smallest diameter we used in this study (20.13  $\mu$ m) was chosen as a compromise to avoid these effects.

We plan to develop a special calibration target composed of dots of known diameters, similar to that applied by Kashdan et al. (2003) but in the size range below  $20 \,\mu\text{m}$ . Unlike the droplets, the position of the target in the sampling volume would be under control.

## Specific comments

1. Page 1 line 19: You mention that researchers have to tackle intrinsic difficulties when using in situ and remote sensing observations. You could consider giving some examples describing the main challenges of in situ and remote sensing observations.

We listed a few examples for each of the two approaches:

Researchers employing both strategies tackle with intrinsic difficulties. For instance, in-situ methods often face the dependence of the sample volume on particle size or air flow velocity, nonlinearity of Mie scattering intensity with respect to droplet size, aerodynamic effects related to the flow around or inside the instrument or aircraft, harsh conditions (incl. icing, wetting, temperature changes), necessity for handling large datasets or instantaneous data processing. Remote sensing provides the information with limited spatial resolution, hence microphysical properties represents only the average or integral over relatively large volumes which might be too simplistic to characterize inhomogeneous or multilayered cloud fields. On top of that, the retrievals are often dependent on the assumptions of specific size distribution or specific vertical structure of the atmosphere.

 Page 2 line 44: Holographic systems have also been applied on balloon-borne platforms; e.g. HoloBalloon Ramelli et al. 2020, https://amt.copernicus.org/articles/13/925/2020/

We included this study in the short review of cloud droplet sizing instruments given in the introduction. We also updated the citation concerning the HALOHolo instrument. Together with Schlenczek et al. (2017) who actually performed ground-based measurements, we cited Schlenczek (2017) (PhD thesis) and Lloyd et al. (2020).

3. Page 2 line 48: Here you compare the sampling volume and frame rate of holographic and shadowgraph instruments. I would recommend to compare the sample volume rate as this is more meaningful (e.g. 15 cm3\*5 fps = 75 cm3 s-1; 0.04 cm3\*400 fps = 16 cm3 s-1).

We agree, those values were mentioned in the revised text. In case of holography we used the value of 6 fps in this estimation as this is the true frame rate of e.g. HALOHolo and HoloGondel. Additionally, we cited the recent advancements with regard to the sample volume rate achieved in HoloBallon by Ramelli et al. (2020).

4. Page 2 line 50: You write that the shadowgraph technique did not gain common use in cloud measurements. This is misleading, as CPI is frequently used on aircrafts. Here are some examples: Lawson et al. 2001: https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/2000JD900789 Lawson et al. 2010: https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/2009JD013017 Stith et al. 2002: https://journals.ametsoc.org/jamc/article/41/2/97/16088 Woods et al. 2018: https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/2017JD028068

We did not formulate the point clearly. What we actually meant is that shadowgraphy is not common in cloud droplet measurements in comparison to other methods. Additionally, we cited the first from the papers listed because it describes the capabilities of the CPI, and the last one to give the most recent specification on the size range measured by this instrument.

The corrected sentence reads:

Nevertheless, despite both its simplicity and many insightful laboratory experiments, e.g. concerning droplet collisions (Bordás et al., 2013; Bewley et al., 2013), shadowgraphy is not the first choice method in cloud droplet measurements.

5. Page 7 line 146: In Fig. 1 the flow was vertically aligned. Did you use a different flow direction for the experiments? Please specify.

Fig. 1. illustrates the components of the setup. The photograph was taken during another short test performed with the same instruments but different mounting of the pipe. It is neither analyzed nor discussed in the paper. During the measurements described in the paper, the outflow from the pipe was horizontally aligned. Unfortunately, no picture of this particular configuration is available.

Appropriate explanation was added to the text in sec. 3.1:

The flow velocity was estimated to be of the order of  $10 \text{ cm s}^{-1}$  and the direction of the flow was aligned horizontally from left to right (i.e. the direction of the pipe exit was perpendicular to what is shown in Fig. 1).

6. Page 9 Fig. 4: On page 7 line 142 you state that the poly-dispersed water droplets in the stream were in the size range 2-20 μm. Is there a way to verify this? Why do you observe a lot of droplets larger than 20 μm in Fig. 4?

Unfortunately, we do not have another droplet sizing instrument. The given approximate range comes from the works of P. Korczyk and others (Korczyk, 2008; Korczyk et al., 2012) who used the same source of droplets, i.e. an ultrasonic humidifier. They measured the size spectrum by collecting sedimenting droplets on a glass plate covered with an oil film to prevent evaporation and analyzing the microscopic photographs. Despite the same source, the delivery system and ambient thermodynamic conditions are somewhat different in our case, which might result in small changes in the actual droplet size distributions due to possible collisions and evaporation. Note also that we collected much larger statistics (i.e. total number of counts) than was feasible with their method, hence very infrequent large droplets are better represented in our dataset.

We added the explanation in the text:

A dense stream of poly-disperse water droplets was generated with the use of an ultrasonic humidifier, the same as in the study of Korczyk et al. (2012) who measured the droplet size to be mostly in the range of  $2-20 \,\mu\text{m}$  in diameter (Fig. 1 there). Differences in delivery method and in ambient conditions could result in a little different spectrum.

7. Page 14 line 280: I only see the gradual decrease from the center to the sides for x1 in the horizontal direction. It seems like you have more particles on the left side compared to the right side (maximum is not at 0 µm). Is this relating to the non-uniformity of the plume or to instrumental flaws?

Our interpretation of the logarithmic plot given in the text was too simplistic. We changed the quantity presented in panels (a), (b) of Fig. 8 by normalizing  $N_V(x)$  or  $N_V(y)$  by the mean concentration to show the relative changes. After this correction it is clear the maximum is located left from the center. We suppose this might be related to the non-uniformity of the plume as well as the instrumental flaws. Therefore, we cannot claim the specific instrumental issues based on the results.

## The corrected description reads:

Figure 8 in panels (a) and (b) shows normalized droplet concentration  $N_V(x)/N_V$  and  $N_V(y)/N_V$  suitably integrated over size and over other dimensions, divided by the total concentration in order to highlight the relative dependence on position inside the sample volume. The values mostly decrease gradually from the maximum (located left from the center) to the sides in case of horizontal direction and from the bottom to the top in case of vertical. The relative differences, except from very close to the edges, are of the order of 10 %. They are small enough to be possibly caused by the non-uniformities of the plume.



Figure 8 corrected. Dependence of droplet concentration on (a) horizontal, (b) vertical and (c) axial position.

8. Page 14 line 284: You speculate that the decrease in concentration in vertical direction for x1 is due to instrumental reasons (e.g. non-uniform illumination). Can you verify/quantify that based on the images? The shadow images for x1 in Fig. 12 don't show indications for non-uniform illumination. Please comment on that.

The VisiSize software has an option of background normalization. The user can select one image which serves as a background and all the further frames collected in the measurement are divided by it. Such an option was used in the detection experiment described in sec. 3 (Fig. 3). We speculate the background normalization ensures proper performance of the thresholds ( $T_h$  and  $T_p$ ) but does not substantially improve the signal-to-noise ratio in originally darker (weakly illuminated) regions. There, only a small part of the grayscale levels (256) is used.

We cannot verify/quantify this issue directly for our detection experiment because in the course of long runs, the images are not saved but instantly processed to derive particle statistics (see the list of them in Table 2). If one prefers to keep the captured images, the duration of an uninterrupted measurement is limited by the available computer RAM which in our particular configuration meant about 2000 frames (corresponding to about 66 seconds).

In the series shown in Fig. 12 some gradient of brightness is visible in case of lens setting x1, for instance the right edge seems to be slightly darker than the center and the left. The orientation of the gradient is then different than in the detection experiment (sec. 3) which might result from manual position adjustment. We recall our observation that the satisfactory adjustment for this lens setting is rather difficult to achieve. Importantly, most of the droplets measured during the sizing experiment (sec. 4) were close to the central part of the SV. Therefore, our findings are not affected by the illumination non-uniformity.

9. Page 14 line 292: You explain that the axial dependence is more difficult to evaluate. Fig. 8.c shows a sharp decrease of the concentration with increasing z-distance, which you attribute to the miscounting of the smaller droplets. Would it be possible to perform a similar experiment as in Sect. 4 where you generate a mono-dispersed particle distribution? Or are the concentrations produced by the FMAG too small?

The concentrations produced by the FMAG are indeed much smaller  $(0.3-3 \text{ mm}^{-3})$  in comparison to the dense plume in the first experiment. We speculate the exact value depends non-trivially on the settings of the FMAG and the conditions on the way from the nozzle exit to the sample volume of the shadowgraph (flow properties, evaporation, collisions). Moreover, the generated stream of droplets is quite localized which implies non-random probing of the sample volume, including the axial direction. The essential component of the detection experiment with the dense plume (sec. 3) was the close-to-uniform filling of the SV. In our opinion, appropriate study of detection properties involving FMAG-generated droplets would require precise control of the position and orientation of the droplet stream with respect to the focal plane of the shadowgraph which seems to be challenging.

 Page 16 line 335: For the size calibration experiments, you produced droplet diameters of 20.13, 39.35 and 57.55 μm. Can you explain how these diameters were selected?

We intended to select 3 different sizes covering the range which is available with the particular FMAG generator at our disposal using UPW (17-69  $\mu$ m, Duan et al. (2016)). As explained in our reply to the general comment above, the smallest diameter (20.13  $\mu$ m) was chosen as a compromise in order to avoid spectrum broadening. Such effect is most probably related to the high vibration frequency which together with the liquid flow rate controls the output size.

We added a short explanation of the choice of the smallest size in the text:

The smallest diameter was chosen to ensure a relatively narrow spectrum as we observed significant broadening for high vibration frequency which is necessary to generate yet smaller sizes. Presumably, this results in less accurate breakage of the fluid stream or more frequent collisions.

11. Page 18 line 353: You state that the left-side skewness of the tails in Fig. 11 implies partial evaporation. I only see a left-skewed tail for x2 and x1 for the diameters 39.25 and 57.55 μm, but not for x4 and the smaller size (20.13 μm). I would expect that the evaporation effect is largest for smaller particles, but this does not seem to be the case. How can you explain this pattern?

In our opinion, the observation made by the reviewer might be related to the size of the sample volume which increases with effective pixel size (decreases with magnification) as well as with the particle size (i.e. from the top-left to the bottom-right panel in Fig. 12). Other processes which were not controlled in our experiment might have also contributed to the observed results, e.g. ambient air properties, velocity of the droplets or some interactions among them.

Our speculation was explained in the text:

Left-side skewness of the tails suggests partial evaporation. Although in general the effect of evaporation is expected to be more significant for small droplets, the skewness is evident for 39.25 and 57.55  $\mu$ m measured with the lens settings x1 and x2. We speculate it might be related to the size of the sample volume which increases with the effective pixel size (decreases with magnification) as well as with the particle size (i.e. from the top-left to the bottom-right panel in Fig. 12). The position of the nozzle exit was adjusted so that the

center of the FMAG-generated droplet stream is as close as possible to the focal plane of the shadowgraph. We expect the droplets more distant from the central axis of the stream to be more likely partially evaporated because of the longer travel and exposure to the dry air blown from the area around the nozzle. Those can be detected in case of considerable sample volume but not in case of smaller SV. Importantly, this is only one of the effects which could have contributed to the observed result together with the ambient air properties, velocity of the droplets or some interactions among them.

- 12. Page 21 Fig. 14: Why do you have multiple times the same symbol in a, b and c? Are these different experiments? Yes, there were multiple measurement rounds performed with the same settings. Each symbol represents the mean diameter or the 1st-peak diameter obtained in a single measurement. The caption of the figure was updated accordingly.
- 13. Page 22 Fig. 15: Were other cloud probes deployed at the measurement site? On page 20 line 381 you say "After laboratory tests, the shadowgraph VisiSize D30 has been used for the first time to measure droplets in atmospheric clouds [...] to compare it with other probes already in service in cloud physics studies [...]". If other cloud probes were deployed at the same time, I would add the size distribution of additional probes in Fig. 15 for validation of the VisiSize D30 shadowgraph system. Alternatively, I would recommend performing a comparison campaign with other cloud probes in the future.

During two observational periods in July and August 2019, the size distribution was measured also by the Phase Doppler Interferometer (PDI). The two instruments, intentionally located side-by-side were not simultaneously operational in all the runs but we have data from overlapping periods. This data is still subject to processing and detailed analysis. In case of the PDI, the sample volume depends non-trivially not only on droplet size but also on the wind velocity, highly variable in the turbulent experimental conditions. Therefore, we are working on proper systematic comparison which considers those issues and shows some limitations of ground-based measurements with the instruments developed for high true air speeds . We intend to describe the details and submit it to the AMT as soon as the analysis and interpretation is finished.

## **Technical comments**

- 1. Page 5 line 93: I would suggest to write "[...] a method compensating for the effects[...]"
- 2. Page 6 Fig. 2: Consider choosing different colors for Th and Tp for better distinction.
- 3. Page 6 line 120: 'z' should be written in italic.
- 4. Page 8 line 165: 'z' should be written in italic.
- 5. Page 8 line 167: 'z' should be written in italic.
- 6. Page 15 line 308: capitalize 'D' in "2-dimensional (z, D) maps"

7. Page 19 Fig. 12: Consider adding different lines for single droplet/double collision/triple collision similar as in Figure 13 or at least of the FMAG diameter.

We agree with the technical comments and applied the specific corrections according to the reviewer's suggestions.

### **Response to the Anonymous Referee #2**

## **General comments**

Would it be possible to use the shadowgraph system to obtain some 3D information about the position of the cloud droplets?

Unfortunately, the diameter-dependent sample volume is generally too small to obtain the information about the 3D arrangement of the droplets in a typical atmospheric cloud. For instance, in Fig. 5. one can see the DOF for the droplet of a diameter 20  $\mu$ m is about 200  $\mu$ m (twice the  $z_{max}$ ). Multiplied by the FOV (see Table 1) it results in the SV of about 5.90, 1.50 and 0.35 mm<sup>3</sup>, for magnifications x1, x2 and x4, respectively. Even if the concentration was 1000 cm<sup>-3</sup> and all the droplets had 20  $\mu$ m, we would expect on average around 5.9, 1.5 or 0.35 counts per frame, respectively. For smaller diameters, the DOF and hence the SV as well as the expected counts per frame are accordingly much smaller. For the field measurement described in sec. 5, the concentrations were estimated to about 928 cm<sup>-3</sup> and 798 cm<sup>-3</sup>. During the first round with magnification x2, there were on average only 0.7 counts per frame. During the second round with magnification x4, there were on average only 0.08 counts per frame.

Another issue might be the shape of the SV resembling more a slice than a cube as the z-dimension is much smaller than x and y. On top of that, only the absolute value of z coordinate of a droplet is estimated. It is unknown, on which side of the focal plane the object is.

Could you give any estimate for the largest detectable number concentration? In case of large number concentrations, for example, it might be possible that droplets "hide" one after the other.

Due to the small slice-shaped SV, the problem of the largest detectable concentration is not relevant for typical atmospheric clouds. Similarly to the previous question, we may consider mono-dispersed droplets of a diameter 20  $\mu$ m and concentration 1000 cm<sup>-3</sup>. Then, the expected number in the single cylinder of a diameter 40  $\mu$ m and the height equal to the DOF (around 200  $\mu$ m) is only 2.5  $\cdot 10^{-4}$ . For smaller droplets, it would be accordingly smaller. Therefore, the coincidence of two droplets at similar (*x*, *y*) position seems highly improbable.

In the field measurements, there was on average even below 1 count per frame. The concentration in the plume used during the laboratory experiment was about two orders of magnitude larger which gave 18-54 counts per frame (depending on magnification). In the limited number of the inspected test images we did not spot coincidences. However, in the course of the longer

measurement the images are not stored but processed in real time (see sec. 2.1) which makes the problem of coincidences hardly possible to verify afterwards.

On the other hand, what we observed during the laboratory experiment is that the dense plume filling the considerable space between the camera and the laser but outside the SV can influence the average brightness of the image. The thresholds implemented in the software adapt to the average brightness but still such shadow diminishes signal-to-noise ratio of individual particles as discussed for the magnification x1 in sec. 3.5. In our opinion, the effect is not significant in atmospheric clouds due to smaller concentration, hence limited scattering.

Concerning the application of the shadowgraph system in the atmospheric cloud (Section 5): Has there been any other cloud probe which measured the size distribution at the same time as the shadowgraph system? If so, could you present a comparison of the obtained droplet size distributions and its moments, respectively? If not, I would suggest, for a future study, to have a comparison to another cloud probe.

During two observational periods in July and August 2019, the size distribution was measured also by the Phase Doppler Interferometer (PDI). The two instruments, intentionally located side-by-side were not simultaneously operational in all the runs but we have data from overlapping periods. This data is still subject to processing and detailed analysis. In case of the PDI, the sample volume depends non-trivially not only on droplet size but also on the wind velocity, highly variable in the turbulent experimental conditions. Therefore, we are working on proper systematic comparison which considers those issues and shows some limitations of ground-base measurements with the instruments developed for high true air speeds. We intend to describe the details and submit it to the AMT as soon as the analysis and interpretation is finished.

## Specific comments

1. Page 7, line 145-146: Looking on Fig. 1, I would think that the direction of the droplet flow was vertically aligned.

Fig. 1. illustrates the components of the setup. The photograph was taken during another short test performed with the same instruments but different mounting of the pipe. It is neither analyzed nor discussed in the paper. During the measurements described in the paper, the outflow from the pipe was horizontally aligned. Unfortunately, no picture of this particular configuration is available.

Appropriate explanation was added to the text in sec. 3.1:

The flow velocity was estimated to be of the order of  $10 \text{ cm s}^{-1}$  and the direction of the flow was aligned horizontally from left to right (i.e. the direction of the pipe exit was perpendicular to what is shown in Fig. 1).

2. Fig. 5c: For diameters larger than 30  $\mu$ m,  $z_{max}$  deviates significantly from the analytical approximation  $z_{95}$ . Do you have any explanation for that?

The discrepancy relates to the poor statistics of large droplets. Each point in the plot represents one diameter bin and shows the distance from the focal plane corresponding to the furthest detected droplet from that bin. Diameters larger

than 30  $\mu$ m are very scarce in the plume (see the size distribution in Fig. 7), so the number of detections is also small and they do not fill equally the entire sample volume. Therefore, it can happen that all of them randomly appear at the positions closer to the focal plane than the maximum possible distance  $z_{95}$ . For instance, in the case of the measurement with the magnification x4 presented in Fig. 5c, there were only 3 counts in the last bin and only 15 in the one before last. The sample volume decreases strongly with magnification implying the limited chance of detecting infrequent large droplets which explains why the effect is most pronounced for x4.

We added a short comment to the text:

The discrepancy observed for largest sizes is related to the poor statistics, i.e. small number of counts of large droplets which are infrequent in the measured plume.

3. Fig. 7 and Fig. 15 and the corresponding text: In Fig. 7 only DSDs applying methods"def" and "ind" are shown, for given reasons. However, later in Fig. 15, DSDs for "def", "cor" and "ind" are shown. Wouldn't it make more sense, to show DSDs for methods"def", "cor" and "ind" in Fig. 7 and explain that there are only small differences between methods "cor" and "ind" and then only show DSDs for "def" and "ind" in Fig. 15?

We added the missing method to Fig. 7 and decided to keep all of them in Fig. 15 for consistency. The caption of Fig. 7 was modified accordingly.

4. Page 14, line 280: This gradual decrease from the center to the sides is obvious for x1, but not really for x2 and x4. Could you please comment on that?

Our interpretation of the logarithmic plot given in the text was too simplistic. We changed the quantity presented in panels (a), (b) of Fig. 8 by normalizing  $N_V(x)$  or  $N_V(y)$  by the mean concentration to show the relative changes. After this correction it is clear the maximum is located left from the center.



Figure 8 corrected. Dependence of droplet concentration on (a) horizontal, (b) vertical and (c) axial position.

## The corrected description reads:

Figure 8 in panels (a) and (b) shows normalized droplet concentration  $N_V(x)/N_V$  and  $N_V(y)/N_V$  suitably integrated over size and over other dimensions, divided by the total concentration in order to highlight the relative dependence on position inside the sample volume. The values mostly decrease gradually from the maximum (located left from the center) to the sides in case of horizontal direction and from the bottom to the top in case of vertical. The relative differences, except from very close to the edges, are of the order of 10 %. They are small enough to be possibly caused by the non-uniformities of the plume.

5. Page 14, line 284-291: I think the speculation given here is reasonable. However, could this also be true for x2 and x4? But here more (smaller) droplets are detected, both in a smaller FOV compared to x1 which compensates this feature?

As far as we understand, the reviewer suggests the suspected y-dependent concentration concerns large droplets regardless of the magnification setting used. This effect is then supposedly not clearly visible for x2 and x4 because of the dominant number of small droplets detected in a more uniform manner.

We verified this claim to be true. The concentration  $N_V(y)$  of the droplets larger than 12 µm only is indeed more ydependent than the same quantity for the droplets smaller than 12 µm, in case of both magnifications x2 and x4. We cannot exclude the contribution of gravity sorting to that effect. However, the relative decrease of the  $N_V(y)$  of large droplets from the bottom to the top is significantly smaller than over the same distance in case of the magnification x1.

The exact influence of the non-uniform illumination on the results of DSD and DNC is hardly possible to be estimated with the available data. We emphasize the importance of precise alignment of the laser and camera lens positions. This is done manually, so the positioning can differ slightly between the experiments. As stated in the paper, achieving close to uniform illumination is much easier for magnifications x2 and x4 than x1 because of the smaller FOV to be fitted into the same laser beam.

## We modified the text:

The y-dependence of the concentration is more pronounced for large droplets above 12  $\mu$ m (not shown) which might be the influence of gravity sorting. However, in case of the series recorded with lens setting x1, the concentration falls with height y by a factor of more than 10 from the bottom to the top of the FOV. We speculate this effect is of instrumental origin, as the difference is rather too large (almost exponential) and the timescale too short to allow for the explanation only by gravity sorting of the droplets in the plume.

 Page 15, Line 318: Could you please provide a size range where you would not use x1. As later said, x1 makes sense for larger droplets in the drizzle and rain size range. Please make clearer under which circumstances you would avoid using x1, and vice versa.

We estimated the minimum diameter for uniform detection to  $12 \ \mu m$  so the magnification x1 is not recommended to measure any droplets smaller than that. Another limit, related to the issue of illumination, is not so precise because the image quality depends to some extent on the accuracy of manual alignment. In order to avoid poor signal-to-noise ratio

we would suggest, as a rule of thumb, a diameter an order of magnitude larger than the effective pixel size which results in around 40  $\mu$ m. On the other hand, the objects need to fit into the FOV, so the largest measured size should be a few times smaller than the shorter dimension (4.1 mm). Eventually, we expect the setting x1 to perform well in the range of roughly 40-400  $\mu$ m. So far, we have not collected much data on drizzle to confirm this expectation experimentally. Nevertheless, in the sizing experiment (sec. 4) we had indeed no issues for 39.35  $\mu$ m droplets but still some inaccuracies for 20.13  $\mu$ m.

We included this information in the text:

As a consequence, the authors discourage using lens setting x1 in further studies of cloud droplets. The large minimum particle size for uniform detection reported earlier ( $\sim$ 12 µm) makes this option of limited utility anyway. Instead, x1 can possibly serve well for the measurements of drizzle drops. In order to avoid the illumination and signal-to-noise issue described above, we would suggest, as a rule of thumb, the lower limit an order of magnitude larger than the effective pixel size (3.69 µm). On the other hand, the objects need to fit into the FOV, so the largest measured size should be a few times smaller than the shorter dimension (4.1 mm) which eventually results in the conservative range of roughly 40-400 µm. Notwithstanding, so far insufficient data on drizzle has been collected to confirm this expectation experimentally.

7. Fig. 9 and respective text: Does the discrepancy in the first and second z-bin has any consequences on the calculation of the DSD and DNC?

We consider the question relevant however cannot estimate the effect quantitatively based on the available data. In order to investigate this issue we increased the number of z-bins from 20 to 80 which implied the changes in Fig. 8c and Fig. 9. This operation allowed to identify a lower limit on the distance z and the probable reason for the discrepancy observed earlier - diffraction around the objects close to the focal plane. In the paragraphs added to the text we discuss the mechanism and speculate the influence on the DSD and the DNC within the valid size range is rather minor. In our opinion, one solution to systematically eliminate the problem would be to add the term representing diffraction to Eq. (1). This requires repeating the calibration procedure to obtain the correct new values of  $a_1$  and  $a_2$ . At best, also implementing the corrected equation in the software processing routine.

We corrected the interpretation of Fig. 8c:

Interestingly, the first z ranges contain a much smaller number of droplets than the maximum located further from the focal plane, regardless of the lens setting.

We added the paragraphs discussing the issue:

On the other side, the minimum distance  $z_{min}$  for a given diameter is well approximated by assuming the halo area in the focal plane equal to the diffraction term ( $A_h = \pi D a_5$ ), analogously to Fig. 4, and solving Eq. (1) for z (blue line).



Figure 9 corrected. Variability of droplet detection properties in size-space domain for lens setting (a) x1, (b) x2, (c) x4.

(...)

In experimental runs with higher magnifications, i.e. lens setting x2 and x4, the (z, D) map features the decrease of concentration with z from the maximum located a bit above  $z_{min}$ . The same effect was noticed in Fig. 8 panel (c). Most probably, it relates to the diffraction which is not included in the modeled dependence of the halo area  $A_h$  on the diameter D and distance z in Eq. (1). Namely, the equation is not correct in the limit of small z because it implies the object in the focal plane (z = 0) should be ideally sharp  $(A_h = 0)$ . Consequently, in this limit the calculated z position is overestimated with respect to the true one. The counts representing droplets standing very close to the focal plane are shifted to the further z-bins in Fig. 9. The extent of this shift is probably not constant but decreases with the true z. Hence, the counts cumulate at some point above  $z_{min}$  creating a maximum in  $N_V(z)$ . We expect the shift to decrease because the calibration constants  $a_1$  and  $a_2$  were fitted by the manufacturer in the procedure resembling Kashdan et al. (2003) so that the Eq. (1) performs satisfactorily in the range of defocus distances and particle diameters typical for industrious applications, i.e. a bit larger than analysed here. Therefore, the estimated z should approach the true one with increasing defocus and droplet diameter.

The shift of the estimated z positions with respect to the true ones is most pronounced in case of the small distances from the focal plane. Importantly, it should have no effect on the accuracy of the sample volume calculation, hence the DNC and the DSD, as long as the  $z_{95}$  is not overestimated but represents the true distance at which the droplets are no longer counted. We expect this condition to be met if the largest possible defocus  $z_{95}$  is significantly higher than the smallest  $z_{min}$ . For instance, they differ by a factor of two for diameters larger than 8.0, 6.3, 4.8 µm in case of lens settings x1, x2 and x4, respectively. Those values are close to the minimum diameter for uniform detection estimated in sec. 3.4. It implies the influence on the DSD and the DNC within the valid size range is probably minor. However, we cannot quantify it with high confidence based on the available data.

## **Minor comments**

- 1. Page 2, Line 50: I would suggest to write either "shadowgraphy" or "the shadowgraph technique".
- 2. Page 3, line 70: Please add "the" at the beginning of the sentence: "The two main parts[...]".
- 3. Page 6, line 120: Since "z" is a parameter it should be given in italic type font.
- 4. Page 7, line 137: Please write "[...] inverting Eqs. (1) and (2)".
- 5. Page 7, line 142: It should read "[...] in the range of 2-20 µm [...]".
- 6. Page 7, line 143-144: I would suggest to write: "Care was taken to fill [...]".
- 7. Page 8, line 169: Please add "DOF" in the brackets behind "(depth of field)".
- 8. Fig. 14: I would suggest increasing the size of the symbols in the figures here.

We agree with the minor comments and applied the specific corrections according to the reviewer's suggestions.

## Additional corrections

While revising the manuscript, we found a mistake in Eqs. (6), (7) and (8). As the text explains, the effective cross-sectional area of camera sensor, FOV, is reduced by the margin of the width equal to a diameter under consideration. In the equations, we wrongly used 2D instead of D, confusing diameter with radius. This term was corrected. The change concerns the equations in the manuscript and the captions in Fig. 6. All the calculations were performed correctly.

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# Applicability of the VisiSize D30 shadowgraph system for cloud microphysical measurements

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Abstract. The commercial shadowgraph system, Oxford Lasers VisiSize D30, originally designed to characterize industrial and agricultural sprays, was tested with respect to the application for measuring cloud microphysical properties, such as droplet size distribution and number concentration. Laboratory experiment with a dense stream of poly-disperse cloud-like droplets indicated strong dependence of the depth of field, thus also sample volume, on particle size. This relationship was determined and

- 5 a suitable correction method was developed to improve estimations of droplet number concentration and size distribution. Spatial homogeneity of detection probability inside the sample volume and minimum droplet diameter providing uniform detection were examined. The second experiment with mono-disperse droplets produced by Flow-Focusing Monosized Aerosol Generator (FMAG) verified sizing accuracy and demonstrated reasonable agreement between the instruments. Effects of collisions and evaporation of droplets produced by FMAG were observed. Finally, the instrument was applied to sample atmospheric clouds
- 10 at a ground-based mountain observatory and performed reliably during 3 week long field experiment. Based on the laboratory and field tests, recommendations concerning the use of the instrument for cloud droplet measurements were formulated.

## 1 Introduction

Atmospheric clouds predominantly consist of water droplets. Cloud droplet number concentration (DNC) and size distribution (DSD) constitute the key parameters for quantitative microphysical description of clouds and attract enormous attention in contemporary atmospheric sciences, mostly due to their crucial importance for cloud lifetime, radiative effects and rain formation

(Devenish et al., 2012).

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There exist two general approaches to measuring cloud microphysical properties: in situ sampling from airborne platforms or at ground-base ground-based stations and remote sensing which involves applying inverse retrieval techniques to data collected by satellites, radars and radiometers. Researchers employing both strategies tackle with intrinsic difficulties. HoweverFor

20 instance, in situ methods often face the dependence of the sample volume on particle size or air flow velocity, nonlinearity of the Mie scattering intensity with respect to droplet size, aerodynamic effects related to the flow around or inside the instrument or aircraft, harsh conditions (incl. icing, wetting, temperature changes), necessity for handling large datasets or instantaneous data processing. Remote sensing provides the information with limited spatial resolution, hence microphysical properties represents only the average or integral over relatively large volumes which might be too simplistic to characterize inhomogeneous or 25 multi-layered cloud fields. On top of that, the retrievals are often dependent on the assumptions of specific size distribution or specific vertical structure of the atmosphere. In general, in situ measurements are considered fundamental, as they offer instrumental access to individual droplets within a sampling volume. The results obtained in situ are then used to derive and validate inversion routines to be used in remote sensing applications.

Among in situ techniques, one can distinguish two branches differing in sampling style:

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- instruments detecting and counting droplets one-by-one but almost continuously in time as they pass through a very small active probe volume provide individual droplet properties and their inter-arrival times,
  - instruments capturing images or other spatial representation of droplets inside a larger sampling volume provide individual droplet properties and information on their spatial arrangement.
- The first branch is represented by a number of spectrometers using light scattering for droplet detection and sizing, e.g. For-35 ward Scattering Spectrometer Probe (FSSP, e.g. Cooper (1988); Brenguier et al. (1998); Gerber et al. (1999); de Araújo Coelho et al. (2005)), Cloud Droplet Probe (CDP, e.g. McFarquhar et al. (2007); Lance et al. (2010); Lance (2012)), Cloud and Aerosol Spectrometer (CAS, e.g. Lance (2012); Glen and Brooks (2013); Barone et al. (2019)), Phase Doppler Interferometer (PDI, e.g. Bachalo and Houser (1984); Chuang et al. (2008); Kumar et al. (2019)). In practice, they sample quasi one-dimensional portion of air passing through the region of active sampling. The exact volume sampled in unit time depends on the velocity
- 40 with respect to the instrument. Therefore, meaningful estimation of DNC requires information on air velocity. Moreover, due to small probe volume cross-section, there exist an upper limit for measurable droplet size. For instance, the range of diameters of the CDP equals 2-50 µm.

Volumetric methods (second branch) usually do not rely on scattering intensity of individual droplet in sizing and their sampling volumes do not depend on the velocity of the flow but consecutive air samples, collected instantaneously, might be

- 45 quite distant from each other, in particular when used on fast moving airborne platforms. This class of techniques is represented e.g. by shadowgraphy and holography. Neglecting the effect of imperfect focusing, shadow image constitutes two-dimensional projection of all the objects onto camera plane and can be rapidly processed to detect particles and obtain relevant statistics. Holograms require extensive processing to digitally reconstruct objects' shape and arrangement in three dimensions (Fugal et al., 2009). Both methods allow to study position, size and shape, thus not only spherical droplets but also e.g. ice crystals.
- 50 Holographic systems have been successfully deployed on research aircrafts (HOLODEC, Fugal and Shaw (2009)and HALOHOLO, Schlenczek et al. (2017)), mountain, and HALOHolo, Schlenczek et al. (2017), Schlenczek (2017), Lloyd et al. (2020)), ground-based mountain observatories (HOLIMO, Henneberger et al. (2013)), mountain cable cars (HOLOGONDELHoloGondel, Beck et al. (2017)) and ground-based mountain observatories (HOLIMO, Henneberger et al. (2013)balloon-borne platforms (HoloBalloon, Ramelli et al. (2020)). Typically, those instruments have resolution of 6 µm, sample volume (SV) of about 15 cm<sup>3</sup>, resolution
- 55 of 6-and take a few holograms per second which results in the sample volume rate (SVR) of around 90 cm<sup>3</sup> s<sup>-1</sup>. The recent HoloBallon setup features SV of 22.5 cm<sup>3</sup> and frame rate of 80 fps which gives SVR of 1800 cm<sup>3</sup> s<sup>-1</sup>. Shadowgraphy has been used e.g. in Cloud Particle Imager (CPI, Connolly et al. (2007)) Lawson et al. (2001), Connolly et al. (2007)), an airborne instrument to observe ice particles and supercooled droplets in the size range 10-2300 of 2.3-2300 µm (Woods et al., 2018). Typi-

cal SV is much smaller than in holography (about  $0.04 \text{ cm}^3$ ), however the frames rate is much higher (400 fps) which gives the

- SVR of about 16  $\text{cm}^3 \text{s}^{-1}$ . Moreover, Rydblom and Thörnberg (2016) have designed a system to investigate icing conditions 60 for wind turbines based on shadow images. Nevertheless, still shadowgraph did not gain common use in cloud measurements despite both its simplicity and many insightful laboratory experiments, e.g. concerning droplet collisions (Bordás et al., 2013; Bewley et al., 2013), shadowgraphy is not the first choice method in cloud droplet measurements.
- In order to explore in detail the advantages and disadvantages of shadowgraphy for cloud microphysical applications, we use the commercial shadowgraph system (VisiSize D30, Oxford Lasers Ltd., Kashdan et al. (2003, 2004)), originally designed 65 for diagnosis of agricultural and industrial sprays, to measure DNC and DSD in warm clouds. Within the study, two series of laboratory experiments were performed which aimed at verifying reliability of detection and accuracy of sizing under conditions resembling atmospheric clouds. They were followed by a field experiment targeting real cloud at a mountaintop observatory.
- 70 The present paper is structured as follows. Section 2 describes the instrument and the measurement principle. Section 3 provides the analysis of detection reliability and homogeneity affecting results on DNC and DSD, investigated with the use of poly-disperse water droplets. Section 4 focuses on sizing accuracy studied with the use of mono-disperse droplet population. Based on the experiments, corrections to the standard algorithm implemented in instrument software are suggested. Finally, in section 5 we present selected results obtained during the first application of the instrument in atmospheric clouds. The last
- section summarizes the findings and discusses the conclusions concerning the further usage of the VisiSize D30 system for 75 cloud research.

#### 2 System overview

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VisiSize D30 is a complete shadowgraph system manufactured by Oxford Lasers Ltd. (Oxon, United Kingdom) designed to characterize particles in various suspensions. Common industrial applications include among others the characterization of: agricultural sprays, paint sprays, consumer aerosols, fire extinguishers and automotive fuel injectors.

#### 2.1 Hardware description

Two The two main parts of the set-up are infrared diode pulse laser with diffuser and digital camera with suitable lens objective (see Fig. 1). Lens magnification can be changed manually to adjust the resolution and extent of the sample volume to the object of study. For three selected options, instrument calibration was performed by the supplier. Capabilities of the system at those settings are listed in Table 1.

The operation of the system is the following. The region of interest is illuminated from behind by diffused (incoherent) expanded laser light beam, then shadow images of droplets are collected at up to 30 frames or pairs of frames per second with a digital camera. The laser and the camera are jointly triggered so that a single laser pulse "freezes" the motion of droplets present within the measurement volume during each frame capture. Droplets detected inside the depth of field (DOF) are then

measured based on their shadow images, and statistics regarding concentration or size distribution are built by processing a 90

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**Figure 1.** Experimental setup for studying detection properties (see sec. 3 for details). Main parts of the VisiSize D30 are: infrared pulse laser with a diffuser (top-right), CCD camera with lens objective enclosed in a water-proof housing (top-left). Water droplets produced with an ultrasonic droplet generator are measured while passing through the sample volume (top-middle) located close to the camera.

Table 1. Properties of the VisiSize D30 system for three different lens magnification settings provided by Oxford Lasers Ltd.

VisiSize D30 specifications							
Camera chip [pix x pix]		1952 x 1112					
Camera pixel [µm]		5.5					
Frame rate [fps]		30					
Wavelength [nm]		808					
Laser pulse duration [µs]	0.1 - 5.0						
Lens setting	x1	x2	x4				
Magnification	1.49	2.97	6.12				
Effective pixel size [µm]	3.69	1.85	0.90				
Resolution [µm]	6.9	3.7	2.0				
Field of view [mm x mm]	7.20 x 4.10	3.62 x 2.06	1.75 x 1.00				
Depth of field [mm]	50.0	16.6	5.2				
Sample volume [cm <sup>3</sup> ]	1.48	0.123	0.0092				
Volume rate $[\text{cm}^3 \text{ s}^{-1}]$	44.3 3.71 0.28						

series of images. Worth to mention, there is also an option to measure droplet velocity in the imaging plane by comparing pairs of consecutive frames and measuring droplet displacement between them. The captured images are either processed in real-time to determine particle positions and sizes (live mode) or stored as graphic files (capture mode). In case of the former,

the output is only a list of particles together with their properties (droplet file). Specific quantities included in the droplet file
are explained in Table 2. The second complementary output file contains system settings used and measurement summary with some basic statistics of the recorded droplet set (summary file). For the capture mode, one can analyse captured images later tuning some parameters or access the raw genuine view of the particles (see Fig. 3 in sec. 3.1). However, then the total time of uninterrupted measurement is limited by available computer RAM.

Table 2. Explanation of the parameters reported in output droplet file together with corresponding symbols used to denote them in the text.

Parameter	Symbol	Units	Description
Frame	j	-	Number of the image containing current particle
Particle ID	i	-	Particle number, unique across dataset
Area	Α	$\mu m^2$	Estimated cross-sectional area, calculated from $D$
Diameter	D	μm	Estimated particle diameter, calculated based on $A_p$ and $A_h$
Shape Factor	$S_f$	-	Measure of sphericity equal to the ratio of the principle moments of inertia of
			the shadow, taking values from 0 to 1, with 1 representing a perfect circle
X	x	pix	Horizontal position in camera plane
Y	y	pix	Vertical position in camera plane
Pixel Area	$A_p$	pix <sup>2</sup>	Number of pixels in particle shadow with brightness below threshold $T_p$
Pixel Halo	$A_h$	pix <sup>2</sup>	Number of pixels in particle shadow with brightness between thresholds $T_h$ and
			$T_p$ where $T_h < T_p$
Distance to Focal	z	μm	Estimated axial position in the sample volume, calculated based on $A_p$ and $A_h$ ;
Plane			insensitive to direction, thus always positive

## 2.2 Principle of droplet detection and sizing

- 100 The measurement principle applied in VisiSize system was explained by Kashdan et al. (2003, 2004). It stems from the basic observation that with increasing defocus (particle displacement out of the focal plane of the camera lens) the image of the object is more and more blurred which hinders the proper estimation of the size of the shadow. On the other hand, the range of axial positions guaranteeing acceptable sharpness of the shadow is usually quite limited. As a consequence, the probability of detecting particle inside such restricted SV might be often insufficient to collect meaningful statistics of the suspension in
- 105 reasonable time. To overcome difficulties described above, Kashdan et al. (2003) applied the a method compensating for the effects of imperfect focus. Basically, the displacement from the focal plane is estimated from the degree of image blurring, specifically the gradient of brightness at the edge of inner dark shadow. In their implementation, two threshold limits are determined for each analysed picture based on the histogram of pixel brightness. Both of them lie between the background intensity and the level corresponding to dark centers of particle shadows (see Fig. 2). The upper threshold  $T_p$  separates the
- 110 background from the particle image. All pixels below this value are assumed to belong to effective total particle image area

 $A_p$  (with the equivalent diameter  $D_p$ ). The lower threshold  $T_h$  distinguishes between the dark shadow interior and an outer gray "halo". Pixel with intensities between  $T_h$  and  $T_p$  are counted to produce the estimate of particle halo area  $A_h$  (with the equivalent width  $D_h$ ). Therefore,  $A_h$  belongs to  $A_p$  and has to be smaller.



**Figure 2.** Schematic representation of threshold method showing shadow image of two sample droplets, in-focus droplet (left) and defocused droplet (right), alongside grayscale intensity profiles below them illustrating applied thresholds.

With increasing defocus distance, the total area of particle image grows due to blurring until at some point it fades away 115 into background making the object no longer distinguishable. Worth to mention, halo area  $A_h$  grows faster than total area  $A_p$ because it extends both into outer and inner direction, taking over respectively the background and the interior pixels. Ideally, for object standing exactly in the focal plane  $A_h$  should tend to zero. However, it is never the case due to intrinsic diffraction caused by finite aperture of the lens. This effect might be particularly important for small object of the size close to the optical resolution of the system.

Particle total area A<sub>p</sub> and halo area A<sub>h</sub> can be regarded as directly measured quantities. Both true particle diameter D and estimated defocus distance z are derived from them. The exact conversion is determined with experimental calibration, separately for each lens setting. The pictures of calibration targets of known sizes are taken at known distances away from the focal plane. Applying the sizing algorithm to the collected data, some function is fitted to approximate the relationship A<sub>p</sub>(D, z) and A<sub>h</sub>(D, z). For the present VisiSize D30 model, calibration was performed by Oxford Lasers for three possible lens magnification settings (x1, x2 and x4) and incorporated into the software.

In principle, for perfect focusing the halo area  $A_h$  should tend to zero, since then the image consists only of a dark disk with sharp edge around. Certainly  $A_h$  grows with the distance between actual particle position and the focal plane. The halo is formed exactly around the dark interior of the shadow, i.e. each point on an initially very sharp edge spreads equally in all directions, smoothing the intensity gradient and creating halo of area which scales with the perimeter length and the amount

130 of defocus. For objects with shape close to a circle, the perimeter is proportional to the diameter which leads to a scaling

 $A_h \sim Dz$ . However, even a point source, while being out of the focal plane is mapped to a circle of confusion in image plane (camera chip). Therefore  $A_h$  should be growing function of z - z even in case of size D tending to zero. The simple linear relation satisfying the above properties is:

$$A_h = a_1 D z + a_2 z \tag{1}$$

135 where  $a_1$  and  $a_2$  are calibration constants for a given lens magnification.

As described above, with increasing defocus distance z, halo around the dark interior develops at the cost of both the background pixels and the inner shadow pixels. The whole  $A_h$  is included in total particle pixel area  $A_p$ , so only the outward growth affects the value of  $A_p$ . Outward part of the shell is larger than inward one, hence the proportionality factor would be close to but slightly higher than 0.5. Obviously, in focal plane the image area is dependent on the true cross-section of the physical object, but  $A_p$  is composed of pixels of finite size which disturbs the perfect representation of smooth shapes. Consequently, effective diameter length might be diminished by the amount of the order of pixel size. Moreover, even for the focal plane diffraction comes into play and hinders perfect imaging - objects always appear slightly larger than in reality which is important especially for small droplets. The enlargement of diameter due to diffraction should be of the order of the optical resolution of the lens system. All the effects can be taken into account in the formula of the form:

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$$A_p = a_3 A_h + \frac{\pi}{4} \frac{(D + a_4 + a_5)^2}{\text{pix}^2}$$
 (2)

where the constants  $a_3$ ,  $a_4$ ,  $a_5$  and pix describe halo blurring, pixelization, diffraction and effective pixel size, respectively. Note that  $A_p$  is specified as the number of pixels, hence the unit conversion factor pix<sup>2</sup> is needed. Having the values of  $A_h$  and  $A_p$  obtained from the image of the specific particle, both the diameter D and the defocus distance z can be calculated by inverting the EqEgs. (1) and (2).

## 150 **3 Detection properties**

## 3.1 Diagnostic experiment

First diagnostic laboratory experiment was carried out to characterize instrument performance in terms of detection probability and homogeneity which affect statistics of DNC and DSD. A dense stream of poly-disperse water droplets (known-was generated with the use of an ultrasonic humidifier, the same as in the study of Korczyk et al. (2012) who measured the droplet

- 155 size to be mostly in the range of 2-20 μm in diameter , Korczyk et al. (2012))was generated with the use of an ultrasonic humidifier. The (Fig. 1 there). Differences in delivery method and in ambient conditions could result in a little different spectrum. In our setup, the cloud of droplets was delivered from the humidifier into the SV with 4 cm wide, 70 cm long circular plastic pipe(see Fig. 1). Care was exercised. Care was taken to fill the whole shadowgraph SV with the stream of droplets though both the flow and the particle field were not exactly homogeneous. The flow velocity was estimated to be of the
- order of  $10 \text{ cm s}^{-1}$  and the direction of the flow was aligned horizontally from left to right  $\frac{1}{2}$  (i.e. the direction of the pipe exit was perpendicular to what is shown in Fig. 1). For each of the three lens settings (x1, x2, x4) laser power was adjusted to reach

optimal background brightness of the pictures and 10 min long measurement in live mode was performed. Figure 3 presents an example image captured during the experiment. Statistics reported by the software for each run are listed in Table 3.

Lens setting	x1	x2	x4	
Video frames	17705	17670	17695	
Empty frames	3390	4508	8156	
Total counts	602232	951642	324625	
Counts/frame	34.0	53.9	18.3	
Min. diameter $[\mu m]$	7.9	4.6	3.2	

Table 3. Statistics of the laboratory experiment with poly-disperse droplets for different lens magnification settings.



**Figure 3.** A typical shadow image of droplets produced by ultrasonic droplet generator taken during laboratory tests with the camera lens magnification x2.

## 3.2 Focus rejection and depth of field

165 Results of the experiment show that both halo  $A_h$  and total particle area  $A_p$  cannot take any values but their admissible range is limited by certain conditions (see Fig. 4). At sufficiently large distance away from the focal plane particle image starts to fade away into background making it no longer distinguishable. Simultaneously, halo takes over almost whole particle image. To avoid measuring objects whose signal-to-noise ratio does not allow for proper sizing, a simple rejection limit is exercised Importantly, deactivation of this option is not available from the level of the user in the software version 6.5.39.

The condition poses an upper limit for the range of values  $A_h$  can take. On the other side, the minimum halo area can be estimated by the diffraction on particle edge because diffraction effects cannot be avoided even in case of the perfect in-focus placement. Indeed, the product of the perimeter length  $\pi D$  times diffraction constant  $a_5$  approximates the lower limit for halo

(3)

- 175
- well (see Fig. 4). Note the large values of  $A_h$  are attained only by relatively large droplets and at far defocus. Accordingly, for a given diameter the range of total particle area  $A_p$  is also limited. It can grow with defocus distance z, but only to the point where halo constitutes 95 % of the image. Otherwise, such particle would have been rejected.



Figure 4. Range of halo areas  $A_h$  observed in measurement results with changes in diameter size D and defocus distance z for different lens magnification setting: (a) x1, (b) x2, (c) x4.

According to Eq. (1) and (2) halo grows linearly with droplet diameter while total image area grows quadratically. It means that for a given defocus distance z, halo should constitute larger fraction of the whole image for small droplets in comparison
to the large ones. With increasing z-z the halo would fill the image much sooner in case of small objects and their shadows would sooner fade away into background, making them no longer detectable. Such qualitative reasoning explains the intuitive fact that the range of distance z-z within which the object can be detected depends on the object size. For instance, effective SV depends on cloud droplet diameter with all the adequate consequences for measuring DSD.

Kashdan et al. (2003) showed that, to a reasonable accuracy, the range of possible defocus distance (depth of field, DOF)
[-z<sub>def</sub>, +z<sub>def</sub>], grows linearly with particle diameter. The proportionality factor comes from the experimental calibration. The measurement volume V is then equal to the product of the default DOF z<sub>def</sub> and effective area of camera sensor S (field of view, FOV).

$$z_{def} = a_6 D \qquad V = 2a_6 D S \tag{4}$$

In a summary file generated by the software, this formula is applied to calculate the volumes  $V_k$  corresponding to the consecu-190 tive size bins  $[D_k, D_{k+1})$ , where integers k = 1, 2, ..., K denote bin number.

However, the volumes calculated with such a method are not correct for small particles as typical droplets in atmospheric clouds. The reason for this fact is that the focus rejection condition  $(A_h < 0.95A_p)$  impose a limitation on the acceptable defocus distance z. Hereafter this limit will be denoted  $z_{95}$ , in contrast to previously introduced  $z_{def}$ . Specific value can be obtained by using Eq. (1) and (2) to expand inequality of the focus rejection criterion in Eq. (3).

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$$z < z_{95} = \frac{0.95}{(1 - 0.95a_3)(a_1D + a_2)} \frac{\pi}{4} \frac{(D + a_4 + a_5)^2}{\text{pix}^2}$$
 (5)

The effect of the above condition is illustrated in Fig. 5. Indeed, for all the droplets measured in the diagnostic experiment, Eq. (3) leads to much stronger limitation of the effective SV than Eq. (4). Predicted ranges of defocus distance agree very well with maximum values  $z_{max}$  found in the experimental dataset. The discrepancy observed for largest sizes is related to the poor statistics, i.e. small number of counts of large droplets which are infrequent in the measured plume. It can be shown, that focus rejection defines the true SV for particles of small diameter whereas it usually has no effect in the case of large particles like rain drops (then  $z_{95} > z_{def}$ ). The exact critical size depends on the lens settings and the respective calibration. When one considers the relative difference between the two values it can be calculated that it drops below 10 % for particles larger than 210, 260 and 50 µm for lens settings x1, x2 and x4, respectively. Considering the population of such droplets, the choice of the DOF limit would have only minor effect on the results.



**Figure 5.** Maximum range of defocus distance z for each size bin: derived from the volumes listed in summary file (crosses), and the actual maximum values  $z_{max}$  found in the experimental data (circles), together with two analytical approximations - default  $z_{def}$  and corrected  $z_{95}$ . Measurement series for lens magnification settings: (a) x1, (b) x2, (c) x4.

## 205 **3.3** Effective sample volumes

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Size-dependent DOF has to be accounted for while calculating DSD based on shadowgraph images. Moreover, image processing procedure includes border rejection mechanism which excludes all the objects touching the outer edge of the picture because sizing of such objects would be strongly biased. Thus, the effective cross-sectional area of camera sensor, FOV, is reduced by the margin of the width equal to a diameter under consideration. Cloud droplets are orders of magnitude smaller

210 than the whole FOV, therefore such correction, although reasonable, would not exert much influence on the final results and often might be neglected. Yet, it is significant for large drops, i.e. rain. Eventually, the *default* SVs in calculations of DSD are:

$$V_{k}^{def} = 2z_{def} \mid_{D_{k}} (L_{x} - 2D - D_{k})(L_{y} - 2D - D_{k})$$
(6)

where  $L_x$  and  $L_y$  denote the size of the FOV in  $\mu$ m and k is the number of a size bin.

However, as explained above the default solution does not account for DOF limitation due to the focus rejection condition. 215 Hence, the *corrected* SVs can be defined as follows.

$$V_k^{cor} = 2z_{95} |_{D_k} (L_x - 2D - D_k) (L_y - 2D - D_k)$$
(7)

If the total number of bins *K* is small or the spread of droplet diameters present in a dataset is particularly large, then the range of sizes for objects belonging to one bin might be significant. For that reason, it would be sensible to introduce SV prescribed exactly for the particular droplet of interest. The method of calculating DSD with the use of *individual* volumes of should be more precise from the physical point of view. On the other hand, it requires the access to the list of all droplets whereas corrected and default methods need only the accumulated number of particles within the bins.

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$$V_i^{ind} = 2z_{95} \mid_{D_i} (L_x - 2D - D_i)(L_y - 2D - D_i)$$
(8)

According to the above discussion, the effect of choosing different maximum range of defocus distance on effective SV around a droplet is shown schematically in Fig. 6.



Figure 6. Schematic illustrating the difference between the default and the corrected/individual sample volume having the same field of view,  $S = (L_x - 2D_i)(L_y - 2D_i)S = (L_x - D_i)(L_y - D_i),$ but differing in depth of field ( $z_{def}$  vs.  $z_{95}$ ).

#### Correction to concentration and size distribution 225 3.4

Proper quantitative measures of particle concentration and distribution of their sizes in a given suspension should allow for meaningful comparisons between the measurement series, different instruments and experimental conditions. As explained, effective SVs depend on object sizes. Additionally, practical choice of size bins usually involves widths growing with increasing diameter. In order to characterize droplet spatial arrangement and size differentiation, we introduce a number of counts nor-

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malized with respect to both spatial position and size, denoting it as  $N_{VD}(x, y, z, D)$ . This quantity has units of mm<sup>-3</sup> µm<sup>-1</sup> and should be interpreted as local probability density function (PDF) of droplet diameter at point (x, y, z) normalized to sum up to local total droplet concentration (number in a unit volume, DNC) at this point. Then

$$N_{VD}(D) = \frac{\int \int \int N_{VD}(x, y, z, D) dx dy dz}{\int \int \int dx dy dz}$$
(9)

corresponds to the global PDF of droplet diameters and

$$235 \quad N_V = \int N_{VD}(D) dD \tag{10}$$

to global DNC.

Ideally,  $N_{VD}(x, y, z, D)$  should describe physical reality and it cannot be easily estimated from the measurement just by binning the results  $(x_i, y_i, z_i, D_i)$ , because due to size-dependent DOF the instrument is able to detect the particle of given size D only inside the limited range of z. If one divides the ranges of the four variables into bins to construct 4-dimensional grid cells, counts entries in each cell and normalizes by cell 4D volumes, then some of the grid cells in such experimentally 240 obtained  $N_{VD}^m(x, y, z, D)$  would be intrinsically missing information. For the same reason, the limits of the integral in Eq. (9) along dz should depend on D.

When DSD is concerned,  $N_{VD}(D)$  has an advantage over simple PDF since its values can be compared between measurement series. For the same phenomenon, different lens setting should bring the same results. In order to estimate  $N_{VD}(D)$ , the number of particles within given size range needs to be divided by respective size-dependent volume. Specifically, for the three 245 methods depicted in the previous subsection:

$$N_{VD}^{def}(k) = \frac{M_k}{F\Delta D_k V_k^{def}} \tag{11}$$

$$N_{VD}^{cor}(k) = \frac{M_k}{F\Delta D_k V_k^{cor}} \tag{12}$$

$$N_{VD}^{ind}(k) = \frac{1}{F\Delta D_k} \sum_{i: D_k \le D_i < D_{k+1}} \frac{1}{V_i^{ind}}$$
(13)

where  $M_k = |\{i : D_k \le D_i < D_{k+1}\}|$  is the number of counts within bin  $k, \Delta D_k = D_{k+1} - D_k$  is the bin width and F simply 250 the number of frames included in an analysed series.

DSDs obtained with the formulas Eq. (11)-(13) for the laboratory experiment indicate (see Fig. 7) that the values of  $N_{VD}(D)$ are significantly underestimated by the default method. It is not surprising, since using Eq. (4) generates SVs which are much larger than the true ones. On the other hand, there is no significant difference between corrected and individual SVs (not shown in the figure). This observation suggests that even without having data for individual droplets usually contained in a particle file, still reasonable DSD can be obtained by correcting accumulated counts listed in a summary file.

Figure 7 compares size distributions of droplets generated with the same device and measured with different lens settings. If each configuration had resolved the whole range of diameters present in a spray, the lines would follow each other. Instead, results approximately agree only for larger droplets which is explained by the instrument resolution improving with the magnification used. The plot suggest that the true minimum droplet size for uniform detection is  $\sim 6 \mu m$  for x2 and  $\sim 12 \mu m$  for x1. As expected, those values are greater than 3.7  $\mu m$  and 6.9  $\mu m$ , respectively, reported by the producer as the resolution corresponding to the vicinity of the focal plane. Unfortunately, the results do not allow to determine such limit for the lens magnification x4. It can be speculated to equal roughly 4  $\mu m$ , while definitely stays inside the range defined by the focal plane resolution (about 2  $\mu m$ ) and the above limit for lower magnification (6  $\mu m$ ).



Figure 7. Comparison of droplet size distributions obtained with two three methods, default (def), corrected (cor) and individual (ind), in the course of the three measurement series differing in lens magnification settings. Same data plotted in (a) linear and (b) logarithmic scale.

Total DNC  $N_V$  can be calculated by suitable integration with respect to the whole range of diameters which in practice turns out into the sum over bins in case of default and corrected method or the sum over individual counts in case of individual method:

$$N_V = \sum_k N_{VD}(k)\Delta D_k \qquad N_V = \frac{1}{F} \sum_i \frac{1}{V_i^{ind}}$$
(14)

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Having the information about DNC and DSD, one can calculate simple statistics characterizing the cloud, such as mean droplet diameter  $\overline{D}$  or higher order moments of the distribution (mean surface diameter  $D_2$ , mean volume diameter  $D_3$ , effective diameter  $D_e$ ). Liquid water content (LWC) can be estimated by summing up volumes of the droplets measured. Because the DSD differs between the methods and lens settings, the resulting mean diameters and LWC will also vary. Table 4 summarizes the results obtained for the poly-disperse stream produced by an ultrasonic generator. DNC is about three to four times larger for corrected and individual method with respect to the default one. In all the cases, statistics of diameter are

smaller for new methods in comparison to the default. It can be explained by noting that the largest relative difference in SVs between the methods appears for smallest droplets (see Fig. 5), so it is their contribution to the DSD which changes at most.

Lens setting		x1		x2			x4			
Method	def	cor	ind	def	cor	ind	def	cor	ind	
$N_V  [\mathrm{mm}^{-3}]$	2.1	8.6	8.6	37.3	112.2	112.3	114.1	331.6	330.1	
$ar{D}$ [µm]	14.1	13.7	13.7	8.3	7.9	7.9	6.0	5.7	5.7	
D <sub>2</sub> [μm]	14.5	13.9	14.0	8.6	8.2	8.3	6.4	6.0	6.0	
D <sub>3</sub> [μm]	14.9	14.2	14.3	9.1	8.6	8.6	6.9	6.4	6.4	
$D_e = D_3^3 / D_2^2 \; [\mu m]$	15.7	14.9	15.0	10.0	9.4	9.4	8.0	7.3	7.3	
LWC $[g m^{-3}]$	3.6	13.0	13.2	14.5	37.3	37.7	19.6	45.7	46.2	

Table 4. Results of the laboratory experiment with poly-disperse droplets: comparison of different methods used for estimating sizedependent sample volumes.

## 3.5 Homogeneity of detection

One of the important properties characterizing particle sizing instrument, apart from the extent of the SV, is homogeneity of detection inside that volume. Perfect homogeneity can be defined as the case when the probability of detecting the given particle of interest is the same everywhere, as long as the particle appears to stand at the position belonging to the respective SV, regardless of how complicated its boundaries are. If the probability depends on the position in space, the results obtained with the system might be biased. It is particularly important for calculating the measures of droplet spatial arrangement, e.g. nearest neighbour distance or radial distribution function (Larsen and Shaw, 2018).

Within the present study, quality of detection is evaluated for shadowgraph system VisiSize D30 based on the long record
of the plume of water droplet produced by an ultrasonic droplet generator. As stated earlier (sec. 3.1), the stream of droplets was supposed to extend further than the SV in each direction. Unfortunately, no independent information is available about whether the DNC inside the visible plume was uniform. One can expect that it drops from the maximal value in the center towards the edges where the intensive mixing takes place between cloudy and clear air portions. Nevertheless, the mixing zone was observed to be outside the SV - at least on average, because in general the outflow was quite dynamic. Therefore, statistics
integrated over time will be analysed in this section. To allow for drawing conclusions regarding the detection, it is assumed that on average the real physical conditions are homogeneous, i.e. during the experiment the flow filled each part of the entire SV with identical concentration of droplets having identical properties (e.g. DSD).

Indeed, results of the experiment integrated over size, time and distance to the focal plane mostly provide relatively constant average droplet concentration with respect to the two principal directions of the camera sensor - horizontal and vertical. The

- 295 calculated average concentration falls down significantly close to the edges of the FOV (not shown) which is expected concerning border rejection procedure. Figure 8 in panels (a) and (b) shows droplet concentration  $N_V(x)$  and  $N_V(y)$  normalized droplet concentration  $N_V(x)/N_V$  and  $N_V(y)/N_V$  suitably integrated over size and over other dimensions, divided by the total concentration in order to highlight the relative dependence on position inside the sample volume. The values mostly decrease gradually from the <u>center maximum (located left from the center)</u> to the sides in case of horizontal direction and from the bot-
- 300 tom to the top in case of vertical. The relative differences are up to about, except from very close to the edges, are of the order of 10 %, i. e. They are small enough to be as well assigned to possibly caused by the non-uniformities of the plumeinstead of instrumental flaws. The y-dependence of the concentration is more pronounced for larger droplets (above 12 µm, not shown here) which might be the influence of gravity sorting. However, in case of the series recorded with lens setting x1, the concentration falls with height y by a factor of more than 10 from the bottom to the top of the FOV. We speculate this effect is
- 305 of instrumental origin, as the difference is rather too large (almost exponential) and the timescale too short to allow for the explanation by possible only by gravity sorting of the droplets in the plume. Probable reason is nonuniform illumination of the scene. Although non-uniformities can be compensated for by background normalization (such function is built-in in the software), signal-to-noise ratio of an individual particle shadow might still depend on the position with consequences for detection probability. Due to extensive halo some of the particles might have been rejected. Such problem with illumination might stem
- 310 from imperfect manual alignment between the laser and the camera, yet we note achieving the satisfactory light conditions with lens setting x1 and lower is challenging. There are no such issues for higher settings, as the FOV is smaller then and it fits easily inside the uniform core of the laser beam.



Figure 8. Dependence of total-droplet concentration on (a) horizontal, (b) vertical and (c) axial position.

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Axial dependence is more difficult to evaluate, because the extent of the acceptable DOF depends on both particle size and lens setting. As expected, average concentration decreases with *z*, since for further distances only the droplets which are large enough can be counted (see Fig. 8 panel (c)). This trend is a result of coexisting effects of DOF limitation and the shape of real DSD where large droplets constitute minor part of the total number. Interestingly, the first *z* range contains ranges contain a much smaller number of droplets than the secondmaximum located further from the focal plane, regardless of the lens setting. Such unphysical behaviour might stem from the sizing algorithm, which calculates *z* position out of the halo area.

Small particles, like the ones observed in the experiment, are blurred even in the focal plane which makes them being faulty positioned at higher distances than the true ones. Equation (1) assumes no halo for the focal plane z = 0 which cannot be

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realized due to diffraction. Then, non-zero halo  $A_h$  is measured and it results in z > 0 for all the shadows.

Moreover, it can be noted that for larger distances z the concentration rises with pixel size (falls with magnification, dependent on the lens setting chosen, see Table 1). This fact refers to the DOF limits (Eq. (4)) and respective curves plotted in Fig. 5. Comparison between the three magnifications implies that for any fixed distance z higher than roughly 117  $\mu$ m, the lower limit of the detectable size range increases with the magnification, i.e. wider spectrum of droplets can be counted at setting x1 than x2 and x4. For example, at z = 150 um, the lens setting x1, x2 and x4 allow for the detection of droplets larger than 21.4, 22.2.

26.4 µm in diameter, respectively.

However, the probability of detection might depend both on the position in space and on the particle size. Therefore, crosscorrelated dependence was examined and illustrated in the form of 2-dimensional (z,d) (z,D) maps in Fig. 9. It is easy to 330 recognize the limitation introduced by the focus rejection criterion which also controls the effective SV (red line). On the other side, the minimum distance  $z_{min}$  for a given diameter is well approximated by assuming the halo area in the focal plane equal to the diffraction term  $(A_h = \pi Da_5)$ , analogously to Fig. 4, and solving Eq. (1) for z (blue line).

What is more, the DSD seems to change with the distance z from the focal plane, in particular for the lens setting x1. Precisely, it is a simple consequence of the focus rejection criterion analysed above that the increasing portion of smaller sizes

- are not detected with increasing distance z but the concentration of sizes which are well above this limit should ideally not change with the distance along the optical axis of the system. It clearly does in In case of lens setting x1. Large, large droplets are present only far from the focal plane and the concentration of droplets of a given size grows with z. Such behaviour suggests that the sizing procedure for the measurement with lens setting x1 might have not work properly. Possibly, due to insufficient brightness of the pictures the halo area was overestimated at the cost of inner particle shadow. According to sizing Eq. (1) and
- 340 (2), this leads to overestimation of defocus distance together with underestimation of the diameter.



Figure 9. Variability of droplet detection properties in size-space domain for lens setting (a) x1, (b) x2, (c) x4.

As a consequence, we the authors discourage using lens setting x1 in further studies of cloud droplets. The large minimum particle size for uniform detection reported earlier ( $\sim$ 12 µm) makes this option of limited utility anyway. Fortunately, in Instead,

x1 can possibly serve well for the measurements of drizzle drops. In order to avoid the illumination and signal-to-noise issue described above, we would suggest, as a rule of thumb, the lower limit an order of magnitude larger than the effective pixel size

345 (3.69 μm). On the other hand, the objects need to fit into the FOV, so the largest measured size should be a few times smaller than the shorter dimension (4.1 mm) which eventually results in the conservative range of roughly 40-400 μm. Notwithstanding, so far insufficient data on drizzle has been collected to confirm this expectation experimentally.

In experimental runs with higher magnifications, i.e. lens setting x2 and x4, the  $\frac{\text{DSD}}{\text{DSD}}$  behaves like expected – large droplets appear with similar concentration both in the middle and at the sidelines of the SV. The occurrence of the smaller ones is

- 350 limited by the focus rejection criterion (red line). There is only a discrepancy between first and second (z, D) map features the decrease of concentration with z -bin. The same effect was noticed in Fig. from the maximum located a bit above  $z_{min}$ . The same effect was noticed in Fig. 8 panel (c). Most probably, it relates to the diffraction which is not included in the modeled dependence of the halo area  $A_b$  on the diameter D and distance z in Eq. 8 panel (c). (1). Namely, the equation is not correct in the limit of small z because it implies the object in the focal plane (z = 0) should be ideally sharp ( $A_b = 0$ ). Consequently,
- 355 in this limit the calculated z position is overestimated with respect to the true one. The counts representing droplets standing very close to the focal plane are shifted to the further z-bins in Fig. 9. The extent of this shift is probably not constant but decreases with the true z. Hence, the counts cumulate at some point above  $z_{min}$  creating a maximum in  $N_V(z)$ . We expect the shift to decrease because the calibration constants  $a_1$  and  $a_2$  were fitted by the manufacturer in the procedure resembling Kashdan et al. (2003) so that the Eq. (1) performs satisfactorily in the range of defocus distances and particle diameters typical
- 360 for industrious applications, i.e. a bit larger than analysed here. Therefore, the estimated *z* should approach the true one with increasing defocus and droplet diameter.

The shift of the estimated z positions with respect to the true ones is most pronounced in case of the small distances from the focal plane. Importantly, it should have no effect on the accuracy of the sample volume calculation, hence the DNC and the DSD, as long as the  $z_{95}$  is not overestimated itself but represents the true distance at which the droplets are no longer counted.

365 We expect this condition to be met if the largest possible defocus  $z_{95}$  is significantly higher than the smallest  $z_{min}$ . For instance, they differ by a factor of two for diameters larger than 8.0, 6.3, 4.8 µm in case of lens settings x1, x2 and x4, respectively. Those values are close to the minimum diameter for uniform detection estimated in sec. 3.4. It implies the influence on the DSD and the DNC within the valid size range is probably minor. However, we cannot quantify it with high confidence based on the available data.

## 370 4 Sizing accuracy

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## 4.1 Diagnostic experiment

Second diagnostic experiment was carried out to characterize instrument performance in terms of particle sizing which affects the relevant statistics of cloud droplets including mean droplet diameter and DSD. Monodisperse water droplets were generated with Flow Focusing Monodisperse Aerosol Generator 1520 (FMAG) manufactured by TSI Inc. and measured with VisiSize D30. FMAG uses periodic mechanical vibration to break a narrow jet of a liquid into droplets of desired size (within the range of

15 to 72 µm in diameter). Efficiency and accuracy of droplet generation is enhanced by aerodynamic flow focusing and charge neutralizer making FMAG a common tool applied for calibration of aerosol spectrometers and droplet sizing instruments (Duan et al., 2016).

In the course of the experiment, pressurized N<sub>2</sub> (1.0 psi) was used as the flow focusing gas, and Ultra-purified H<sub>2</sub>O as the liquid. Droplet size was controlled by adjusting two parameters: liquid flow rate and vibration frequency. Three different settings were applied resulting in droplet diameters of 20.13, 39.25 and 57.55 µm as obtained with the formula provided by the manufacturer (see the settings listed in Table 5). Geometrical standard deviation among the generated droplet population is supposed to be 1.05 or smaller (Duan et al., 2016). The smallest diameter was chosen to ensure relatively narrow spectrum as we observed significant broadening for high vibration frequency which is necessary to generate yet smaller sizes. Presumably, this results in less accurate breakage of the fluid stream or more frequent collisions.

Experimental setup was arranged in two different configurations (see Fig. 10). The first without a cylindrical case over the nozzle, with shadowgraph SV as close as possible to the nozzle head (about 4.5 cm). The second with the case and the dilution air (flow rate of  $5 \text{ Lmin}^{-1}$ ) forcing the droplets to leave the cylinder. The shadowgraph SV was then located above the cylinder exit (about 18.5 cm over the nozzle head).

Table 5. Settings of the FMAG droplet generator used during sizing tests.

FMAG 1520 settings								
Inlet air [psi]	10.0							
Dilution air $[L \min^{-1}]$	5.0							
Focusing air [psi]	1.0							
Flow rate $[mL h^{-1}]$	2.0	8.0	18.0					
Vibration frequency [kHz]	130 70 5							
Droplet size [µm]	20.13	39.25	57.55					

## 390 4.2 Results

Sample shadow images of droplets produced by the FMAG and captured by the VisiSize D30 are reproduced in Fig. 11. The configuration of the setup was as in panel (c) of Fig. 10, i.e. without the case covering the nozzle. Despite the population of droplets is supposed to be mono-disperse, one can see the range of sizes. For each droplet size specified by the parameters of FMAG, a series of images was taken by the shadowgraph with three different lens magnification settings. Results are presented

in Fig. 12 in the form of probability density functions (PDF) (equivalent to  $N_{VD}(D)/N_V$ ). Such measure was chosen in order to compare the DSD between different magnifications and measurements substantially different in total droplet concentration which itself is not quantity of interest in the current analysis.



**Figure 10.** Experimental setup for studying sizing performance: VisiSize D30 located above the exit of FMAG in order to measure outgoing droplets (a) in configuration with the cylindrical case above the nozzle (b) or without it (c).

Strikingly, all the histograms contain multiple peaks which suggests quite frequent collisions between droplets on their way from the nozzle to the SV of the shadowgraph. The position of the first peak corresponds well with the generated droplet size in most of the cases. Its width can be attributed to the inevitable spread of true generated droplet diameters as well as to the imperfect imaging and sizing of droplet shadows. Left-side skewness of the tails <u>implies suggests</u> partial evaporation. Although in general the effect of evaporation is expected to be more significant for small droplets, the skewness is evident for 39.25 and 57.55 µm measured with the lens settings x1 and x2. We speculate it might be related to the size of the sample volume which increases with the effective pixel size (decreases with magnification) as well as with the particle size (i.e. from the top-left

- 405 to the bottom-right panel in Fig. 12). The position of the nozzle exit was adjusted so that the center of the FMAG-generated droplet stream is as close as possible to the focal plane of the shadowgraph. We expect the droplets more distant from the central axis of the stream to be more likely partially evaporated because of the longer travel and exposure to the dry air blown from the area around the nozzle. Those can be detected in case of considerable sample volume but not in case of smaller SV. Importantly, this is only one of the effects which could have contributed to the observed result together with the ambient air
- 410 properties, velocity of the droplets or some interactions among them.



Figure 11. Sample shadow images captured by VisiSize D30 showing droplets of different size generated by FMAG.

Taking into account geometric standard deviation of generated droplets (1.05), histogram bin width (0.5 µm) and evaporation, the sizing by shadowgraph is pretty accurate. Only in case of smallest droplets (20.13 µm) and lowest lens magnification (x1) the reported diameters seem to be significantly biased. This issue is probably of instrumental origin and might correspond to non-uniform illumination of the FOV which also caused inhomogeneity of detection of relatively small objects for that

415 particular lens setting (see sec. 3.5).

> The fact of droplet collisions is further corroborated with measurements conducted in the configuration as in Fig. 10 panel (b), i.e. with the cylindrical case over the nozzle and longer distance between nozzle exit and the shadowgraph SV (around 18.5 cm). Longer path enhances the chance of collisions and effects of evaporation. The former can be clearly seen in histograms presented in Fig. 13. Consecutive peaks correspond to single generated droplet, double collision and triple collision as calculated simply by summing the volumes. Here, lens magnification setting x1 was selected due to largest SV, thus best

statistics which can provide better estimation of less frequent events in the probability distribution. More distant peaks can be traced better in case of largest droplet size suggesting the probability of high-order collisions might increase with size. This can be expected recalling extensive studies on cloud droplets collision kernels (Devenish et al., 2012; Grabowski and Wang, 2013). Left-skewed tails are also visible, in particular for the first peak, which is the consequence of evaporation on the way of

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Finally, the accuracy of sizing is evaluated more quantitatively in the scatter plot in Fig. 14 which compares initial droplet size as specified on FMAG with the results obtained with the shadowgraph – mean droplet diameter reported by the software and the position of the first major peak in the size histogram. Obviously, mean diameter is larger than the first peak as it includes



Figure 12. Probability distributions of droplet size measured by VisiSize D30 in configuration as in fig. 10 panel (c) (distance between sample volume and nozzle head is  $\sim 4.5$  cm), for different camera lens magnifications and different FMAG output droplet sizes. Initial size of the generated droplets as well as the size after double and triple collisions are marked with vertical lines.

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the contribution of collided droplets. Excluding the problematic case of magnification x1 for size 20.13 µm, the position of the first peak deviates up to 2 µm from the original value and the relative error ranges up to roughly 5 %. However, with regard to the accuracy of shadowgraph instrument, it is only an estimation of the upper bound for those quantities, as the generated size is also subject to intrinsic uncertainty. Hence, as far as the comparison with FMAG allows to say, the sizing by shadowgraph is indeed pretty accurate, apart from small droplets observed with magnification x1. Interestingly, in most cases the first peak estimation is mostly higher than the FMAG one while the opposite can be expected due to evaporation effects. 435 This fact suggests that the shadowgraph slightly overestimates the sizes in relation to FMAG in most cases.

5 **Field measurements** 

After laboratory tests, the shadowgraph VisiSize D30 has been used for the first time to measure droplets in atmospheric clouds - in order to test the instrument performance under harsh environmental conditions, compare it with other probes already in service in cloud physics studies, and to study microphysical properties of warm (liquid) orographic clouds. The measure-



**Figure 13.** Probability distributions of droplet size measured by VisiSize D30 in configuration as in fig. 10 panel (b) (distance between sample volume and nozzle head is  $\sim$ 18.5 cm), for camera lens magnification setting x1. Initial size of the generated droplets as well as the size after double and triple collisions are marked with vertical red lines.



**Figure 14.** Comparison of droplet sizing between VisiSize D30 and FMAG: scatter plot illustrating the position of the first peak in size histogram and arithmetic mean diameter with respect to FMAG generated droplet sizes for different lens magnification settings (left). Red boxes (a, b, c) are enlarged in panels on the right. Each pair of points (filled and empty) represents a single measurement.

- 440 ments were performed in a ground-based mountain observatory Environmental Research Station (Umweltforschungsstation) Schneefernerhaus located on the southern slope of Zugspitze in Bavarian Alps – during two observational periods in July and August 2019. Typical meteorological conditions at this place together with respective cloud and turbulence properties are described in detail by Siebert et al. (2015) and Risius et al. (2015).
- Comprehensive analysis of the field experiment alongside with the results obtained with the shadowgraphy imaging technique is going to be covered in a separate article while here we present example observations of cloud microphysical properties representing the range of conditions typical at the place. The data was collected on 13 July 2019 when the clouds covered the sky for most of the day (7-8 oct). However, due to the wind and complex terrain the observatory is usually exposed only to intermittent portions of cloudy air. Two measurements series, each 15 min long, recorded within relatively homogeneous conditions were selected. The first was performed in the afternoon (14:46-15:01 LT) using lens magnification setting x2, the
  second in the late evening (23:19-23:34 LT) using x4. Throughout the day temperature varied around 0° C. Wind was predominantly westerly, coming over the saddle in the mountain range located west from the observatory. It was stronger for the first measurement series, with velocity of around 5 m s<sup>-1</sup> and fluctuations of 2 m s<sup>-1</sup>, than for the second one (about 1.5 m s<sup>-1</sup> and 1 m s<sup>-1</sup> of mean velocity and fluctuations, respectively). There was no precipitation noticed in the afternoon, while light rain occurred in the evening shortly before the measurement.
- Figure 15 presents normalized DSDs calculated with the methods described in sec. 3.4 for the two recorded events. Basic microphysical statistics calculated based on the DSDs are listed in Table 6. Naturally, the results cannot be directly compared between the series as the conditions were slightly different. However, DNC as well as mean diameter and LWC resemble typical properties of non-precipitating continental clouds and stay close to the range of conditions reported by Siebert et al. (2015) for Schneefernerhaus observatory based on the measurements with completely different instrument (Phase Doppler Interferometer).

Time (Lens setting)	14:46-2	15:01 LT	(mag. x2)	23:19-23:34 LT (mag. x4)			
Method	def	cor	ind	def	cor	ind	
$N_V  [\mathrm{cm}^{-3}]$	359.4	927.0	928.7	341.6	797.9	797.8	
$ar{D}$ [µm]	10.8	10.6	10.6	9.0	8.8	8.8	
D <sub>2</sub> [µm]	11.0	10.8	10.8	9.1	9.0	9.0	
D <sub>3</sub> [µm]	11.3	11.0	11.0	9.3	9.1	9.1	
$D_e = D_3^3 / D_2^2  [\mu m]$	11.7	11.5	11.4	9.6	9.4	9.5	
LWC $[g m^{-3}]$	0.27	0.65	0.65	0.14	0.32	0.32	

Table 6. Results of the cloud observations performed with the VisiSize D30 at Schneefernerhaus on 13 July 2019.

The cloud observed in the afternoon seems to contain more significant portion of larger droplets (10-15  $\mu$ m) with respect to the evening one. Yet, in both cases the droplets were produced mostly by condensation as the maximum diameters measured



**Figure 15.** Droplet size distributions observed with the VisiSize D30 in clouds at Schneefernerhaus observatory during two events on 13 July 2019 - afternoon (14:46-15:01 LT) and late evening (23:19-23:34 LT), the former taken with lens setting x2, the latter x4. Three methods of calculating the results are marked with different linestyle: default (def), corrected (cor) and individual (ind) (see sec. 3.4 for explanation). Same data plotted in (a) linear and (b) logarithmic scale.

do not correspond to the sizes capable of efficient rain formation by collisions. It should be noted, that moments of the DSD (mean diameter statistics) are most probably moderately overestimated because the portion of small droplets might not be 465 properly detected in the whole relevant volume. This limit of minimum diameter for uniform reliable detection was estimated to be  $\sim 6 \mu m$  for lens setting x2 (see sec. 3.4) and unfortunately not found exactly for lens setting x4 (though definitely stays between 2 and 6  $\mu m$ ). Obviously, the degree of the bias decreases with magnification (improving resolution). For the same reason, the total DNC might be moderately underestimated with respect to the true one. Nevertheless, all the instruments suffers from similar issues whenever the range of detectable diameters is finite. The relative differences between the three 470 methods of DSD calculation are very similar to what was stated in case of laboratory experiment (sec. 3.4).

The comparison between the two example observations discussed in this section also illustrates the trade-off regarding the choice of magnification. Larger one (x4) provides better resolution and proper representation of the left tail of the DSD (below 6  $\mu$ m), though the right tail of relatively scarce large droplets is then poorly statistically represented because the total number of counts is quite modest. Specifically, despite the similar DNC and duration of the measurement, roughly 10 times more droplets counts were recorded in the first series (x2), simply due to the larger SV.

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## 6 Conclusions

The shadowgraph imaging system – Oxford Lasers VisiSize D30 – has been tested and characterized with respect to cloud microphysical measurements, i.e. number concentration and size of cloud droplets. The instrument captures images containing

shadows of multiple particles, counts them and estimates sizes correcting for image blurring due to out-of-focus position.

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Although developed for industrial applications, the system can be applied for cloud physics studies. Nevertheless, diagnostic laboratory experiments pointed out important limitations which need to be considered.

First, the sample volume within which a droplet is detectable depends on its size because blurring caused by defocus differently affects images of particles of different size. This fact has to be always taken into account when estimating droplet concentration (in a unit volume). The solution implemented in the software assumes linear relation between depth of field and particle diameter which is efficient for relatively large objects (>260 µm, exact value depends on the lens magnification).

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and particle diameter which is efficient for relatively large objects (>260 µm, exact value depends on the lens magnification). However, in case of small droplets, like the cloud ones, additional focus rejection criterion impose much stronger limit on acceptable depth of field. It affects relevant sample volume and leads to underestimated number concentration. Therefore, we developed correction method using sample volume based on that limit.

Second, the analysis of detections in a dense poly-disperse stream of droplets implied that the minimum droplet size for reliably uniform detection is significantly larger than the resolution in the focal plane. It was estimated to be ~6 µm and ~12 µm for the camera lens magnification settings x2 and x1, respectively. Potentially, it can be enhanced by careful data conditioning, i.e. with strong limit on the estimated distance from the focal plane, but at the cost of decreasing sample volume. Furthermore, detection probability was found to be satisfactorily homogeneous across the field of view, except for the small magnification setting (x1). Minor issues were revealed with respect to axial direction, probably caused by substantial diffraction effects on small droplets.

Third, the test of sizing accuracy was performed using mono-disperse droplet generator (FMAG 1520, TSI). Substantial effects of droplet collisions and evaporation were observed in the size histograms obtained with the shadowgraph. Notwith-standing, after filtering out collisions by selecting first major peak the results indicated reasonable agreement between diameters reported by the shadowgraph and those supposed to be generated by the FMAG. The relative difference was not larger than 2  $\mu$ m or 5 %, again except for the lens magnification setting x1 which caused difficulty in uniform illumination of the scene.

Finally, the system under study was applied to sample atmospheric clouds in a ground-based mountain observatory. It performed satisfactorily well under windy, cloudy, humid weather conditions and provided quite an extensive set of microphysical data which is intended to be presented and discussed in detail in a separate publication. The results of selected observations analysed here comply with the expected conditions and previous independent measurements performed at that location.

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To sum up, the VisiSize D30 can be successfully applied for cloud microphysical measurements. However, relevant quantities like droplet size distribution, number concentration, mean diameter or effective diameter need to be calculated with care, accounting for size-dependent sample volume. While conducting the experiment, one should pay an attention to appropriate adjustments of the laser and the camera in order to assure uniform illumination of the field of view. We recommend to avoid low magnifications (e.g. x1) as they make the proper illumination adjustments more difficult and are of limited utility for cloud

510 studies due to large limit on minimum diameter for satisfactory detection. Those might be instead advantageous for sampling drizzle and rain which is a topic currently under study.

*Author contributions.* J.L.N., M.M. and S.P.M. designed the study. J.L.N. planned and carried out the detection experiment (sec. 3). M.M. planned and carried out the sizing experiment (sec. 4). M.M. carried out the field experiment, while M.M. and J.L.N. analysed the collected data. J.L.N. and M.M. wrote the manuscript with contributions from S.P.M.

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