Upper Tropospheric Humidity: A Report on an International Workshop

12-15 June 2007, Karlsruhe, Germany

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Background

Why does ice at the lowest atmospheric temperatures sometimes appear not to nucleate in aerosol particles? Why do thin cirrus clouds at the lowest atmospheric temperatures sometimes appear not to absorb water vapour despite considerable supersaturation?

These and related questions were discussed by 38 scientists from 9 nations during an international workshop co-sponsored by SPARC in Karlsruhe, Germany, in June this year. Less than seven years after the WAVAS report[1], SPARC's "Assessment of Upper Tropospheric and Stratospheric Water Vapour", there is renewed interest in the question of water vapour measurements, in understanding and judging their reliability and in estimating consequences of deviations from our traditional "textbook" understanding of cirrus cloud-driven dehydration processes.

The discovery of pronounced supersaturations with respect to ice in upper tropospheric cloud-free air and inside cirrus clouds calls into question our understanding of the physics of ice cloud formation. These findings represent potentially important modifications in our characterisation of upper tropospheric and stratospheric water and energy budgets, with implications for cloud formation, fluxes of water and radiation, and atmospheric chemistry. At the core of understanding processes in cirrus clouds is the requirement for accurate measurements of water vapour and total water concentrations under field and laboratory conditions. Currently applied and newly developed instruments with improved sensitivity and time resolution require elaborate calibration procedures. However, recent observations of unexpectedly high supersaturations using different kinds of instruments warrant close scrutiny of the various hypotheses put forward as explanations by laboratory experimentalists, cloud modellers and ice theoreticians.

The workshop was held in sessions devoted to field observations and instrumental issues, laboratory work on single particles and bulk proxies, and microphysical and large-scale modelling.

General

The relative humidity with respect to ice, also called ice saturation ratio S_{ice}, is defined as

$$S_{ice} = \frac{p_{H2O}}{p_{H2O,\,sat}(T)}$$

where p_{H2O} is the partial pressure of water in the gas phase and $p_{H2O,sat}$ is the vapour pressure of ice, which according to the Clausius-Clapeyron equation is a strong function of temperature. Given sufficient time inside an ice cloud we expect that – according to our traditional understanding – the partial water pressure will equilibrate to the vapour pressure through growth/evaporation processes of the ice particles, until $S_{ice} = 1$ is reached.

Traditionally one assumes that the timescale for water uptake by ice particles in supersaturated air is in accordance with molecular diffusion of the water molecules from the gas phase to the ice surface. One further assumes a mass accommodation α of the water molecules on the ice surface in the range $\alpha \gtrsim 0.1$ (*i.e.*, at least every tenth H_2O molecule hitting the ice surface adsorbs and is accommodated on the surface, while the others are rejected and return to the gas phase).

We know from laboratory studies that, irrespective of composition, ice nucleates homogeneously in aqueous aerosol particles at $S_{ice} > S_{nuc}$, *i.e.* ice nucleation above a critical relative humidity which is well established by laboratory and theoretical work, $S_{nuc} = 1.4...1.8$ for $T = 240...180$ K (Koop *et al.*, 2000).

According to this general framework, recent field and laboratory observations of S_{ice} can be classified in the three groups shown in **Figure 1**:

(a) $S_{ice} \lesssim S_{nuc}$ outside and S_{ice} 1 inside cirrus clouds (the "text book case", upper grey panel in Figure 1 (the corresponding ice saturation ratios are shown in the lowermost panel),
(b) $S_{ice} > S_{nuc}$ without obvious ice formation (centre grey panel in Figure 1),
(c) $1 \ll S_{ice} < S_{nuc}$ inside cirrus clouds without a clear tendency to equilibration (lower grey panel in Figure 1).

Cases (b) and (c) are in apparent violation of traditional cloud microphysics. Much of the workshop discussion revolved around whether or not measurements were sufficiently accurate to actually constitute a violation of traditional microphysics in the rapidly changing environment of the upper troposphere, including small-scale temperature fluctuations and lofting of air above convective systems.

Field observations and instrumental issues

Relative humidity can be determined either by directly measuring the partial pressure p_{H2O}, *e.g.* by Lyman-α or tuneable diode laser absorption spectroscopy (TDL), or by measuring the frost point (*i.e.* the temperature at which $p_{H2O,sat}(T) = p_{H2O}$) using a frost point hygrometer. In both cases the ambient temperature needs to be measured, from which $p_{H2O,sat}(T)$ is derived in order to calculate S_{ice}. **Karen Rosenlof** introduced the topic by providing an overview of humidity measurements, from Alan Brewer's early frost point hygrometer measurements in 1943, to the SPARC WAVAS report and

[1] http://www.atmosp.physics.utoronto.ca/SPARC/WAVASFINAL_000206/WWW_wavas/Cover.html

modern measurement techniques. Ambient temperature measurements on aircraft or balloon sondes have an accuracy of about ± 0.5 K, with main errors resulting from radiation corrections on sondes, and from static pressure corrections on aircraft. This results in an uncertainty in S_{ice} of about ± 10 %. Although this uncertainty could explain a fraction of the observed unusual S_{ice} data, the larger part must have other reasons.

Cornelius Schiller provided evidence for $S_{ice} > 1.4$ inside a cold (180-190 K) 2 km thick cirrus layer at 16-18 km on 19 November 2005 north of Australia, observed by the stratospheric research aircraft Geophysica. **Martina Krämer** provided further evidence for ice supersaturations inside and outside of cirrus from many flights in arctic, mid-latitude and tropical field campaigns. She estimated that the range of observed S_{ice} does generally not exceed the possible range bounded by traditional ice growth theory (Korlev and Mazin, 2003). This result was controversially discussed by the participants. She also highlighted ambiguities connected with in-flight cross-calibration of the two Lyman-α instruments on board the Geophysica – one for gas phase the other one for total water measurements. Care must be taken that calibration efforts do not lead to an additional bias, *i.e.* an enhancement of the calculated supersaturations. Often, optical instruments such as the FSSP only provide little help to discriminate between clear air and in-cloud situations, because very high cirrus clouds may be very thin so that the counting statistics of optical instruments is too low to detect these clouds.

Jessica Smith reported on several campaigns from Costa Rica, the Houston experiments, and Crystal-Face from Florida. Evidence from *in situ* observations on board the research aircraft WB-57 of supersaturations in clear air and in cirrus from the mid-latitudes to the tropics was provided. There is a potential high bias in S_{ice} of 5 % inside clouds.

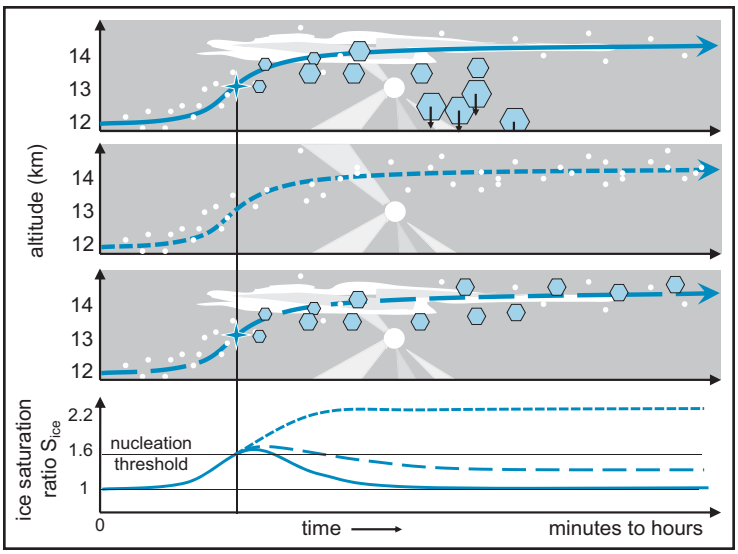

Figure 1: Ice cloud formation in rising air. The top three panels sketch three scenarios for the formation of ice clouds along an ascending air parcel trajectory; the bottom panel sketches the effect of these scenarios on supersaturation. According to conventional understanding, ice particles nucleate (star), grow, and reduce the supersaturation (solid curves). Recent observations suggest suppressed nucleation (short dashed curves) or suppressed growth (long dashed curves) in large parts of the atmosphere. Adapted from Peter et al. (2006).

The picture emerging from the European and US observations is that measurements at $T > 200$ K appear to be in general agreement with homogeneous ice nucleation and ready growth of ice particles, leading to the absence of high supersaturations, and general agreement with the upper panel in Figure 1. Conversely, at $T < 200$ K very high S_{ice} occur, reaching Sice $> S_{nuc}$, (compare **Figure 2**). Widespread regions supersaturated with respect to ice (with S_{ice} = 1.1-1.6) are observed by MOZAIC, a programme with five A340 passenger aircraft that since 1994 has obtained more than 250,000 hours of Humicap-H Vaisälä data of relative humidity. The MOZAIC climatology identifies about 30% of the upper tropospheric air as supersaturated, as was shown by **Herman Smit**. In contrast, much lower supersaturations are found by CARIBIC, also operating from the A340, as reported by **Andreas Zahn.**

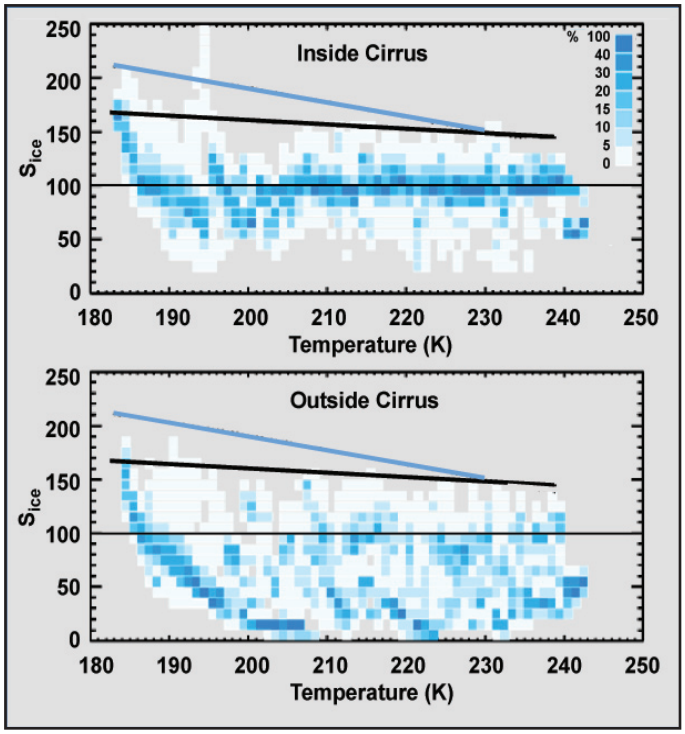

Figure 2: Frequency of occurrence of ice saturation ratios, observed inside and outside of Arctic, mid-latitude and tropical cirrus. The data set represents about 13 h of airborne in-situ observations inside and 16 h outside of cirrus (data are sorted in 1K temperature bins; black line: homogeneous freezing threshold, blue line: water saturation line). Adapted from Krämer et al. (2008).

Sean Davis showed that rocket exhaust plumes could constitute an excellent opportunity for constraining the accuracy of water vapour measurements, given that the unique chemical environment of the plumes and their water content are well known. **Liz Moyer** asked how measurements of isotopic composition can help to clarify the supersaturation issue. Current instruments can distinguish fresh convective outflow cirrus from *in situ* formed cirrus. In future, it may be possible to distinguish *in situ* condensation of ice at equilibrium vapour pressure from ice formed under diffusion-limited non-equilibrium conditions.

Daniel Cziczo started the session on water/trace gas interactions by focusing on the effect of organic species on atmospheric ice formation. To this end single particle mass spectrometry is an ideal *in situ* and real time method to qualitatively determine the chemical composition of particulate matter in the atmosphere. Internally

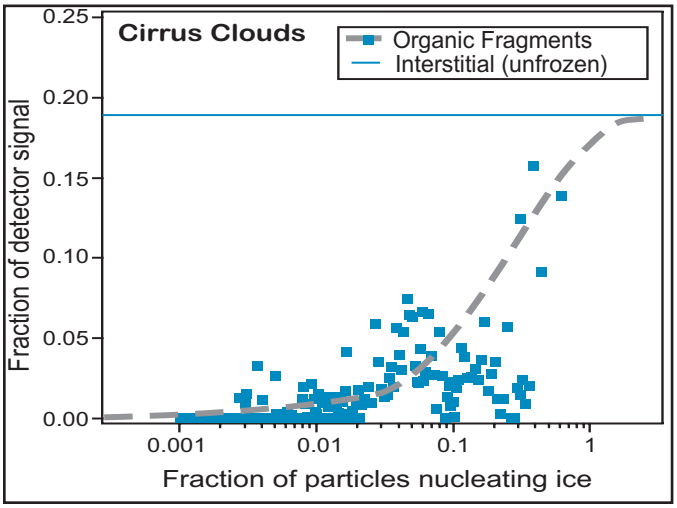

Figure 3: Average fraction of the aerosol mass spectrometer signal attributed to organic fragments as a function of the fraction of the background aerosol that was nucleating ice (observed during CRYSTAL-FACE with ice crystal densities from 0.001 to 300 cm^{-3}). Ice nucleation happens preferentially in particles with little or no organic content. Only when almost all pre-existing particles were nucleating ice was also the organic fraction activated, as shown by the blue horizontal line. Adapted from Cziczo et al. (2004).

mixed sulphates and organics dominate the free tropospheric aerosol in terms of mass, and they represent the major fraction of all aerosol particles leading to homogeneous ice nucleation under very cold conditions. However, under conditions of moderately low temperature and low saturations ($S_{ice} \gtrsim 1$) refractory particles, such as mineral dust, fly ash or metallic particles, dominate. In addition, there is also evidence that under homogeneous nucleation conditions ice preferentially nucleates in sulphate-rich particles, while the organic-rich fraction stays preferentially in the interstitial aerosol (see **Figure 3**). This behaviour could be caused by changes in the relationship between solute mass fraction and water activity of the supercooled liquid phase, by modifications of the accommodation coefficient α for water molecules, or by a combination thereof (Kärcher and Koop, 2005). In turn, the changes could be due to organic films, but this would have to lead to a reduction in α as low as 5×10^{-5} to 8×10^{-3} (depending on cooling rate) in order to explain the observed high RH in the upper troposphere, and the presence of just a few uncoated particles would negate this effect.

David Fahey and **Christiane Voigt** directed the discussion on observations of HNO_3 on ice and its potential implications for humidity equilibration in cirrus clouds and contrails. **Figure 4** by Gao *et al.*, (2006) shows one of those measurements which were most discussed during the workshop, but which was also met with concern over data accuracy. A contrail self-sampling experiment, using data from a contrail formed and sampled by the NASA WB-57F high-altitude aircraft, showed an average $S_{ice} \approx 1.31$ at temperatures of 195–200 K within the contrail. This is 31% higher than expected over pure hexagonal ice, while the combined uncertainty of the measurements is only ± 11 %. One explanation is that HNO_3 adsorbed as nitric acid trihydrate (NAT) on the ice surface might block growth sites and enhance the equilibrium S_{ice} in the low-temperature contrail, as described by Gao *et al.* (2004). This could result in a lower mass accommodation, *i.e.* a kinetic effect. While HNO_3 in contrails has also been found in the ice phase of thick anvil cirrus and in subvisible cirrus, *e.g.* during the European SCOUT-O3 campaign, the blocking by NAT so far is just a hypothesis that will require laboratory testing of ice growth in the presence of HNO_3 under a range of microphysically relevant atmospheric conditions of temperature and humidity.

These field data triggered a scientific debate about whether it was at all possible that such high supersaturations in ice clouds could persist. **Tom Peter** noted that "persistence" should mean that the supersaturation was maintained significantly longer than would be the case by uptake *via* diffusion of H_2O molecules through the gas phase, and their subsequent mass accommodation with $\alpha \approx 0.1$. Conversely, brief excursions to very high S_{ice} are not necessarily an indication for unusual physics. This was illustrated by **Klaus Gierens** with the example of the wake of aircraft wings, where $S_{ice} \approx 10$ can be reached leading to aerodynamic contrail formation.

The following discussion ranged from the assertion that the observations were severely affected by a measurement offset, to the opposing statement by one laboratory scientist that high supersaturations must occur below 200 K based on experimental evidence, and that the field observations were wrong if they did *not* show pronounced supersaturations.

Recent intense laboratory calibrations of the Lyman-α hygrometer flown on board the WB-57 were detailed by Jessica Smith. Multiple calibration standards, tied to independent physical properties of water vapour, provide the means to minimise systematic errors in the calibration system, suggesting an accuracy of 5 % and a precision of better than ± 0.2 ppmv. In particular, the calibration work resulted in no evidence of an offset in flight.

Holger Vömel showed balloon-borne observations of $S_{ice} \gg 1$ in cirrus layers that were 1 to 5 km thick, just below the tropical tropopause over Biak in Indonesia. These measurements are corroborated by very similar observations from FLASH-B, a balloon-borne Ly-α hygrometer, revealing high supersaturations in 2 km thick cirrus layers below the tropical tropopause in West Africa, as presented by **Vladimir Yushkov.**

In addition, Holger Vömel reported on attempts to address the issue of disagreements between various *in situ* instruments measuring H_2O in this altitude region.

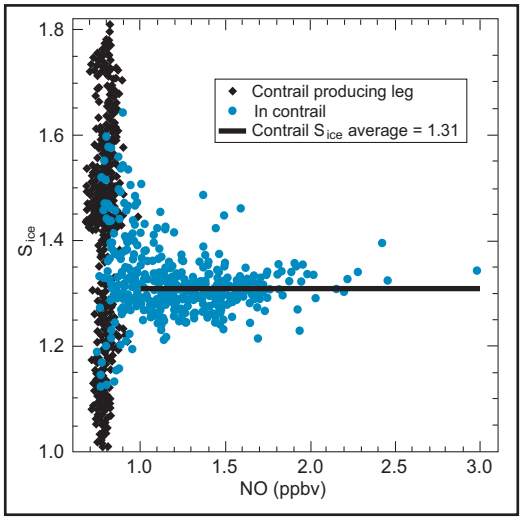

Figure 4: Derived S_{ice} as function of nitrogen monoxide (NO) on contrail-producing (CP, black diamonds) and contrail-sampling (CS, blue circles) legs measured onboard the WB-57 during a contrail self-sampling flight. The background NO values are nearly constant at 0.7 ppbv. NO values found above this value on the CS leg indicate contrail air affected by the CP leg. The black horizontal line represents the average $S_{ice} \approx 1.3$ found on the CS leg. Adapted from Gao et al. (2006).

Figure 5 shows measurements from a balloon launch at Midland, Texas, which was coordinated with water vapour measurements on board NASA's WB-57 high-altitude research aircraft (Vömel *et al.*, 2007). In this comparison, the WB-57 spiralled between 12 km and 18 km during the balloon ascent and descent, with the Harvard Lyman-α hygrometer and a NOAA/CSD aircraft frost point hygrometer on board. The figure shows all descent and ascent profiles of the WB-57 instruments and the descent profile of the balloon instrument (there was severe contamination on ascent). Throughout the entire altitude region, the Harvard Lyman-α hygrometer shows values more than 50 % above those measured by the balloon instrument. This large discrepancy cannot be explained in terms of the known instrumental uncertainties. While there was no progress during the workshop concerning the source of this difference, "the closer agreement of the two frost point instruments may indicate that the difference is not related to the measurement platform, but rather to the techniques or instrumental implementations of the technique" (Vömel *et al.*, 2007).

Also, at the end of the workshop, debate remained about these discrepancies. While all instruments clearly indicate substantial supersaturation within cirrus clouds, which is presently unexplained, there are significant differences between the various instruments, with the aircraft measurements being generally higher than the balloon instruments in direct comparisons. And while the frost point hygrometer could be regarded as the canonical instrument, the sophisticated calibration work done by the Harvard group on their Lyman-α does not allow for any conclusion on the source of the error, but calls for similar calibration efforts for all instruments.

Less controversial was the last part of this session introducing new developments. **Ulrich Bundke** reported on new fast frost-point measurements for use in nucleation chambers or airborne applications, and **Frank Wienhold** on a novel radio-sonde payload to study UTLS aerosols and clouds. The latter reported on the new development of a lightweight aerosol backscatter sonde, which may eventually have similar characteristics as the Rosen and Kjome (1991) sonde, but be light enough to fly on regular radio-sondes. **Volker Ebert** reported on the new development of an absolute humidity sensor without gas sampling based on tunable diode laser (TDL) absorption spectroscopy and its application to in-cloud supersaturation measurements in the AIDA cloud chamber. Finally, **Marc Zondlo** showed calibrations and first test flights of the HIAPER vertical cavity surface emitting laser (VCSEL) near 1854 nm.

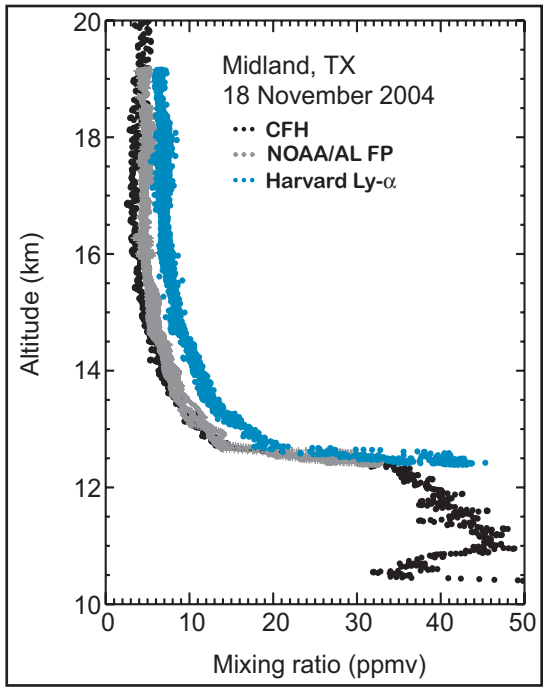

Figure 5. Comparison between the balloon-borne frost point hygrometer (black dots) and the WB-57F instruments Harvard Lyman-α (light blue dots) and NOAA/CSD frost point hygrometer (grey crosses). Adapted from Vömel et al. (2007).

Laboratory studies of ice nucleation and growth

Thomas Leisner, host of the meeting, opened the session by talking about fundamentals of ice nucleation as derived from electrodynamic levitator techniques applied to single aerosol particles. By means of levitation experiments with supercooled water microdroplets he showed that ice nucleation is a process that is volume-dominated, at least as long as the droplets are larger than 4 μm in radius. Only for smaller droplets might surface nucleation become important and possibly dominant, if at all. Indirectly, this finding on pure water droplets makes it less likely that the formation of an ice nucleus could be hampered by surface contamination and that impurities in the surface region of water droplets (*e.g.* surfactants) could actually be responsible for the observed suppressed nucleation tendencies.

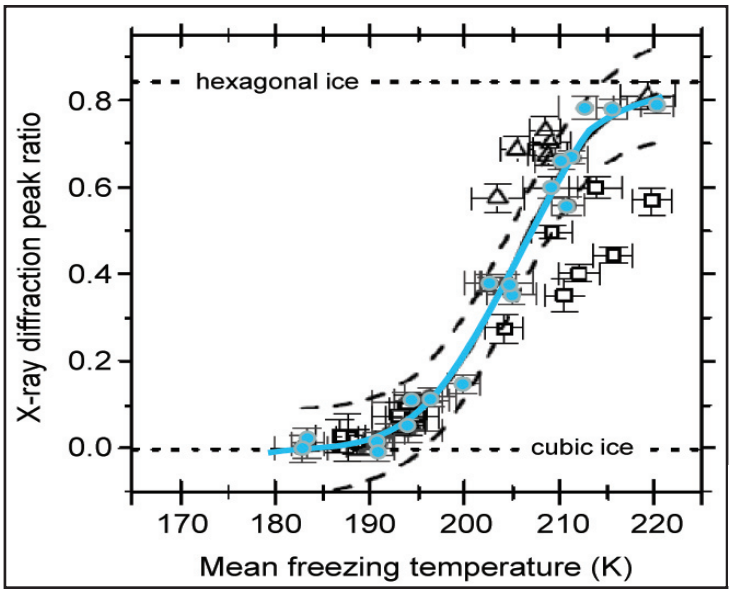

Figure 6. X-ray intensity peak ratio of the intensities of the exclusive hexagonal peaks and the peak intensity common to cubic and hexagonal ice. The data are grouped into three size bins, 2–5 μm (Δ), 5–10 μm (•) and 10–20 μm (□) Adapted from Murray and Bertram (2007).

Dennis Lamb presented laboratory work using an electrodynamic particle trap to measure cycles of growth and partial evaporation of an ice particle under conditions of periodically varying super- and subsaturation (from $S_{ice} < 1$ to about 1.2). These experiments led to the perplexing result that the mass accommodation coefficient of the water molecules on the ice surface could be as low as $\alpha \approx 0.006$, *i.e.* only 6 out of every 1000 molecules are involved in vapour deposition to the small ice particles (Magee *et al.*, 2006). This result has important implications for cirrus clouds and for maintaining high supersaturations in the UT. The physics behind these extremely small mass accommodation coefficients needs to be studied much further.

The crystallisation of aqueous droplets at extreme tropopause temperatures was also

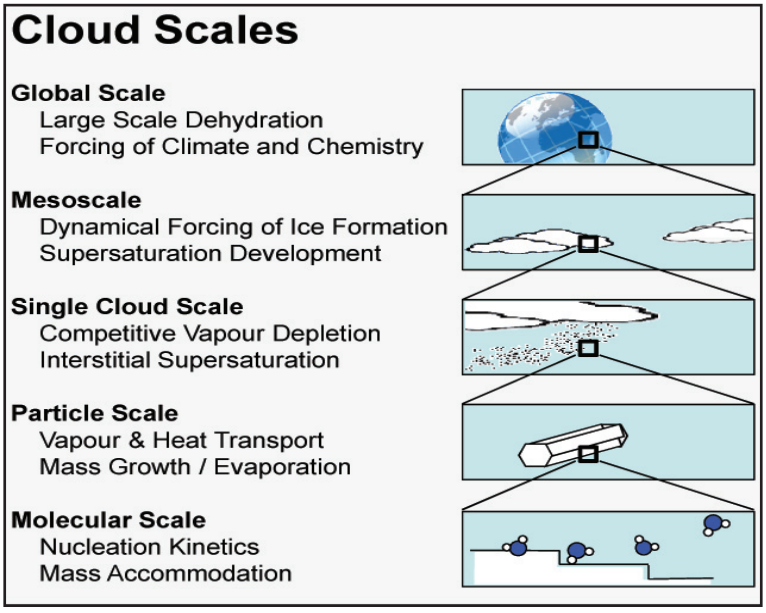

Figure 7. Various cloud scales addressed during the workshop.

studied using droplets suspended in emulsions. **Ben Murray** showed that cubic ice is the dominant product when solution droplets freeze below ~190 K, so that at least a part of the high S_{ice} below 200 K could be accounted for by cubic ice. The crystallisation of solution droplets is a complex process (see **Figure 6**), and the cubic-to-hexagonal phase transformation can be solvent mediated. This transformation may be blocked when the solution becomes very viscous. At the lowest temperatures the presence of organics may further enhance the viscosity of the solution within droplets, leading to the formation of a glass, which in turn may suppress ice nucleation altogether, as was suggested by **Claudia Marcolli** and **Thomas Koop** based on emulsion experiments using differential scanning calorimetry.

Cirrus simulation studies in the aerosol and cloud chamber AIDA (Aerosol Interactions and Dynamics in the Atmosphere) revealed that organic coatings on solid particles may suppress their activity as heterogeneous ice nuclei. By means of cirrus formation experiments **Ottmar Möhler** showed that ice nuclei, such as mineral dust and soot, may partly or even completely lose their ice nucleation activity when they are covered by organic coatings. Interestingly, switching from the deposition mode (with direct ice nucleation on the solid dust or soot particles) to the immersion mode (with ice formation on a solid nucleus surrounded by an aqueous liquid), may reduce their activity, a process that is not easily understood in microphysical terms. Subsequently **Harald Saathoff** reported on the general possibility of a systematic intercomparison and systematic testing of humidity and water vapour instrumentation within AIDA. This discussion was resumed again in the last part of the workshop, preparing the Aqua Validation and Instrument Tests Intercomparison Campaign of Water Vapour Measurement Techniques (AquaVIT), which took place 8-26 October 2007 at the AIDA facility Karlsruhe (see below).

Modelling of ice processes and their impact on global humidity

What happens on the molecular scale when ice nucleates in a solution droplet and what happens with the solute during the subsequent crystallisation process? How does this affect cloud formation all the way to the global scale (**Figure 7**)?

Lubos Vrbka showed molecular dynamics simulations of water freezing suggesting that the initial event of homogeneous nucleation is preferentially in the subsurface region stimulated by salt concentration fluctuations. This finding relates to the experimental work by Leisner mentioned above, by emphasising the necessity for experimental work on submicron droplet freezing.

Also, the growth process of ice crystals at extremely low temperatures is presently not well understood. One way to model the ice surface and the mass accommodation of the water molecules approaching the surface is to assume that whenever a crystal plane has been completed during a growth phase, the start of a new plane requires overcoming an energy barrier. In this model, growth occurs *via* repeated nucleation events, and the H_2O mass accommodation coefficient becomes a function of relative humidity, $\alpha = \alpha(S_{ice})$, see Wood *et al.,* (2001). Two groups (**Thierry Corti** and **Beiping Luo**; **Marcia Baker**, **Jon Nelson** and **Jennifer Kay**) took this approach. Parameters of the lattice plane nucleation are adapted from laboratory experiments (*e.g.* Magee *et al.,* 2006). A first result from this modelling work is that simulations with constant α yield either excessively high ice particle number densities if $\alpha \ll 1$, while $\alpha \approx 0.1$ cannot explain the observed high S_{ice}. Future work will show whether $\alpha = \alpha(S_{ice})$ can overcome this dilemma.

The workshop also prompted a new debate about how and to what degree coatings by foreign molecules on ice surfaces may change the ice vapour pressure. Of course, foreign molecules on ice surfaces may change the kinetics of the ice growth and evaporation, but can they also change the vapour pressure, *i.e.* thermodynamic state? One focus of this discussion, led by **Dan Murphy**, was the so-called Δ-ice, which had been put forth by Gao *et al.,* (2004) as a concept for ice with HNO_3 surface impurities, leading to nitric acid trihydrate (NAT) clusters. This, in turn, may lead to "step-pinning", *i.e.* the growth of the nucleated steps is hindered by the foreign molecules, in the present case by the NAT-clusters. Dan Murphy discussed possible interpretations of Δ-ice: the HNO_3 could lead to a previously unknown phase of ice (neither hexagonal nor cubic), but this in not very likely; the HNO_3 could change the kinetics of growth of hexagonal ice, but this would maintain $S_{ice} > 1$ only in the case of continuous cooling; Δ-ice could also simply be cubic ice, but this would explain at most ~ 11 % of the observed supersaturation. Finally he stressed that HNO_3 can *not* change the relative rates of evaporation and condensation of ice, as this would violate the second law of thermodynamics.

Alexei Korolev concurred with the assessment of how our lack of understanding of ice nucleation and growth rates represents a main obstacle in our understanding of high S_{ice} in ice clouds. However, based on present theoretical understanding he also concluded that the relative humidity in ice clouds must be expected to increase with decreasing temperatures, and that there was no need for new microphysical mechanisms to explain high S_{ice}, a conclusion that was vividly debated.

Eric Jensen analysed CRAVE water vapour and subvisible cirrus observations, revealing the existence of hexagonal particles

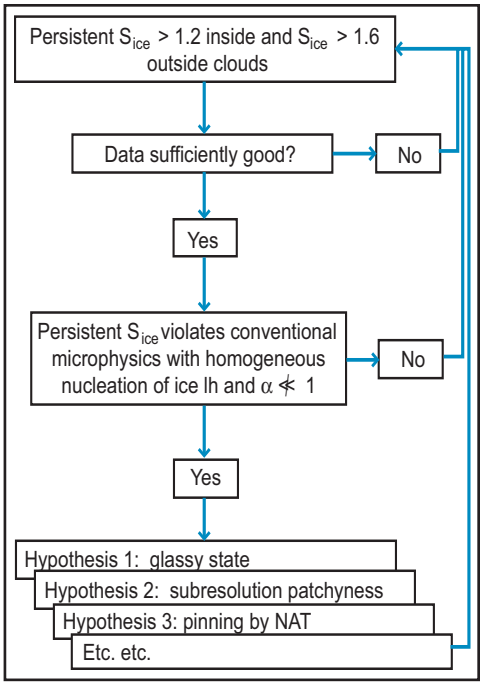

Figure 8. Stream of arguments applied during the workshop.

as large as 100 μm in diameter a few hundred metres below the tropical tropopause. Model calculations suggest that in order to grow such large ice particles extreme supersaturations around $S_{ice} \approx 2$ were required. This latter conclusion would speak for a clear need for new microphysical mechanisms to explain the observed high S_{ice} in cold tropical cirrus clouds. On another day of the CRAVE campaign more conventional ice particles were found and modelled by **Iulia Gensch** using a detailed microphysical box-model forced along backward trajectories. She found best agreement when assuming heterogeneous ice nucleation, relatively little water, and rapid accommodation of water on ice ($\alpha \approx 1$).

Mesoscale models may bridge the gap between global models and backward trajectory analyses. **Federico Fierli** showed that such models with horizontal resolutions in the range of 10-20 km are able to reproduce supersaturation in the recent outflow; however that S_{ice} may be severely underestimated in aged outflow. This may be related to insufficient resolution which does not allow including appropriate physical and small-scale dynamical processes responsible for the maintenance of high S_{ice}. To this end, cloud-resolving models with horizontal resolutions ~ 100 m have an advantage, as was illustrated by **Peter Spichtinger.** High patchiness in cirrus clouds – unresolved by aircraft-borne measurements – could be induced by internal dynamics of cirrus clouds, which may dominate the properties of cirrus clouds (ice number and mass concentrations) including the maintenance of high relative humidities (S_{ice} up to 1.6) inside cirrus clouds.

Finally, **Ulrike Lohmann** widened the perspective by asking to what degree low mass accommodation coefficients (α) would influence cirrus cloudiness in a global circulation model. Preliminary calculations suggest that a low accommodation coefficient (α = 0.005) for H_2O on ice may increase S_{ice} under clear-sky conditions and reduce the overall cirrus cloudiness. Once cirrus clouds form, their crystal number density and mass is increased, thus lowering S_{ice} inside cirrus.

Final discussion and outlook

The rapporteurs, to whom we are most grateful, were **Thierry Corti**, **Thomas Koop**, **Claudia Marcolli**, **Liz Moyer**, **Karen Rosenlof**, **Cornelius Schiller** and **Peter Spichtinger**. **Figure 8** summarizes some aspects of the final discussion. Persistent $S_{ice} > 1.2$ inside and $S_{ice} > S_{nuc}$ (with $S_{nuc} \approx 1.6$ at 200 K) outside clouds were conditions regarded as calling for special attention. However, even after three days of intense discussion there was no general agreement amongst the workshop participants whether or not the measured data were sufficiently good to warrant a call for "unconventional" microphysics. The point that high supersaturations must occur below 200 K based on laboratory evidence was also reiterated, and that therefore there could be little doubt that pronounced upper tropospheric supersaturations must be expected.

Amongst the many possible explanations for the observed high supersaturations discussed during the workshop, formation of a glassy state of the preexisting aerosol or formation of viscous hexagonal/cubic mixtures were emphasised again to explain the lack of nucleation outside of clouds. Mesoscale subresolution patchiness, step pinning by HNO_3 deposition on ice forming NAT, low mass accommodation of H_2O on ice, and cubic ice were seen as most promising hypotheses to explain the lack of H_2O uptake inside of cirrus clouds.

The workshop was closed by an open planning meeting led by **Harald Saathoff**, **Cornelius Schiller** and **Volker Ebert**, for an AIDA water vapour intercomparison campaign. There was broad agreement that such an instrument intercomparison could bring us a good step forward, even if the intercomparison conditions are different from those onboard an aircraft or a balloon. Aims are to determine the instrument performances for static conditions (constant p, T, H_2O), dynamic conditions (changing parameters), with and without clouds. In the meantime the SPARC-cosponsored AquaVIT campaign (Aqua Validation and Instrument Tests) took place, 8-26 October 2007, Karlsuhe, Germany. Amongst the participating instruments were all the WB-57, Geophysica and balloon instruments mentioned above plus several others. The experiments were coordinated by Harald Saathoff, Cornelius Schiller and Volker Ebert and run as blind tests, supervised by **David Fahey** and **Ru-Shan Gao** from NOAA, Boulder, and by **Ottmar Möhler** from Forschungszentrum Karlsruhe who acted as referees. First intercomparison results are expected to become available in spring 2008.

Acknowledgments:

The international workshop on "Upper Tropospheric Humidity", 12-15 June 2007, Karlsruhe, Germany, has been sponsored and financially supported by:
- WCRP: The World Climate Research Program;
- SPARC: Stratospheric Processes and their Role in Climate (a WCRP core project);
- EUROCHAMP: A European Union FP6 project;
- SCOUT-O3: A European Union FP6 project;
- ETH Zurich: Institute for Atmospheric and Climate Science;
- FJ: Forschungszentrum Jülich, Institute of Chemistry and Dynamics of the Geosphere;
- FZK: Forschungszentrum Karlsruhe, Institute of Meteorology and Climate Research.

We are grateful for their support.

Acronyms:

AIDA: Aerosol Interaction and Dynamics in the Atmophere
AquaVIT: Aqua Validation and Instrument Tests Intercomparison Campaign of Water Vapour Measurement Techniques
CARIBIC: Civil Aircraft for the Regular Investigation of the Atmosphere Based on

an Instrument container
CRAVE: Costa Rica. Aura Validation Experiment
FSSP: Forward Scattering Spectrometer Probe
FLASH-B: Fluorescent Advanced Stratospheric Hygrometer (Balloon version)
HIAPER: High-performance Instrumented Airborne Platform for Environmental Research
MOZAIC: Measurement of ozone, water vapour, carbon monoxide and nitrogen oxides aboard Airbus in-service aircraft
NAT: nitric acid trihydrate
TDL: tuneable diode laser absorption spectroscopy
VCSEL: Vertical-Cavity Surface-Emitting Laser
WAVAS: SPARC Assessment of Upper Tropospheric and Stratospheric Water Vapour

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SPARC water vapour initiative

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In the year 2000, SPARC published its Assessment of Upper Tropospheric and Stratospheric (UTS) Water Vapour (SPARC Report No. 2, WCRP-113, WMO/TD No. 1043), which was coordinated and edited by Dieter Kley, Jim M. Russell III and Celine Phillips. The key topic addressed in this report was the analysis and the assessment of the long-term changes of UTS water vapour, with an emphasis on the observed increase of water in the stratosphere. The report had a strong focus describing and comparing relevant data sets using in-situ hygrometers and remote sensing instruments from laboratories all over the world in order to create a suitable data set, including historical data back to the 1940s. Data presented is the report are available at the SPARC data centre at http://www.sparc.sunysb.edu/. The distribution and variability of UTS water vapour, the relevant processes, and the impact of the increased water vapour on radiation, dynamics and chemistry were discussed. However, a quantitative explanation of the analysed changes was not possible in 2000.

Following the recommendations of this report, climatological measurement programmes have continued, new campaigns to investigate UTS water vapour have been carried out, new satellite observation programmes have been launched, and many model and laboratory studies have been made since 2000 to explain the observations and to identify previously unknown processes. Emerging from the new observations, an additional “puzzling” question became apparent in that unexpected high relative humidities were observed, largely in the cold tropopause region both inside and outseide of clouds (see contribution by Peter, Krämer and Möhler, this issue). Data quality, in particular knowing the absolute accuracy and not simply the relative discrepancies between different sensors, has become crucial issue if were are to assess these questions. These accuracy issues have lead to of the need to cross validation of established and recently developed hygrometers, both in the field and in the laboratory.

In light of these developments, it seems timely to update the SPARC water vapour assessment of 2000. In particular, there is a need to summarise the relevant results over