Atmos. Meas. Tech. Discuss., 5, C3364–C3369, 2012
www.atmos-meas-tech-discuss.net/5/C3364/2012/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Application of linear polarized light for the discrimination of frozen and liquid droplets in ice nucleation experiments" *by* T. Clauss et al.

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Received and published: 20 December 2012

We would like to thank Referee 2 for his positive review of our paper and the useful suggestions and questions. In the following, we give the answers to the comments. The comments of Referee 2 are marked in **bold**, our respective answers in normal font.

General comments

The manuscript extensively describes a new optical particle counter, TOPS-Ice,

devoted to the detection of ice particles. In general the paper is well written and it is clear the huge amount of experimental and computational work behind it. In my opinion the main interest of this instrument is based on measuring the cross polarized component of the scattered light at 42.5 degrees (scattering angle) as a diagnostic tool for identifying spherical/irregular ice crystals. As mentioned in the manuscript, the performance of the measurements at such scattering angle provide a better signal to noise ratio that at exact backward direction (180 degrees scattering angle). It is mentioned in several parts of the manuscript (e.g. abstract, and conclusions section) that the cross polarized component of the scattered light by spherical droplets is not vanishing at 42.5 degrees (scattering angle). Strictly speaking that sentence is not correct. For spherical particles the cross polarized component (proportional to $(F_{11} - F_{22})$) is equal to zero at all scattering angles since the F_{11} element is equal to F_{22} at all scattering angles (see e.g. Yang et al, 2003 and Zakharova and Mishchenko 2000; both references are included in the reference list). In my opinion, as explained in detail below, the reason for not obtaining the expected results are not due to an erroneous choice of the detection angle (42.5 degrees) but to a not optimum design of the instrument. In any case the value of the current version of the instrument is proved by the presented results. I encourage the authors to improve the design of TOPS-ICe for future work and to clarify in the current manuscript the points detailed below.

The reason for the non-vanishing cross-polarized component of the spherical particles in the specific set-up of TOPS-Ice will be given in the specific comments below. However, the detection of the spherical particles has the advantage that they are counted as well in the same optical channel. The original idea was to detect only ice within PMT C and measure the number of all particles from PMT A/B. By measuring both ice and droplets within the same channel, we can be sure, that they are detected within the same sample volume. Of course, some improvements of the instrument can be done in the future. For example, a different scattering angle (100°) may rise the signal of the ice particles and increase the difference between the droplet and the ice particles distributions. Also a broader field of view of PMT C could be considered (different fiber in front of PMT C). These improvements will be added to the Conclusions of the paper.

Specific comments

- The scattering experiment can be described as follows (see e.g. Hovenier, Van der Mee and Domke, Transfer of polarized light in planetary atmospheres, Section 2.2): We assume the origin of the right-handed Cartesian coordinate system located inside an arbitrary particle that is illuminated by a plane-parallel monochromatic wave traveling in the positive z-direction. [...] In the particular case of homogeneous spherical spherical particles: $F_{12} = F_{21}$ and $F_{11} = F_{22}$ at all scattering angles (see e.g. Bohren & Huffman 1983; Mishchenko, Travis and Lacis 2002; Hovenier, Van der Mee and Domke 2004). The main problem is that detector B is too close to the scattering volume and/or it has a too broad detection area. In that case, as mentioned in Section 3 of the manuscript (Eqs. 3, and 4), the detected signal must be integrated over θ and ϕ. The azimuthal dependence must be specified with respect to the scattering plane taken as reference in the experiment ($\phi = 0$). That implies a multiplication for the Mueller matrix that rotates the plane of the scattered beam to the reference plane and that is the reason for a cross polarized component different from zero. I miss a more detailed discussion of this point in the manuscript. As mentioned, the sentence "the cross-polarization linear component of the light scattered by spherical particles is not completely suppressed (which is only possible for true back-scattering) is not correct unless you clearly explain the reason for that. Moreover, it would be interesting to provide the lineal

dimensions of the TOPS-Ice i.e., distances laser-scattering volume, scattering volume-detectors. Is it possible to close the detector (by using a pinhole in front of the PM tube) and/or locate the detector at a distance from the scattering volume large enough so that the azimuthal dependence can be neglected?

Considering the scattering plane for $\varphi = 0$ which is normal to the polarization of the incident light, the cross polarized component vanishes completely for spherical particles (as the Referee suggests). In TOPS-Ice, however, the acceptance angle of the scattering detection optics is not non-zero in the azimuthal direction (φ differs from -12.7° to +12.7°), which has to be accounted for the scattering matrix. The Muller Matrix in the new scattering plane inclined by an angle φ with respect to the original scattering plane and considering the projection on the analyzer can be defined as:

$$F = R(-\varphi)T(42.5^\circ - \vartheta)MR(\varphi) \tag{1}$$

with:

$$R(a) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2a & \sin 2a & 0 \\ 0 & -\sin 2a & \cos 2a & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \tag{2}$$

$$T(b) = 0.5 \begin{pmatrix} \cos^2 b + 1 & \cos^2 b - 1 & 0 & 0 \\ \cos^2 b - 1 & \cos^2 b + 1 & 0 & 0 \\ 0 & 0 & 2\cos b & 0 \\ 0 & 0 & 0 & 2\cos b \end{pmatrix} \tag{3}$$

and M the Muller Matrix of the scatterer. For spherical particles, $M_{12} = M_{21}$ and $M_{11} = M_{22}$ is valid. In general, $F_{12} = F_{21}$ and $F_{11} = F_{22}$ is not valid for spherical particles. Therefore, p. 5759, line 1 was changed to "vertically polarized (normal to the drawing plane in Fig. 1)" and the text was rewritten so that is more clear that the light passes firstly the fiber with the aperture of 25.4° and then is detected by the PMTs.

- Section 2.2. paragraph 15: "For the discrimination, the change of the polarization state of the incident light during the scattering process is used". Can you specify in which way the state of polarization of the incident light is changed and what is the main purpose of that change?

This is of course a false formulation. We rephrase the statement in the following way: "To discriminate between the spherical and non-spherical particles, we exploit the property of light scattered by spherical droplets to preserve the original polarization state within the scattering plane. This is generally not true for the non-spherical particles. The measurement of the cross-polarized component can therefore be exploited for distinguishing between spherical and non-spherical particles."

- It is not clear whether the instrument is devoted to measure a single particle (as mentioned in several parts of the manuscript), an ensemble of particles (as Section 4 seems to indicate) or both. Please, clarify.

The instrument detects the signal of a single particle, either a droplet or an ice particle. For the data evaluation, the measured values of signal height and width from many single particles are combined in a histogram. From that histogram, the number of droplets and the number of ice particles is determined. This explanation was add to the beginning of Sect. 4.

- Section 4. I find highly interesting the amplitude-width distribution analysis presented in this section. As shown in Figures 8 and 9 the analysis of the signal pulse width of the depolarization channel can be used for determining

the ice fraction. Related to that I wonder if the bi-modality of the width pulse when measuring a mixture of liquid droplets and ice particles might be related to different size distributions of droplets and ices, respectively instead of a different nature of the particles. Do you have an estimate of the size of the ice particles? do they have sizes of the same order as the water droplets presented in the sample? Have you performed experimental tests with different size distributions of droplets and ices?

Actually, it can not be excluded that the size of the ice particles differs slightly from the size of the droplets. Even if the time from the nucleation event until the detection within TOPS-Ice is quite short ($<$1.6 s, as the absolute maximum, most ice particles have much less time to grow), the size of the ice particles can be in maximum about double of the size of the droplets depending on the experiment. Additionally, the ice particles have a much more broad optical size distribution, measured with PMT A, than the droplets. This is due to the different orientations and depends on the time of the individual nucleation event within LACIS. The droplets also evaporate very slightly on their way from the outlet of LACIS to the sensing volume of TOPS-Ice. Depending on the nucleation time, the size of the ice particles also seems to differ in different experiments. For example, during the experiments with kaolinite, the size of droplets and ice particles is quite similar, in contrast to experiments with, for example, SNOMAX. Discussion on the sizes of droplets and ice particles will be added to the Sections 2.1 and 4. Nevertheless, we can be sure that the ice particles are not smaller than the droplets, and this is always the case in every laminar freezing experiment with a monodisperse droplet population. But this fact would only support the differentiation of droplets and ice particles. We also performed measurement with different droplet sizes and this didn't show a difference in the evaluation method. The whole paragraph will be added to the manuscript.

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