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Quantitative measurement of the microphysical and optical properties of cirrus clouds with four different in situ probes: Evidence of small ice crystals

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[1] Original microphysical and optical measurements were obtained in cirrus clouds on the Southern and Northern hemispheres during the INCA experiments using four independent techniques: (1) the Counterflow Virtual Impactor, (2) the PMS FSSP-300, (3) the PMS 2D-C and (4) the Polar Nephelometer probes. The combination of these four techniques provides a description of particles within a diameter range varying from a few micrometers (typically 3 μm) to 800 μm. Because of the presence of small ice crystals in cirrus clouds, it is particularly important to overcome the limited accuracy of the sensors used in the experiments for the cloud microphysical measurements. Representative examples of combined results suggest that the available measurements are reliable and can be used for the ongoing comparison between the results from the SH and NH campaigns. The results give the definite picture that the observations of numerous (5 to 10 cm^{-3}) small ice crystals in cirrus clouds are a relatively common microphysical feature. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0394 Atmospheric Composition and Structure: Instruments and techniques. **Citation:** Gayet, J.-F., F. Auriol, A. Minikin, J. Ström, M. Seifert, R. Krejci, A. Petzold, G. Febvre, and U. Schumann, Quantitative measurement of the microphysical and optical properties of cirrus clouds with four different in situ probes: Evidence of small ice crystals, *Geophys. Res. Lett.*, *29*(24), 2230, doi:10.1029/2001GL014342, 2002.

1. Introduction

[2] Microphysical parameters are critical in determining the optical properties of cirrus in both the visible and infrared spectral range, and thus influence both the modeling of climate processes and the interpretation of observations from satellite measurements. Accurate determination of the microphysical parameters is also crucial for a better understanding of the cirrus formation and evolution, including potential effects of anthropogenic emissions [*Ström and Ohlsson*, 1998; *Kristensson et al.*, 2000]. Evidence of small ice particles (i.e. smaller than 20 μm) in cirrus clouds has been presented in several studies (see among others, *McFarquhar and Heymsfield*, 1996) but the lack of reliable measurements of such ice crystal properties crucially hampers data interpretation and modeling validation. For instance, *Arnott et al.* [1994] showed from calculations based on in situ observations that small particles can contribute significantly to and sometimes dominate both the solar extinction and infra-red emission. One possibility to overcome these limitations is to combine various independent techniques for the measurements of number and bulk densities. During the European INCA experiment (INterhemispheric differences in Cirrus properties from Anthropogenic emissions) four instruments with independent measurement techniques were mounted onboard the German research aircraft Falcon operated by Deutsches Zentrum für Luft- und Raumfahrt: the Counterflow Virtual Impactor [CVI, *Noone et al.*, 1993] operated by the Stockholm University, the PMS FSSP-300 [*Baumgardner et al.*, 1992] operated by DLR, the PMS 2D-C [*Knollenberg*, 1981] and the Polar Nephelometer [*Gayet et al.*, 1997] both operated by the LaMP. The combination of these four techniques provides a description of particles within a diameter range from a few micrometer (typically 3 μm) up to 800 μm. This paper presents representative examples of combined results, and discusses the reliability of the available measurements.

2. Description of the Probes

[3] The CVI provides the inertial separation from the surrounding atmosphere of crystals larger than approximately 5 μm diameter and smaller than 60 μm diameter (aerodynamic size). The residual particles that remain after evaporation of the condensed water and volatile material are counted using a TSI-3010 condensation particle counter (CPC manufactured by TSI, Inc.). The crystal number density is derived assuming a one-to-one ratio between the number of residual particles and the number of ice crystals. The ice water content (for IWC less than about 30 mg m^{-3}) can also be derived from the Lymanα detector connected to the CVI probe. The operation of the CVI in cirrus cloud has been thoroughly described by *Ström and Heintzenberg* [1994].

[4] The PMS FSSP-300 optical particle counter basically measures particles from 0.3 to 20 μm in diameter [*Baumgardner et al.*, 1992]. Because the small ice particles are not spherical (a typical asymmetry parameter of 0.77 was measured during INCA with the Polar Nephelometer, see below) the size calibration for aspherical particles proposed by *Borrmann et al.* [2000] was considered. *Borrmann et al.* [2000] calculated size bin limits for a refractive index of

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Table 1. Random Uncertainty Estimates on Microphysical Parameters for Two Particle Concentrations, A: 5 cm^{-3}, B: 0.5 cm^{-3}

Parameter	Particle concentration			Size		Extinction Coef.		Ice water content	
Probe	FSSP-300 (1)	2D-C (2)	CVI	FSSP-300 (1)	2D-C	PMS (1)	Nephel (3)	PMS (1)	CVI (4)
Uncertainty A	30%	50%	10%	35%	25%	60%	25%	75%	15%
Uncertainty B	75%	75%	12%	35%	25%	85%	25%	100%	15%

Labels (1), (2), (3) and (4) refer to the works of *Baumgardner et al.*, 1992, *Gayet et al.*, 1996, *Gayet et al*, 2002 and *Twohy et al.*, 1997 respectively. PMS means FSSP-300 and 2D-C.

1.33 using Mie theory (spherical particles) and using the T-matrix method (aspherical particles). Differences in the size response between the calibrations for aspherical and spherical ice particles are little for sizes smaller than 4 μm but then significantly increase with size. Therefore the upper size limit of the FSSP-300 for cirrus measurements is actually 15.8 μm for aspherical particles. In the present study the particles larger than 3 μm diameter have been assumed to be ice crystals with a density of 0.9 g cm^{-3}. Coincidence effects on particle sizing have not been taken into account on data processing because these effects are hypothesized to do not significantly affect the ice crystal size spectra [*Baumgardner et al.*, 1992].

[5] The PMS 2D-C probe provides information on crystal size and shape for the size range 25–800 μm. The method of data processing used in this study has already been described in detail by *Gayet et al.* [1996]. We recall that the method provides, at 1 Hz frequency, the size spectrum distributed over 32 channels (each having a 25-μm resolution from 25 to 800 (m size range) and the usual microphysical parameters: ice particle concentration, mean particle size, and ice water content. The bulk quantities have been processed assuming empirical crystal mass-size relationship [*Gayet et al.*, 1996]. Because the sensitivity of the probe to small particles decreases with the airspeed (i.e. ~170 m/s with the Falcon aircraft), the six-first channels (up to 150 μm) have been corrected according to the results of *Baumgardner and Korolev* [1997]. We therefore assume that these corrections take also into account the miss and/or under-sizing of the particles evidenced by *Strapp et al.* [2001].

[6] The Polar Nephelometer [*Gayet et al.*, 1997] measures the scattering phase function of an ensemble of cloud particles (i. e., water droplets or ice crystals or a mixture of these particles from a few micrometers to about 800 μm diameter), which intersect a collimated laser beam near the focal point of a parabolic mirror. The light scattered at polar angles from ±3.49° to ±169° is reflected onto a circular array of 44 photodiodes. The laser beam is provided by a high-power (1.0 W) multimode laser diode operating at λ = 804 nm. The direct measurement of the scattering phase function enables us to recognize particle types (water droplets or ice crystals), to calculate the optical parameters (extinction coefficient and asymmetry parameter, see *Gayet et al.*, 2002). Non-absorbing ice particles randomly oriented in the sampling section are assumed in deriving bulk quantities.

[7] The inherent shortcomings on probes and data processing seriously limit the accuracy of derived microphysical parameters as exemplified in Table 1. The rough estimates of random uncertainties (mostly based on published literature) include statistical errors related to sampling statistics, noise errors mainly due to sampling volume determination, uncertainties from assumptions in the inversion of the PMS probe data (shape, density of particles, airspeed corrections, ...) and uncertainties on flow airspeed, pressure and temperature for CVI data. These estimates have been evaluated for a typical particle concentration during INCA of 5 cm^{-3} and for a small particle density (0.5 cm^{-3}), both with a sample duration of 5 sec.. Because the sampling statistics errors become very important for low particle density, much higher total uncertainties are estimated on PMS probe data (75% to 100%). These errors could be reduced considerably when taking averages over longer period.

3. Representative Example of Measurement in Cirrus

[8] The following section will be illustrated with a typical example selected from all the data recorded during the two INCA campaigns. A systematic analysis of the whole data set however, has shown this example to be representative. Figure 1 shows an example of a 1 hour long time-series (1 Hz data rate) of microphysical and optical parameters obtained in a cirrus cloud at an altitude of 9.6 km and a temperature of −46°C (Punta Arenas, Chile, flight on 5 April 2000). The parameters are the following: the FSSP-300 and the CVI derived particle number concentrations, the ice water content from both the PMS probes and the

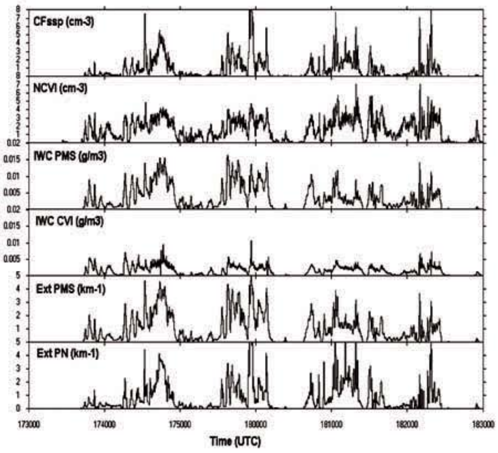

Figure 1. Example of time series (1 Hz) of the following parameters: FSSP particle concentration (*CFssp*), CVI particle concentration (*NCVI*), Ice water content from PMS probes (*IWC PMS*), Ice water content from the CVI (*IWC CVI*), Extinction coefficient inferred from both the PMS probes (*Ext PMS*), Extinction coefficient derived from the Polar Nephelometer (*Ext NP*). These results have been obtained in a cirrus cloud at the 9600 mMSL/−46°C level (Punta Arenas, Chile, Flight 000405a, 5 April 2000).

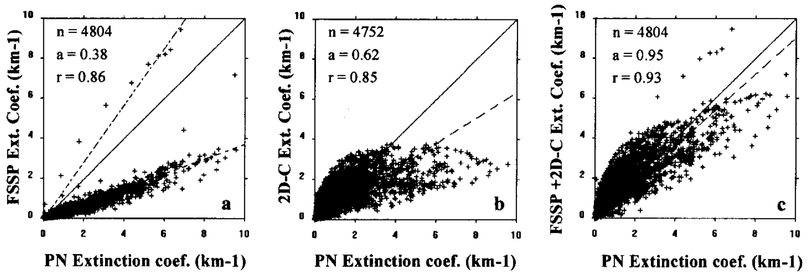

Figure 2. Extinction coefficient measured by the Polar Nephelometer versus the extinction coefficient inferred from: (a) the PMS FSSP-300, (b) the PMS 2D-C, (c) the PMS FSSP-300 & 2D-C probes. The symbols n, a and r refer to the number of data points, the slope of the best-fit curve equation (dashed lines) and the correlation coefficient respectively.

CVI, and the extinction coefficient inferred from both the PMS probes and the Polar Nephelometer.

[9] For FSSP-CVI comparison purposes and according to the CVI diameter thresholds (5 μm < D < 60 μm), the FSSP particle concentration has been calculated for particles larger than 5 μm. The PMS ice water content has been calculated from the FSSP-300 size distribution (D > 5 μm, assuming spherical particles) complemented by the two first channels of the 2D-C size spectrum (D < 60 μm). The PMS extinction coefficient (in the visible wavelengths) includes the contribution of both FSSP-300 particle size distribution (larger than 3 μm diameter, i.e. the lower Polar Nephelometer threshold) and the whole 2D-C particle size spectrum (up to 800 μm diameter). A constant value 2 of the extinction efficiency is assumed (large particle approximation). The parameters inferred from the PMS probes and the Polar Nephelometer have been smoothed by applying a running mean filter of 20 and 5 seconds, respectively, in order to remove small scale fluctuations which are not measured by the CVI (bandwidth reduction due to the evaporator system).

[10] The examination of Figure 1 reveals a noteworthy consistency between each considered parameter. For instance, the fluctuations and the large gradients on microphysical properties evidenced from 17:55 to 18:03 are closely correlated. The ice particle concentration (D > 5 μm) reaches 8 cm^{-3} whereas the subsequent extinction coefficient and ice water content are 5 km^{-1} and 15 mg/m^3 respectively. Below we continue the intercomparison of the probes by considering the different microphysical parameters measured.

3.1. Extinction Coefficient

[11] Figures 2a, 2b, and 2c represent the extinction coefficient inferred from the Polar Nephelometer as a function of the extinction coefficient derived from the FSSP-300 (calibrated for aspherical ice particles), from the 2D-C and from both the two probes respectively. The results show that the small ice crystals (D < 15.8 μm) need to be included in the calculation in order to explain the observed optical properties of cirrus clouds. They contribute in this example to about 38% of the scattering energy. As a matter of fact when the contribution of the particles sampled by the 2D-C is complemented with the contribution of the small particles measured by the FSSP-300 a noteworthy relationship is found between the Polar Nephelometer and the PMS probe measurements (Figure 2c). The slope parameter (0.95) is close to a perfect agreement and the dispersion of the data points is within the uncertainties in Table 1. This is also confirmed by the close agreement between the FSSP-300 and the Polar Nephelometer for the data points which are characterized only by particles smaller than 15.8 μm (fitted by the dotted line on Figure 2a).

3.2. Ice Particle Concentration

[12] Figure 3a represents the comparison of the ice particle concentration measured by the CVI and the FSSP-300. The dispersion of the data points can be explained by the random uncertainties in Table 1, but the non-linear relationship between the two measurements reveals systematic errors. For ice particle concentrations lower than 1 cm^{-3} the CVI overestimates the concentration with respect to the FSSP-300. For the ice particle concentrations most commonly observed with the CVI inside moderately dense cirrus, from 1 to 3 cm^{-3}, the number concentrations derived from the CVI underestimates by a factor up to 3 with respect to the FSSP-300. One possible explanation for this could be systematic errors in the calculated and measured sample flows of the CVI probes. An alternative explanation for the difference in crystal concentration may arise from the fact that the CVI samples particles based on aerodynamic size whereas the FSSP-300 derives the particle size from the scattered light. A small shift in the size of only one or two micrometers may cause large changes in the crystal number concentration. One other obvious explanation, but perhaps

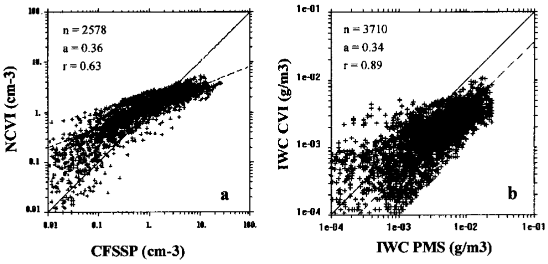

Figure 3. (a) Comparison of the ice particle concentration obtained from the CVI and the FSSP-300. FSSP-300 data include only particles larger than 5 μm (The following relationship is assumed to fit the data: log (y) = a.log(x) + b). (b) Comparison of the Ice Water Content obtained from the CVI and the PMS probes (FSSP-300 and 2D-C).

less likely, is that not all crystals leave behind a residual particle.

3.3. Ice Water Content

[13] Figure 3b represents the comparison of the ice water content derived from the CVI and the PMS (FSSP-300 and 2D-C) probes. For values larger than 1 mg/m3, the CVI underestimates the ice water content by a factor of about 3. This may be linked to the apparent systematic error in the CVI particle sampling mentioned above. The rather large scattering of the data points can be explained by both the random uncertainties in Table 1 and the low response time of the CVI evaporator system as qualitatively evidenced from the time-series in Figure 1. Because the IWC is a direct measurement in the CVI system, transport time in the tubing (3 to 5 s) and memory effects from water vapor adsorbing on wall sides, cause a smoothing of the signal and therefore may also contribute to the underestimation of IWC [*Gerber et al.*, 1998].

4. Discussion and Conclusions

[14] The results of the comparisons between the calculated bulk quantities (particle surface and volume) based on the PMS size distributions and those derived from independent measurements (Polar Nephelometer and Lyman-α) strongly suggest that relatively high (5 to 10 cm^{-3}) concentrations of small ice particles can be observed in cirrus clouds even in moderate vertical velocities of typically ± 0.3 m s^{-1}. Nevertheless, *McFarquhar and Heymsfield* [1996] suggested the FSSP-300 can overestimate ice crystal concentration, especially in the presence of large crystals. One hypothesis (not put forth by McFarquhar and Heymsfield) may be the possibility of ice crystals shattering on the probe inlets (FSSP, CVI, Polar Nephelometer) producing many small ice particles, leading therefore to FSSP-300 overcounting. While it is not possible to rigorously quantify the potential effects of the shattering of large ice particles, or to totally discount their influence, we examined the INCA data set for occurrences of relatively high concentrations of small ice particles in the absence of large ice. An example of a cirrus cloud top that contained a 2-minute average ice particle concentration of 5 cm^{-3}, with no 2D-C particles exceeding 100 μm in size, was investigated on 29 September 2000 at -49 C over the North Sea. This strongly suggests that, in this case, shattering of large ice particles is not responsible for the relatively high concentration of small ice.

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