Atmospheric
Chemistry
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Discussions

Interactive comment on "Discrimination of water, ice and aerosols by light polarisation in the CLOUD experiment" *by* L. Nichman et al.

Anonymous Referee #1

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General comments

This manuscript presents results, primarily taken with the CASPOL instrument, of artificial cloud and secondary organic aerosol (SOA) experiments from the CERN CLOUD chamber. The Cloud and Aerosol Spectrometer with Polarisation detection (CASPOL) measures the total forward-scattered light and orthogonally polarised components of the backscattered light from particles produced in the controlled conditions of the chamber. Based on the relationship between these three measurements a classification scheme for water, ice, and (de)hydrated SOA particles is presented.

The authors discuss the importance of such chamber measurements and describe adequately both the chamber and the CASPOL instrument. The fact that this instrument has been newly installed and the CLOUD chamber has started producing cloud

particles (as opposed to doing purely aerosol studies) makes the results new and of interest, at least to a somewhat limited audience. Specific concerns are listed below however the major two are; firstly that results are presented in a highly specific manner such that its general usefulness has been lessened and secondly, that the quality of the writing hinders the reader's understanding and appreciation of the work presented. The former point has ramifications for satisfying the scope of ACP. From the journal 'Aims and Scope' webpage;

> "The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest."

In its current form I would say that the manuscript does not meet this requirement and may be better suited to submission to a more technical journal.

Specific comments

1. There is a tendency by the authors to present results in a manner that makes them less useful to the wider community. One of the draws of using an instrument like the CASPOL is for comparison with the remote sensing community which does not have access to the forward scattering information. For example, polarisation ratio is presented in a slightly different way to the more common $\beta_\perp/\beta_\parallel$ and the particle classification map is presented as perpendicularly polarised backscatter to total backscatter ratio and forward-scatter ratio. In doing so, the authors have made it impossible to compare these results with a majority of work in the existing literature. The authors also do not explain the reasons for the decision to use these different parameters.

   The authors cite the work by Glen and Brooks [1]. This paper presents maps using the same classification mapping space as in this manuscript however they

also show the same data in backscatter versus polarisation ratio space which is close to that used extensively for lidar return classification. Different plotting space does highlight different characteristics in the same dataset and it is possible that the map presented in this manuscript is the only one that leads to clear separation between the species under study. However this does seem somewhat unlikely given the volume of data presented in the literature in the more conventional way. At the very least the authors should address this, and more helpfully provide classifications with more common axes.

The afore-mentioned work of Glen and Brooks would also provide a nice comparison to the work presented in this manuscript. This could easily be done despite the different definition of polarisation ratio used in the older paper.

2. In page 31441, line 10 it is written that most of the light from particles of interest are scattered in the forward direction. However, based on the ratios in figures 3, 6, and 10, the total backscatter signal is greater than that in the forward direction. Specifics of the parameters used to calculate the ratios are not addressed in the paper so I assume that they are some instrument voltage or count that do not directly represent the scattered irradiance. This makes it difficult for those with different but equivalent instruments (or even updated versions of the same instrument) to utilise or reproduce these results.

3. The discrimination between liquid water and ice with polarised light has a long history so in addition to this, I was hoping to see the identification or classification of different ice habits with this instrument in the chamber. At the end of section 3.1 there is a glimpse of this possibility with a list of different habits formed in the chamber along with the 3V-CPI images in figure 4. Given that all these habits can be created it is unclear why the authors did not make better use them. The sentence on page 31448, line 18 insinuates that there is a limited range of habits within the chamber (at least compared to the real atmosphere). Perhaps the predominant habit cannot be selected with the conditions within the chamber?

Perhaps there was only ever a random selection of habits? If so this should be addressed along with a comment on the distribution of habits used when doing the cluster analysis. If discussion about the impact of different ice crystal habits, or even better, the classification of different habits were included this manuscript would be significantly enhanced.

4. Section 3.2 on the ACPIM modelling does not seem to add appreciably to this manuscript compared to a comment on the chamber wall heating and appropriate citation towards the end of section 2.1. See point **31445.15** below.

5. The Discussion in section 4 is disjointed and very difficult to understand. Please break this into separate paragraphs of related material.

Technical corrections

The general standard of the text is inadequate, in addition to rectifying the specific corrections below, I would encourage the authors to carefully re-read the manuscript and address this issue. Some of the following points are purely technical and some may also be of a scientific nature. Location of items are given as **page.line**.

Title The title is very vague and the use of a generic acronym does not help.

31435.4 This sentence mentions three times that there are both liquid water and ice. Remove the repetition.

31435.8 "...and measure their effects on the backscatter polarisation state." Rephrase the second half of this sentence.

31435.22 Since the relevance of the particle categorisation to TTL data is limited to several sentences at the end of the manuscript, mention of it in the abstract can probably be removed.

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31436.10 SOAs are not referred to in the previous or current paragraph so remove reference to them in this sentence.

31436.11 These complicating artefacts are unlikely to be limited to small ice crystals as stated (although the implications of such artefacts on measurements of the scattered light may indeed be more important) so rephrase these sentences.

31436.14 The optical effects of these defects depend on the orientation of the particle relative to the incident radiation not the flow. If there is a preferential particle orientation in a flow relative to the instrument, and thus laser, this then may lead to systematic measurement bias as mentioned. A reference would be good here along with comments about any preferential orientation or lack there-of in the CLOUD chamber.

31436.21 "Organic components create hygroscopicity variation...", this sentence and the next should be rephrased for clarity.

31437.13 Particles do not rotate the initial polarisation, the polarisation of the incident light is unaffected by the presence of a particle. Rewrite this sentence.

31438.2 "Recently, additional experiment focusing on..." This sentence doesn't make sense.

31439.7 The SIMONE instrument is described as in situ which suggests that the other instruments are not. Is this because the other instruments operate with inlets? If so, a description of instrument inlets and the impact on sampling should be given (with a reference if required). If not, then remove "in situ".

31440.25 "... of the latter..." does not make sense here so the sentence needs rewriting.

31441.1 Although mentioning the CAPS in the body of this section is sensible (if nothing else it allows a reader to find the instrument on the manufacturer's website which does not list CASPOL) including it in the title of this section is just confusing. Rename this "The CASPOL Instrument", "CASPOL Description", or something

like that. In general this section needs to be split up into more than one paragraph and reordered for clarity (eg currently order is forward scattering, backscattering, depolarisation, size and refractive index effects on forward scattering, then effect of qualifier. This does not make for easy reading).

31441.9 It is obvious what is meant, however angles are two dimensional; the instrument collects light over solid angles subtended by the angles given.

31441.10 The near-forward angles are used for what and instead of what alternative?

31441.27 The entire size range of the CASPOL is not given although various subranges or thresholds are mentioned throughout the manuscript. Add this information to this section.

31442.8 Size of aspherical particles is used throughout the manuscript but only receives a very cursory two sentences here. It is unclear how such particles are sized, as only Mie theory is discussed I assume that an equivalent optical diameter is used however it is still unclear what refractive index is used (particularly in cases such as figure 2 where ice and water particle sizes are presented on the same axis). The final sentence needs significant expansion. The bin size (were the manufacturer's nominal diameter bin widths used or was a calibration done as in the Rosenberg et al. [2] paper cited?) varies across the size range of the instrument but these are not given in the manuscript. Was the error assumed or calculated and was it the same for the ice and SOA particles? No uncertainties are given with the diameter data presented and plotted, this should be rectified.

31442.15 This is an unusual manner of expressing the degree of polarisation rotation in the scattered light. It would be good to see the reason that this definition was used compared to the more common one.

31442.16 "... provides a measure of..." perhaps overstates the quantitative utility of the polarisation ratio which varies with one or any combination of the particle characteristics listed.

31442.20 Unless there is precedent, I'd recommend a change of symbol for the perpendicularly polarised backscatter signal. Dpol rolls of the tongue as depolarisation (ratio).

31442.21 This is not entirely obvious. For a spherical particle in the size range of interest, the forward scattering signal will increase with size however spherical particles are somewhat irrelevant here. With aspherical particles the forward scattering may be non-uniform which makes this sweeping statement somewhat uncertain. This is linked to the use of these ratios as discussed in point 1 above but if this ratio is to be used to link size and asymmetry then further explanation and references are required to address the assumptions and subtleties.

31443.8 "...based on polarisation differences..." could cause confusion with a mathematical difference, use "variation" or something similar.

31443.11 Use standard nomenclature (off the website); MATLAB (check journal standards to see if this requires a ®), k-Means, and cite the URL for the `kmeans` function.

31444.4 The use of $d(i, C)$ here is confusing, is it the same as $d(x_j, \mu_i)$ in equation 1? If not, it would be clearer to define $b(i)$ in words only. If it is, then don't reuse i and rework the sentence for clarity.

31444.6 What validation is being referred to here? $s(i) = 1$, $s(i) > x$, or just the use of a silhouette value?

31444.9 Is the red–blue transition automatically generated by the cluster analysis? Are the particle type boundaries given in the map in figure 9 the asphericity thresholds that are referred to here? Are the thresholds set by 100% coverage of the data points, 90%, two standard deviations, etc? The cluster analysis and map is the focus of this manuscript and yet the reader is left to guess at a number of relevant details.

31444.23 See point 3 above for general comments on this section. Specifically, please comment on how representative the images from the 3V-CPI are to the particles sampled by the CASPOL at the same time?

31445.15 Define replicate. There seems to be some significant discrepancies between the model and data for times beyond 60 sec in figure 2c, ie modelled liquid concentration drops quickly to zero around 100 sec and modelled ice concentration remains constant for longer time scales.

31446.5 The meaning of the final phrase of this sentence is unclear. Does this mean that as the particles grew the asymmetry increased faster than the optical equivalent size? If so it would be very interesting to see some supporting data from one of the other instruments or a previous work on the growth of such particles. In any case some clarification is required here. It may be useful to add a plot of the ratio onto a second y-axis of figure 5.

31446.16 "As concentrations decreased below the CASPOL operating threshold of 1300 cm^{-3}..." makes it sound like the concentration is lower than the minimum detection threshold. A slight rewording is required, or even better, remove this as it has already been stated that only concentrations below 1300 cm^{-3} are considered in this analysis.

31446.21 How did cases with significant overlap of the clusters affect the classification map boundaries? Were such cases used for classification? As mentioned in **31444.6**, was there a threshold silhouette value required for a dataset to be added to the classification map?

31447.15 This is an unfortunate choice of words as this sentence is anything but clear. Surely understanding precedes classification?

31447.21 A list of different classification mapping schemes is given and then the map used here is different to these. This is discussed as a major shortcoming of the

manuscript in point 1 above however if nothing else the authors should justify their choice of classification space.

31448.4 Mention is made of segregation by size which would be useful however it has not been done. Further information is required here.

31448.11 This is unclear, does it mean that the boundaries of the regions of liquid water and that of ice are based solely on the particle size? Rewording of sentence required.

31449.8 Figure 5 does not show a PSD.

Table 1 Why are there two values for RH for run 1291.16?

Figure 1 It would be useful to have the x-axis of this plot presented in the same way (and scale?) as those in figure 2.

Figure 2 As mentioned previously, how is the diameter defined here? The colour maps are different to those used in the other concentration contour plots, these should be unified if possible. Remove the blue background from 2b, an increase in the size of the data points would also assist the reader. Add size uncertainty bars to 2b (with associated discussion of their derivation in the text).

Figure 3 Refine size of plots and size of text to make full use of column width. The symbol for the cluster centroid in the caption is incorrect.

Figure 6 Refine size of plots and size of text to make full use of column width.

Figure 9 The labels are almost illegibly small. Several colours (especially the dark blues) are too similar.

Figure S3 Add uncertainties to PSD in S3b. Improve axis labels so that there is more than a single number on the x-axis.

Figure S4 Add uncertainties to PSD in S4a. Improve axis labels so that there is more than a single number on the x-axis.

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References

[1] Glen, A. and Brooks, S. D. "A new method for measuring optical scattering properties of atmospherically relevant dusts using the Cloud and Aerosol Spectrometer with Polarization (CASPOL)", Atmos. Chem. Phys., 13, 1345–1356, doi:10.5194/acp-13-1345-2013, 2013.

[2] Rosenberg, P. D., Dean, A. R., Williams, P. I., Dorsey, J. R., Minikin, A., Pickering, M. A., and Petzold, A. "Particle sizing calibration with refractive index correction for light scattering optical particle counters and impacts upon PCASP and CDP data collected during the Fennec campaign", Atmos. Meas. Tech., 5, 1147–1163, doi:10.5194/amt-5-1147-2012, 2012.

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