

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.

Anonymous Referee #2

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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

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better signal to noise ratio than 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.

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. The Stokes parameters of the incident, S_i and scattered, S_s waves are related as:

$$S_s \propto \mathbf{F} S_i \quad (1)$$

Where \mathbf{F} is the scattering matrix of an arbitrary particle in a certain orientation. The direction of scattering is given by the angle θ which is made with the direction of the incident light and an azimuth angle φ . We assume the plane of reference the scattering plane i.e., the plane containing the incident (laser beam) and scattered beam (detector at $\varphi=0$). In that case the Stokes vector of the scattered beam, S_s , is given by:

$$S_s \propto \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} F_{11} & F_{12} & F_{13} & F_{14} \\ F_{12} & F_{22} & F_{23} & F_{24} \\ F_{13} & F_{23} & F_{33} & F_{34} \\ F_{41} & F_{42} & F_{43} & F_{44} \end{pmatrix} \begin{pmatrix} 1 \\ -1 \\ 0 \\ 0 \end{pmatrix} \quad (2)$$

According to equation (2), the resulting Stokes vector at detector C is given by:

$$\begin{pmatrix} F_{11} - F_{12} + F_{21} - F_{22} \\ F_{11} - F_{12} + F_{21} - F_{22} \\ 0 \\ 0 \end{pmatrix} \quad (3)$$

In the particular case of homogeneous 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 ($\varphi=0$). 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

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volume, scattering volume-detectors. Is it possible to close the detector (by using a pin-hole 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?

- Section 2.2. paragraph 15: “For the discrimination, the change of the polarization state of the incident light during the scattering process is changed”. 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?

- 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.

- 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?

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