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Water vapour measurements inside cirrus clouds in Northern and Southern hemispheres during INCA

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[1] Water vapour data inside cirrus clouds from in-situ measurements with an aircraft-borne frost-point hygrometer are analysed. These data have been obtained during two field campaigns, performed in the Southern and Northern hemisphere mid latitudes. There were many occurrences of ice supersaturation inside the investigated cirrus, with a higher frequency of occurrences in the Southern Hemisphere. The source of the differences in the humidity data from the two hemispheres is not clear, and it is speculated that these differences may be related to different levels of pollution. A distribution law for the relative humidity inside cirrus clouds is inferred. *INDEX TERMS:* 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305)

1. Introduction

[2] The INCA - 'INter hemispheric difference in Cirrus properties from Anthropogenic emissions'- project as described by *Ström et al.* [2001], was devoted to investigate the cirrus properties in polluted and non polluted areas. Then two campaigns were performed, one in the Southern hemisphere (SH) from Punta Arenas (55S, Chile) in March–April 2000, the other in the Northern hemisphere (NH) from Prestwick (55N, Scotland) in September–October 2000. To achieve the objectives, in situ measurements of aerosols and ice particles microphysical properties as well as trace gases and meteorological parameters were made with instruments on board the German Falcon research aircraft. The aim of this paper is to analyse the data from water vapour measurements, and to compare the data from the two campaigns to gain more insight into general characteristics of the humidity inside cirrus clouds. Few data on relative humidity inside cirrus are available. However it has been shown, from recent in situ measurements, that the upper troposphere is frequently supersaturated with respect to ice inside as well as outside cirrus clouds [*Heymsfield et al.*, 1998; *Gierens et al.*, 1999; *Ovarlez et al.*, 2000; *Jensen et al.*, 2001]. The observations indicate that large ice supersaturations are sometimes necessary to initiate ice nucleation. Here, we investigate the differences in water vapor data from the two hemispheres, measured during INCA, and try to understand if these can reflect some differences in terms of cirrus cloud formation.

2. Instrumentation and Data Set

[3] Water vapour mixing ratio was measured by a frost-point hygrometer, coupled to an air pressure sensor. The ambient air flows into the instrument through an air inlet from the aircraft fuselage, that is a modified Rosemount - Goodrich temperature housing (102BW model, unheated, forward facing). Thus gas-phase measurements are made without sampling ice particles larger than 5 microns diameter (Communication from Goodrich Sensor Systems). From the particle measurements described by *Gayet et al.* [2002], the contribution in the ice water content from small ice crystals that could be captured and evaporated into the instrument is evaluated to be lower than 1% in relative humidity. Moreover the hygrometer has shown its ability to make reliable measurements in high humidity conditions at low temperature during POLINAT2 [*Ovarlez et al.*, 2000]. The relative humidity is determined from the Sonntag saturation vapour pressure formula [*Sonntag*, 1994], using the air temperature data provided by the standard instrumentation on board the Falcon, with a 2σ relative uncertainty on relative humidity with respect to ice estimated better than 7%. Ten scientific flights were performed in the Southern hemisphere and nine in the Northern one, providing a large set of data. An example of measurement is shown on Figure 1. The concerned flight was performed in a well established cirrus, as the relative humidity with respect to ice (RHi) remained around saturation over a large range of H_2O mixing ratio and air temperature. Though this is the only such stable case encountered, this shows the reliability of the hygrometer inside cirrus. During all the other flights the water vapour data inside clouds were more changeable and supersaturation was frequently measured.

[4] To investigate the variability in RHi and the levels of the saturation, RHi inside and outside cirrus clouds was considered for the two campaigns separately. The particle phase discrimination (water droplets/ ice particles) was derived from the polar nephelometer [*Gayet et al.*, 1997], and all the clouds investigated in this study were ice clouds. The distinction between the in- and out-of-cloud data was made using the extinction coefficient from the nephelometer by considering the threshold of 0.05 km^{-1} extinction for cloud occurrence, that is the detection limit of the nephelometer (corresponding roughly to a concentration of 0.05 to 0.1 cm^{-3} of particles 5 μ diameter). Moreover, in order to be considered an in-cloud or out-cloud data point, the observed extinction coefficient must have remained above or below

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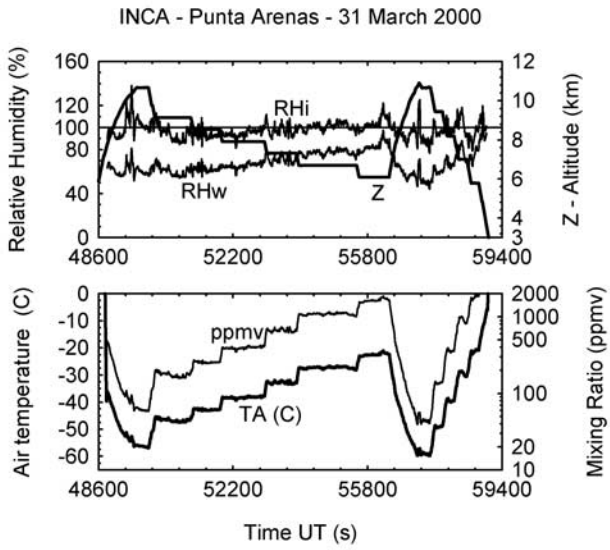

Figure 1. Measurement Inside a thick cirrus cloud from Punta-Arenas, Chile. RHi and RHw are respectively the relative humidity with respect to ice and with respect to liquid water. Here, RHi remains around 100% in the large altitude range investigated, Z (upper panel), as the mixing ratio varies between 50 and 2000 ppmv and the air temperature varies between −20 and −50C (lower panel).

the threshold for at least 4 consecutive seconds. Otherwise the data point was rejected. The 4 seconds used are compatible with the time response of the hygrometer and represent mean cloud parcels of about 700 m. The ascents and descents to and from the measuring altitude have been excluded in the analysis to avoid fast changes in the humidity due to vertical layering. After data reduction, two data sets remained: Punta Arenas, where 42% of the data are outside cirrus, 44% inside cirrus, and 14% rejected; Prestwick, where 59% of the data are outside cirrus, 28% are inside cirrus, and 13% rejected.

3. Results and Discussion

[5] Figure 2 shows the RHi data obtained during SH and NH campaigns as a function of temperature, for in-cloud and out-of-cloud cases separately. Included in the figure are the liquid saturation curve and a line representing a threshold for homogeneous freezing nucleation of micron sized solution droplets, derived by *Koop et al.* [2000]. There are frequent occasions where the humidity exceeds ice saturation and approach the homogeneous nucleation threshold in cloud free air as well as inside cirrus clouds in both hemispheres. Inside clouds, some RHi data even exceed water saturation. These data refer to single events of short duration and large associated increases in water vapor, and could be artefacts possibly from ice crystals accumulating and evaporating in the instrument inlet. They clearly reflect very exceptional situations but were not removed from the data set for further analysis. As they represent only 1% of the whole data set inside cirrus, they have very small weight in the global analysis that is done here.

[6] The frequency distribution separately for NH/SH and in-cloud/out-of-cloud cases, are displayed on Figure 3 as histograms. Outside clouds, the distribution of data is, to some extent, related to the occurrence of cirrus during the measurement flights, as the global cirrus coverage encountered during the two campaigns was different. The distribution for in-cloud cases both peak at RHi = 100% that is equilibrium as one would expect. However, measurements showing supersaturation occurred more often in SH than in NH. For further analysis of the in- cloud RHi distributions the data were divided into a warm (temperature T > −40C) and a cold (T < −40C) regime. In the warm regime the RHi distributions for both hemisphere are rather similar and fairly symmetric around RHi = 100%. However, in the cold regime, where only aqueous solution droplets can exist in

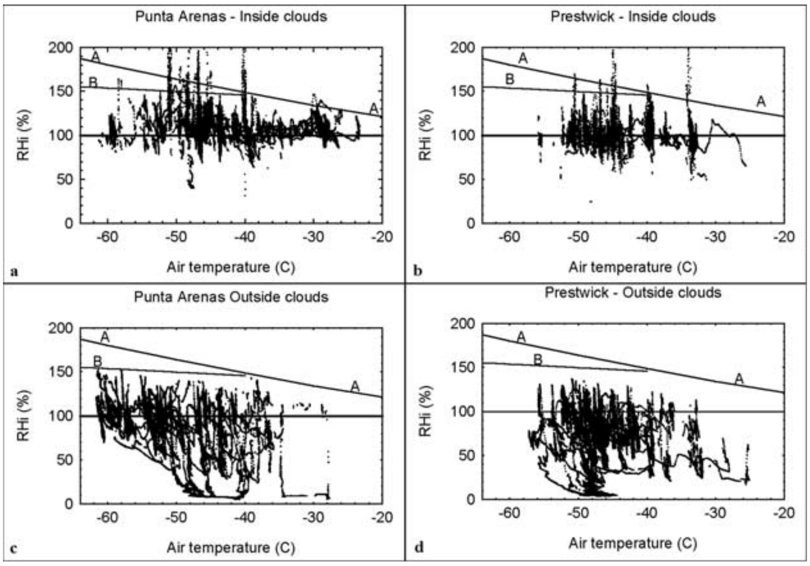

Figure 2. Relation between Relative humidity/ice and the air temperature: Inside cirrus, in SH (plate a) and in NH (plate b). Outside cirrus in SH (plate c), and in NH (plate d). On each plate are shown, the level for saturation with respect to liquid water (line A) and the homogoneous ice nucleation level from Koop et al (line B).

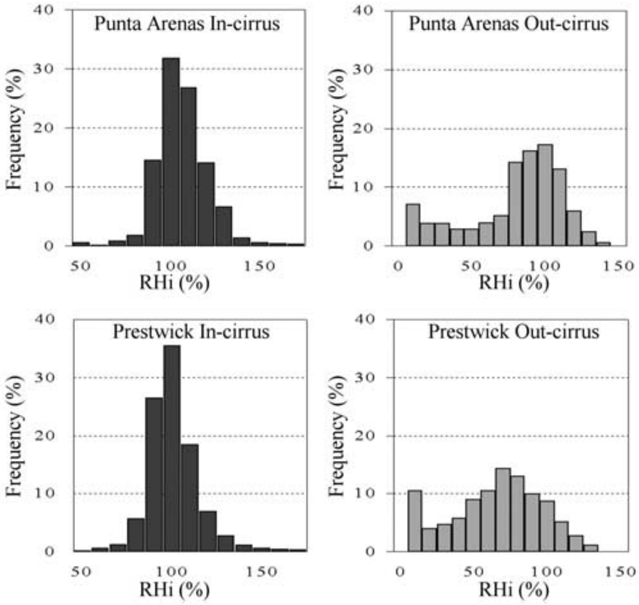

Figure 3. Histograms of RHi measurement from Punta-Arenas and Prestwick, inside and outside cirrus clouds. Inside clouds there are 51% of the data with RHi > 105% at Punta Arenas compared to 31% at Prestwick. For each location, the frequency is with respect to total number of data, inside and outside cirrus respectively.

liquid state, the two distributions differ: the one from the SH is more skewed towards higher RHi. This difference between the SH and NH RHi in cold cirrus could result from a difference in the supersaturation required to form cirrus, or from a difference in the time for cirrus to reach equilibrium (or from both factors), in a clean and a polluted environment respectively. The observations of aerosol properties during INCA show that the loading of particles is, on the average, larger in the NH than in SH by a factor 3 [*Minikin et al.*, 2001]. Moreover the air masses investigated in the SH were all of oceanic origin, whereas the air masses investigated in the NH were a mix of air from oceanic and continental origin. The question is then: could aerosols alter preferred nucleation processes (homogeneous vs heterogeneous), and is this reflected in the humidity observations from cold cirrus?

[7] The relative contribution, below −40C, of heterogeneous and homogeneous nucleation is not well known and still a matter debate since no decisive data on the topic has been published and the numerical modelling of the two processes together is difficult. Studies by *DeMott et al.* [1997] have shown that insoluble core material inside solution droplets lowers the supersaturation needed for crystal formation relative to homogeneous nucleation. On the other hand, the same study has shown that the number of ice crystals formed decreases with increasing number of droplets bearing an insoluble inclusion. On the average ice crystal concentration was measured larger by a factor of 2 in NH than in SH. Then a greater dominance of heterogeneous nucleation in NH is consistent with the lower frequency and lower levels of ice supersaturation observed in the NH compared to the SH, but not with the larger ice crystal concentration which has been measured in NH. So the possible effect of heterogeneous nucleation on the distribution of RHi observed inside cirrus clouds is not straight forward.

[8] If one assumes that homogeneous freezing of solution droplets is the dominant formation path of cold cirrus, then the number of ice crystals formed is expected to depend on the peak updraft velocity, and is relatively insensitive to the number of aerosols or their composition [*Jensen and Toon*, 1994]. The uncertainty on vertical wind speed measurements from the Falcon is quite large. However the statistical analysis from the measurements indicate quite similar distributions for the two campaigns: the higher ice crystal concentration measured in NH does not seem to result from different vertical wind field.

[9] Whatever the relative contribution from nucleation processes in the cirrus investigated, that is unclear at this stage of the analysis of the INCA data, the different RHi distributions between the two campaigns could be due to differences in the cloud particles relaxation time to water vapour equilibrium. That is the characteristic time it takes for ice crystals to reach its equilibrium with the environment (expected to be at RHi ∼ 100%). Numerical modelling by *Khvorostyanov and Sassen* [1998] -KS98- shows that the higher is the number of ice crystals, the larger is the consumption rate of excess water vapor, and the shorter is the relaxation time. In addition the studies from KS98 have put forward a possible high residual equilibrium supersaturation, which, together with the ice crystals relaxation time, increases with the decrease of ice crystals concentration. As pointed out before, on average the crystal number density measured during INCA was higher in the NH compared to the SH, which together with the differences in the humidity distributions would be consistent with the hypothesis by KS98.

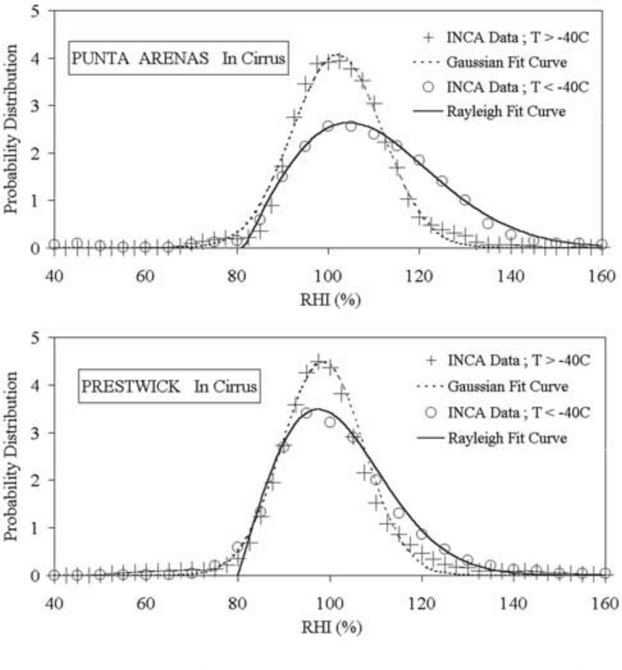

Figure 4. Probability distributions (% per % RHi), fitted by a Rayleigh distribution for T < −40C, and by a Gaussian distribution for T > −40C.

Table 1. Parameters of the Probability Distributions RHi

	Tair < −40C (Rayleigh)		Tair > −40C (Gaussian)	
	SH	NH	SH	NH
RHi max %	104.5	97.5	102.1	98.5
RHi median %	108.5	100.6	102.1	98.5
σ (%)	23	17.4	9.7	8.9

[10] In order to better quantify the differences between the measured in-cloud RHi distributions we have fitted analytical distribution laws to the data. For warm clouds, Gaussian probability densities are appropriate:

$$F(\mathrm{RHi}) = \left[1/(\sigma\sqrt{2\pi})\right] \cdot \exp\left[-(1/2\sigma^2) \cdot (\mathrm{RH_M} - \mathrm{RHi})^2\right]$$

where σ is the standard deviation, $\mathrm{RH_M}$ the mean value. For the cold clouds (below −40C), due to the dissymmetry of the distribution, particularly in SH, it is possible to fit the data with a Rayleigh distribution law:

$$F(\mathrm{RHi}) = \left[(\mathrm{RHi} - \mathrm{RH_0})/\sigma^2\right] \cdot \exp\left[-(1/2\sigma^2) \cdot (\mathrm{RH_0} - \mathrm{RHi})^2\right]$$

where $\mathrm{RH_0}$, set to 80% here, is the lower limit for RHi where the probability to observe cloud particles becomes very low, and then assumed to be zero in the distribution.

[11] The data together with the respective fits are shown in Figure 4, and the parameters given in Table 1. A Student's t test applied to the data shows that the differences found between the NH and SH distributions are significant (the significance level is higher than 99.99% due to the large amount of data in the ensemble). The higher dissymmetry for SH, below −40C, corresponds to more occurrences of nucleation processes which require higher saturation levels or (and) correspond to longer mean relaxation time than in NH as discussed before. The maximum and median value for RHi being higher than 100% in SH could correspond to the concept of a residual equilibrium supersaturation as proposed by KS98.

[12] For the sake of completeness, we mention that the relative humidity in ice-supersaturated air outside clouds follows an exponential distribution as proposed by *Gierens et al.* [1999] and *Spichtinger et al.* [2002].

4. Summary and Conclusion

[13] Water vapor measurements performed inside cirrus of the two hemispheres during INCA show less occurrences and lower levels of supersaturation in NH, than in SH mid latitudes. The probability distribution function proposed for RHi gives a clear indication of the differences between the two hemispheres, below −40C. Those differences could be due to the higher level of pollution in the NH, which we speculate leads to the observed higher ice crystal concentrations and attendant lower ice crystals relaxation times. To understand the processes involved detailed analysis of vertical wind field and aerosol data available from INCA are to be made.

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