Authors' Response to the Anonymous Referee #1

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We are grateful to the Referee #1 for the insightful comments and suggestions on our manuscript. We respond to them in detail below. The original review is given in black, our anwers in blue. The responses mention also specific corrections which were applied to the manuscript.

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 µm 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 µm 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 µm, 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 µm) 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 µm. 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.

2. 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*400fps = 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 $10\,\mathrm{cm\,s^{-1}}$ 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 $\mu\mathrm{m}$. Is there a way to verify this? Why do you observe a lot of droplets larger than 20 $\mu\mathrm{m}$ 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 $\mu\mathrm{m}$ 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 $\mu\mathrm{m}$). 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.

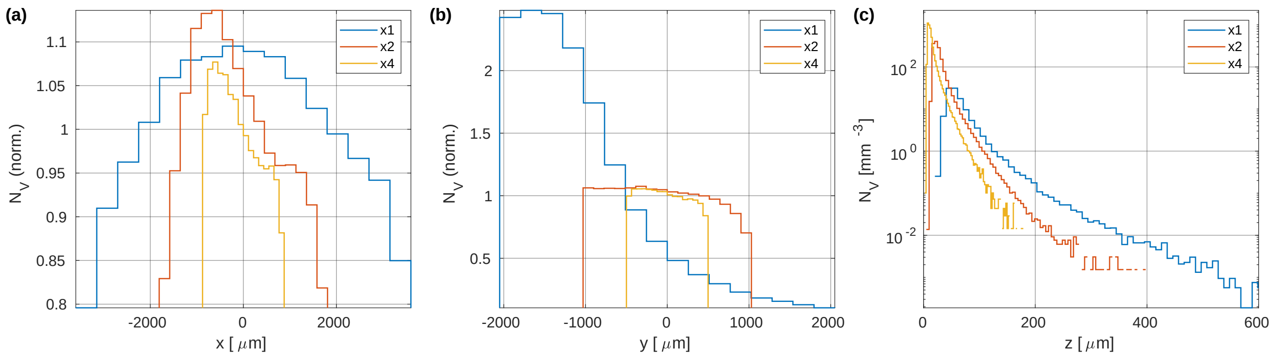

Figure 8 corrected. Dependence of droplet concentration on (a) horizontal, (b) vertical and (c) axial position.

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 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.

10. Page 16 line 335: For the size calibration experiments, you produced droplet diameters of 20.13, 39.35 and 57.55 $\mu\mathrm{m}$. 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 $\mu\mathrm{m}$, Duan et al. (2016)). As explained in our reply to the general comment above, the smallest diameter (20.13 $\mu\mathrm{m}$) 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 $\mu\mathrm{m}$, but not for x4 and the smaller size (20.13 $\mu\mathrm{m}$). 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 $\mu\mathrm{m}$ 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.

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