Summary

The development of a detailed understanding of ice clouds in the atmosphere relies on the combined use of field studies, modelling at a multitude of scales, and laboratory studies that provide the necessary fundamentals. Atmospheric ice is studied by remote sensing methods from the ground, and from airplanes and satellites, using passive spectroscopic and light-scattering methods and active methods such as radar and lidar. In the Troposphere, and also, with greater difficulty, in the Stratosphere, ice is studied in situ by using airborne platforms: aircraft and balloons. In situ measurements in the mesopause region are achieved with rocket-borne instrumentation and are limited to brief sampling times as the rockets ascend and descend through cloud layers. These various methods typically lack sufficient access to fundamental physicochemical parameters of ice particles. Furthermore, the representativeness of these types of studies is always an issue because of the transient character of the involved atmospheric processes. Off-line analysis of collected samples may clarify some aspects, but usually fails for metastable particles or when aging processes are important. In these cases laboratory studies may help. Selected experiments can be performed under well controlled conditions to achieve deeper understanding of underlying processes, e.g. nucleation. Furthermore, under these controlled conditions the impact of individual parameters on the ice formation process can be determined. Theoretical and numerical models are then required to transfer the knowledge of laboratory and field studies into large-scale models using sensible parameterizations.

Ice cloud nucleation is currently insufficiently understood. Ice Clouds form by different mechanisms from water vapour or liquid droplets. In many cases the ice particles cannot form from water alone. Instead some aerosol particles are known to provide surfaces on which the nucleation process is catalysed. These aerosol particles are omnipresent in the atmosphere and can act as ice nuclei, i.e. they help to reduce the height of the nucleation barrier forming stable ice crystals. Many different kinds of aerosol exist, and even individual particles vary strongly. Additionally, most atmospheric particles are internally mixed. The impacts of the aerosol nature on ice nucleation efficiency, ice microstructure and dynamics are one of the least understood parameters in cloud microphysics. The knowledge of chemists, biologists and crystallographers about the aerosol composition has to be combined with the ice dynamic models of physicists, meteorologists and computational modellers to gain a better understanding of the whole process. Only then the global impact of microstructure and dynamics of ice on e.g. the water cycle or the radiation budget can be determined accurately. The ESF research networking programme on the Micro-Dynamics of Ice has provided an ideal platform for such an intersection between the different communities and provided the capability to bring together scientists from the different fields of ice research.

Description of the scientific content and discussion

Short summary of the Session *Laboratory Measurements I*

The first session of the workshop on 'Atmospheric Ice Nucleation' deals with laboratory measurements of ice nucleation properties of various particle types occurring in the atmosphere. First, Bernhard Pummer presented a study on the impact of stress on the IN activity of pollen and fungal spores. His results are for example that starvation of IN increases their activity, while pollution leads to a decrease of ice formation. The latter result might be important especially at Northern latitudes. Next, Lukas Kaufmann compared the ice nucleation behaviour of birch pollen and Hoggar mountain dust for different temperatures. His studies show that only very few best sites on the surface of the birch pollen and mountain dust are responsible for freezing at the highest observed temperatures. As third speaker, Daniel O'Sullivan presented a review of laboratory studies on ice nucleation by a variety of particles (mineral dusts, biological species such as pollen, bacteria, fungal spores and plankton, carbonaceous combustion products and volcanic ash) immersed in supercooled cloud drops. He summarizes that ice nucleation below about -15 °C is dominated by soot and mineral dusts and that above this temperature the only biological particles nucleate ice. Finally, Emiliano Stopelli studied the ice nucleation of biological particles at tropospheric cloud heights with temperatures > -12 °C using atmospheric rain and snow samples from the High Alpine Research Station of Jungfraujoch in the Swiss Alps. The IN abundance in the samples was higher for autumn than for winter samples and southern clouds showed IN concentrations two times higher than clouds coming from the north, but altogether no simple relation between meteorological parameters and IN abundance was found. There was a lively General Discussion at the end of the session, starting with contributions on the variety of feldspar particle compositions occurring in the atmosphere, all showing differing freezing behaviours. Further, the importance of ice nucleation of soot and organic particles are discussed. It was then mentioned that all the studies on ice nucleation properties of atmospheric particles seems not to lead to a consistent picture, but quite in contrary result in a more and more diverse picture which makes it difficult to derive parameterizations of heterogeneous freezing of atmospheric particles for use in process or global models. It was then discussed that this is a similar stage of research than in the last decade for cloud condensation nuclei, which finally leads to the now established Kappa theory. The final discussion was if it is necessary to know all the details of heterogeneous ice nucleation for a better climate forecast or if cloud dynamics play a more important role in ice formation than the aerosol particle composition. With this open question the first session of the workshop was closed.

Short summary of the Session *Laboratory Measurements II*

The main objective of this session was investigation of freezing properties using laboratory experiments. This includes not only classical experiments on single particles but also investigations in huge freezing chambers as the AIDA. In the first talk Kristina Hoehler presented investigations of freezing kinetics on solution droplets as measured in the AIDA chamber. For this purpose, freezing experiments were carried out in order to investigate homogeneous nucleation rates; since homogeneous freezing of aqueous solution droplets is the main pathway for ice production at low temperatures, it is of high interest to know the rates in details. The measurement shows some discrepancies to earlier laboratory measurements as well as to the well-known parameterisation by Koop et al. (2000). In the discussion, the reason for these disagreements could not be determined, although several error sources were discussed. The second talk of the session was held by Naruki Hiranuma. Experiments on heterogeneous freezing of clay minerals and bacteria, carried out in the AIDA chamber were presented; in this case the investigated freezing pathway was immersion freezing. These experiments showed good agreement with former studies at LACIS. As in these studies, the results showed a strong dependence on different types of mineral as well as temperature dependence. The reason for the variability is not known; in the discussion there were some arguments about the importance of surface properties on ice nucleation ability. In the next talk, Nadine Hoffmann presented experiments on contact freezing. The experiments were carried out using levitated droplets in an electromagnetic balance, which are hit by aerosols in a laminar flow. For contact freezing, the collision rates are of high importance; however, it is very difficult to measure these rates. The results showed a strong size dependence of the freezing probability; thus, the contact freezing probability is proportional to the surface area of the droplets. Since, it is quite difficult to distinguish between immersion freezing and contact freezing; several suggestions were made during the discussion in order to investigate the details of this process. In the last talk by Erik Thomson the focus was on the issue of organic coatings and their impact on ice nucleation at low temperatures, i.e. in the cirrus regime. Experiments, using molecular and light scattering were presented. Using these techniques, it is possible to determine the surface coverage, thus the onset of ice nucleation can be determined very precisely. The experiments showed a strong impact of these coating on ice nucleation. However, the results also show surprisingly high ice supersaturation values as thresholds for ice nucleation. This issue was discussed, however without any final conclusion, although it was assumed that kinetic effects might play a role.

Short summary of the Session *Laboratory Measurements III*

The first talk was given by Stefanie Augustin presenting ice nucleation rates of single protein complexes and single macromolecules. Many biological aerosols are extremely good ice nuclei, which are active also at temperatures warmer than -20°C. While mineral dusts are potential ice nuclei at colder temperatures. This biological ice nucleation activity is independent of the fact if these species are dead or alive, but are related to proteins or other macromolecules on or in the outer surface. The so-called CHESS model gives access to the respective ice nucleation rates. In the case of birch pollen two different ice nucleation active macromolecules have been found showing different nucleation rates. Additionally, it was stated that the number of macromolecules is always much larger than the number of mother pollen grains, which multiplies the potential impact of these nuclei.

The same birch pollen samples have also been investigated by Monika Kohn et al. in the Zurich Ice Nucleation Chamber (ZINC) and in its immersion freezing extension (IMCA). This group has reproduced the results of Augustin et al. with their set-up and have also investigated pollen washing water, which models the fact that pollen grains exhibit so-called bursting which means that they spread their inner content out into the environment. In order to monitor these nuclei, a field instrument has been developed for studying immersion freezing also outside the laboratory.

The lower size limit for mineral dust ice nuclei in the immersion freezing mode has been investigated by Andre Welti et al. The lower limit was found at about 100 nm. This is astonishing, since standard literature (Pruppacher&Klett) predicts only a small impact of the size of the ice nuclei on immersion freezing. A logarithmic dependence of the medium freezing temperature on surface area was found. The cut off was close to 100 nm and was depending on chemistry. However, evidence for active sites on the mineral dust particles was not adduced. Important questions were, if the phase composition of the dust is size dependent and what is the best way to determine the surface area of the dust, i.e. geometric area versus specific area.

The last talk of the session was given by Riccardo Iannarelli concerning the phase diagram HCl/H_2O. This talk was very much related to Polar Stratospheric Clouds. Therefore the overlap with the other talks was very restricted and so was the discussion. The discussion was about the kinetics of the film formation. The activation energy is about 20 kJ mol-1 and is proportional to the surface diffusion energy and mediated growth. Thus, the growth of the hydrates is transport limited.

Short Summary of the Session *IN Measurements*

The first talks presented ground based measurements of tropospheric aerosols, relating ice nuclei concentrations with carbonaceous aerosols. The first presentation (by Christina McCluskey) characterized ice nuclei concentrations formed during biomass burning (wildfires and fires in the lab), while the second presentation (by Yutaka Tobo) was focused on primary biological particles and developed a parameterization of ice nuclei populations including these particles.

The latter presentations described measurements of cirrus clouds. Investigations of Anna Luebke were based on campaigns (air plane measurements) and presented the importance of the updraft velocity in determining cirrus microphysical properties. Long term observations of cirrus clouds using the lidar technique were presented by Christian Rolf and yielded a cirrus climatology being representative for mid-latitude cirrus.

Short summary of the Session *IN Characterisation*

This began with Jessica Meyer (Institute for Energy and Climate Research, Germany) describing the occurrence of mixed-phase clouds at mid- and high latitudes. The mixed-phase ice fraction was shown to be determined by ice nucleus (IN) concentrations and the intensity of in-cloud ascent. Mixed-phase clouds were shown to be much more common than ice-only clouds.

The next talk in the session was by Janine Frohlich-Nowoisky (MPI, Germany). This described differences in fungal composition globally, showing contrasts between continental interiors and coastal regions. Discovery of new fungal IN species in air sampled with a filter was presented. These fungal IN nucleated ice at temperatures as warm as -5 degC. The emission of PBAPs after rainfall was observed, with possible release of fungal spores.

The talk by Pierre Amato (Clermont University, France) showed results from another laboratory analysis of PBAPs sampled from the atmosphere. The focus of this talk was on the bacterial species present in the sample, which was from cloud-water. Only 7 out of a total of about 250 strains of bacteria in the sample were observed to be ice-nucleating, as described in the paper by Joly et al. (2013, Atmos. Env). This fraction (3%) matches well with the fraction assumed by the ice-nucleation scheme by Phillips et al. (2008, 2013). The bacterial IN were mostly of the Pseudomonas syringae (Ps) species, though a few other species were also observed. Acidity of water was found to control the activity of these bacterial IN.

The last talk of the session was by B. Sierau (ETH Zurich, Switzerland) who showed ways to create mass-spectrometry 'fingerprints' for field observations of Ps and other PBAPs. The material washed from various spores and pollen was studied for ice nucleation.

Short summary of the Session *IN Modelling*

In the first talk of the session Dennis Niedermeier showed a recently developed simplification of a stochastic model for ice nucleation, i.e. a simplification of the original soccer ball model. In general, it is still not clear if heterogeneous nucleation is a stochastic process or even a deterministic process. From experiments it is difficult to decide the nature of heterogeneous nucleation. As shown in former studies by developing the original soccer model, the authors could show that heterogeneous nucleation can be described by a combination of both models. Since the soccer ball model is quite complicated and expensive in terms of computing time, the authors have developed a simpler version, which again can represent the main features of ice nucleation. The discussion concentrated on some features of the model, especially fitting the parameters. In the second talk of the session Ross Herbert presented a model for modelling heterogeneous ice nucleation, the so-called multiple component stochastic model (MCSM). It was used to determine the impact of nucleation site distributions and temperature dependence of the nucleation rate on the temperature dependence of the ice nucleation ability. Actually, only the temperature dependence of the nucleation rate seems to control temperature dependent nucleation. Additionally, the freezing model was extended for describing single freezing events in continuous flow diffusion chambers and single ice nucleation experiments. In the discussion, some details of the pure stochastic model approach in contrast to the talk before were discussed. Luisa Ickes presented a recently developed ice nucleation parameterisation for the use in large scale models. For this purpose, data from ice nucleation experiments in the laboratory were used to determine realistic nucleation in coarse models. The main focus is here on mineral dust aerosols, acting as heterogeneous ice nuclei in mixed-phase clouds. In the discussion, the test of the model against independent nucleation experiments was suggested as well as a closer look into the physical constraints for the parameterisation. Finally, Matthias Voigt showed large eddy simulations of Kelvin-Helmholtz instabilities in cirrus clouds and the impact of heterogeneous ice nuclei. For the cold temperature regime of cirrus clouds ($T<235K$) homogeneous freezing of aqueous solution droplets is dominant and triggered by local dynamics. Thus, one would expect that heterogeneous IN should not play a role. However, even in the high velocity regime, heterogeneous ice nucleation is able to modify homogeneous nucleation and the microphysical properties of cirrus clouds. In the discussion the low ice crystal number concentrations in the simulations were discussed; this might be an effect of highly fluctuating velocities.

In the final discussion, the use of laboratory experiments for modelling clouds containing ice particles was discussed. Although the basic research on the basic procedure of nucleation is very important for our general understanding, for modelling clouds, the point of view is of high importance. While for coarse resolution models, less detailed models might be appropriate, for simulating very small scales, even more detailed models seems to be necessary. However, there is still lack of understanding of ice nucleation on a very basic level. Actually, we do not understand ice nucleation on a molecular basis; thus, it is important to come up with new theoretical concepts and approaches in combination with measurements on a molecular level.

Impact on future directions of the field

There was a very lively discussion after every talk and after every session and also the final discussion was very fruitful. Important clues have been made by several senior scientists, who have not given own talks. Obviously, there is still a large lack of information concerning the identity of nucleation sites. Only in a few cases, e.g. for Snomax, we have detailed information concerning the chemical signature of these sites. Current models often work with an abstract description of these sites summarizing them e.g. as a contact angle of a facet. This model might be suitable for mineral dust, but poses a problem for organic and biological nuclei.

It was very impressive to see, how many different setups and techniques have been developed in order to investigate the ice nucleation activity of all kinds of aerosols. Although the general knowledge has massively increased during the last years thanks to these measurements, the progress in the understanding of the global atmosphere is still considered to be insufficient. It is of high importance to combine all the information acquired from the laboratory measurements in order to deduce relevant information for climate modelling. It is necessary for theorists and modellers to define, which parameters they really need for their calculations, in order to extract the relevant information out of the data from laboratory experiments and field measurements. This is quite a controversial attempt: On the one hand, a simplification makes calculations easier and different sets of data better comparable, but on the other, it leads to loss of information and to abstraction. For understanding of cloud condensation nucleation, for example, the kappa value was introduced as one simple parameter, although it is considered not satisfactory for every purpose. It is debatable, if an analogous parameter for ice nucleation is both possible and sensible (see contact angle above).

Most essential is the characterization of the decisive properties that make a particle an ice nucleus, since it will allow sensible estimations of ice nuclei generation and distribution in the geosphere. Methods of chemical surface characterization show a high potential to solve this problem in the future.

Annexes

Annex 1: Program of the meeting

Annex 2: Full list of speakers and participants

Annex 1: Program of the meeting

EUROPEAN SCIENCE FOUNDATION

Workshop - Atmospheric Ice Nucleation

Vienna; Austria

6th and 7th of April 2013

Book of Abstracts

TECHNISCHE UNIVERSITÄT WIEN
Vienna University of Technology

Preface

Dear Participant!

I may welcome you to Vienna and I wish you a good time at our workshop. The aim of this workshop is bringing together three communities of atmospheric ice research: field measurements, laboratory studies and modelers. The joint topic is the ice nucleation in clouds. We will focus on methods and open questions concerning the microstructure and dynamics of ice formation processes in the atmosphere discussing experimental and theoretical methods including chemistry and microphysics. A particular focus will be on heterogeneous ice nucleation by biological particles.

Additionally, the organizers of this workshop also offer a session at the EGU conference, which is called “Atmospheric Ice Particles” AS 3.5 and has a tradition of already five years. However, there is no overlap between the workshop and the conference session, since it is the main aim of the workshop to provide a deeper discussion forum for one subject, i.e. ice nucleation, which cannot be discussed at the conference in necessary details. At the workshop, particularly young scientists will be given the opportunity to talk and to present their recent research while senior scientists will be asked to give overviews and questions.

Vienna 6th April 2014

Hinrich Grothe

Organizers

Local Organizers

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Institut für Energie und Klimaforschung (IEK)
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Location

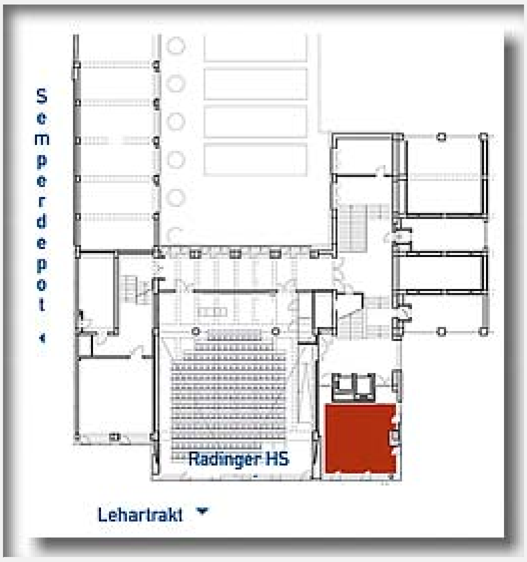
Radinger Hörsaal, Vienna University of Technology

The workshop is held in the Radinger Hörsaal (lecture hall) at the Vienna University of Technology, Getreidemarkt 9, 1060 Vienna. this lecture hall is next to the Institute of Materials Chemistry and is situated in the centre of Vienna only a few yards away from the Karlsplatz and the Naschmarkt, which are well-known tourist attractions.

Radinger Hörsaal (Lecture Hall)

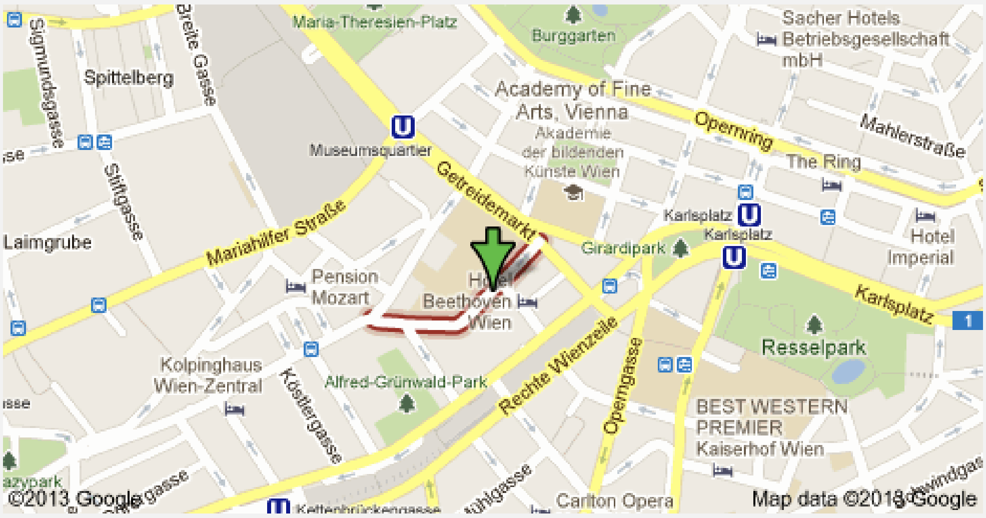
Radinger Hörsaal, Getreidemarkt 9, 1060 Vienna, Austria

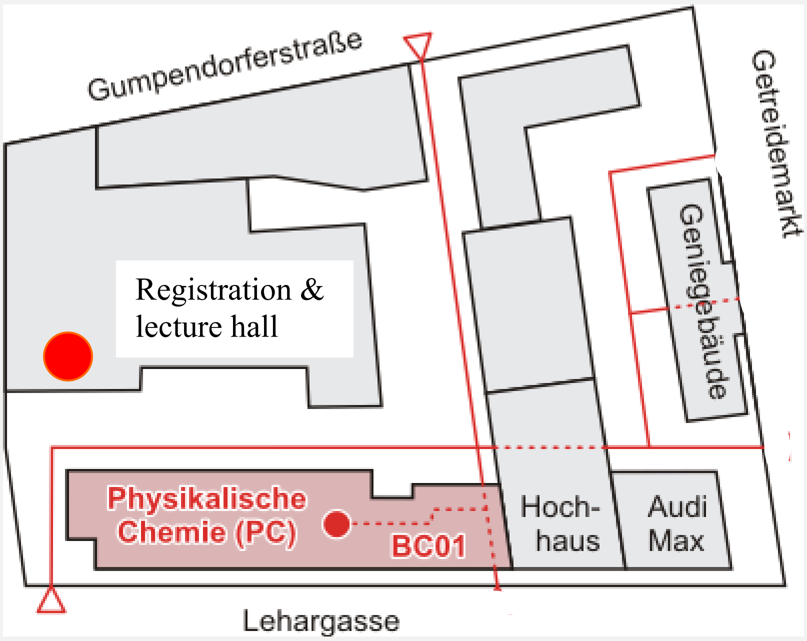
Lecture Hall



Map of Lecture Hall / registration (red square)

City Map Vienna





Sessions

	Saturday 6th April		
11:30 - 12:30	Registration and Buffet		
12:30 - 12:45	Opening of the workshop	Hinrich Grothe	

	Laboratory Measurements I		
12:45 - 12:55	1st Talk	Bernhard Pummer	Stress treatment of pollen and fungal spores - what are the impacts on IN activity?
12:55 - 13:00	Discussion		
13:00 - 13:10	2nd Talk	Lukas Kaufmann	Comparison of ice nucleation between birch pollen and Hoggar mountain dust
13:10 - 13:15	Discussion		
13:15 - 13:25	3rd Talk	Daniel O'Sullivan	Ice Nucleation By Particles Immersed In Supercooled Cloud Droplets: A Review Of Laboratory Studies
13:25 - 13:30	Discussion		
13:30 - 13:40	4th Talk	Emiliano Stopelli	Biological ice nucleation at tropospheric cloud heights
13:40 - 13:45	Discussion		
13:45 - 14:00	General Discussion		
14:00 - 14:20	Coffee Break		

	Laboratory Measurements II		
14:20 - 14:30	1st Talk	Kristina Hoehler	AIDA cloud expansion chamber experiments on freezing kinetics of solution droplets
14:30 - 14:35	Discussion		

14:35 - 14:45	2nd Talk	Naruki Hiranuma	Immersion Freezing of Clay Minerals and Bacterial Ice Nuclei
14:45 - 14:50	Discussion		
14:50 - 15:00	3rd Talk	Erik S. Thomson	Organic coatings, ice nucleation, and water uptake
15:00 - 15:05	Discussion		
15:05 - 15:15	4th Talk	Nadine Hoffmann	Contact freezing of supercooled cloud droplets on collision with mineral dust particles: effect of particle size
15:15 - 15:20	Discussion		
15:20 - 15:35	General Discussion		

15:35 - 15:55	Coffee Break		

	Laboratory Measurements III		
15:55 - 16:05	1st Talk	Steffi Augustin	Ice nucleation rates of single protein complexes and single macromolecules
16:05 - 16:10	Discussion		
16:10 - 16:20	2nd Talk	Monika Kohn	Immersion freezing of biological aerosol particles in the ZINC setup
16:20 - 16:25	Discussion		
16:25 - 16:35	3rd Talk	Welti André	Is there a lower size limit for mineral dust ice nuclei in the immersion mode?
16:35 - 16:40	Discussion		
16:40 - 16:50	4th Talk	Iannarelli Riccardo	Thermodynamics and Kinetics of the low-temperature HCl/H_2O Phase Diagram Revisited
16:50 - 16:55	Discussion		

16:55 - 17:10	General Discussion

17:15	Opening of the Buffet and Wine Reception

Sunday 7th April

	Atmospheric IN Measurements		
08:15 - 08:25	1st Talk	Christina McCluskey	The production and characteristics of ice nuclei from biomass burning in the US
08:25 - 08:30	Discussion		
08:30 - 08:40	2nd Talk	Yutaka Tobo	Temperature dependence of the relation between ice nuclei (IN) and primary biological aerosol particles (PBAPs) at a forested site in Colorado
08:40 - 08:45	Discussion		
08:45 - 08:55	3rd Talk	Anna E. Luebke	The relationship of cirrus ice water content, crystal number, and size to vertical velocity: First results from the MACPEX field campaign
08:55 - 09:00	Discussion		
09:00 - 09:10	4th Talk	Christian Rolf	A mid-latitude cirrus lidar climatology with thin cirrus clouds in the lowermost stratosphere
09:10 - 09:15	Discussion		
09:15 - 09:30	General Discussion		

09:30 - 09:50	Coffee Break		

	IN Characterization		
09:50 - 10:00	1st Talk	Jessica Meyer	Ice and liquid partitioning in mid-latitude and artic mixed-phase clouds: how common is the real mixed-phase state
10:00 - 10:05	Discussion		
10:05 - 10:20	2nd Talk	Janine Fröhlich-Nowoisky	New species of ice nucleating fungi in soil and air

10:20 - 10:25	Discussion		
10:25 - 10:35	3rd Talk	Pierre Amato	Microbial ice-nucleators in cloud water at the puy de Dôme (France)
10:35 - 10:40	Discussion		
10:40 - 10:50	4th Talk	Sierau Berko	Towards the identification of individual biological particles as ice nuclei using single particle mass spectrometry
10:50 - 10:55	Discussion		
10:55 - 11:05	General Discussion		

11:05 - 11:25	Coffee Break		

	IN Modelling		
11:25 - 11:35	1st Talk	Denis Niedermeier	Introduction of a simplified version of the Soccer ball model
11:35 - 11:40	Discussion		
11:40 - 11:50	2nd Talk	R. J. Herbert	A simple method for including cooling rate dependence in a deterministic ice nucleation model
11:50 - 11:55	Discussion		
11:55 - 12:05	3rd Talk	Luisa Ickes	A new freezing parameterization for mixed-phase clouds in ECHAM GCM
12:05 - 12:10	Discussion		
12:10 - 12:20	4th Talk	Matthias Voigt	Impact of different nucleation types on dynamically induced cirrus inhomogeneities
12:20 - 12:25	Discussion		
12:25 - 12:40	General Discussion		

12:40 – 12:45	Closing Remarks	Hinrich Grothe

12:45 - 14:00	Lunch Buffet

13:30 - 13:50	Optional lab tour I
13:40 - 14:00	Optional lab tour II
13:50 - 14:10	Optional lab tour III
14:10	End of the Workshop

Stress treatment of pollen and fungal spores – what are the impacts on IN activity?

B.G. Pummer, L. Atanasova, H. Bauer, J. Bernardi, I.S. Druzhinina, C. Müller, P. Schmitt-Kopplin, K. Whitmore, and H. Grothe

Certain species of pollen and fungal spores have shown significant immersion IN activity in former laboratory studies [Kieft, Pouleur et al., Diehl et al., Pummer et al.]. In contrast to these habitual indoor conditions, bioaerosols in the atmosphere are exposed to numerous stresses, which adulterate them and so might have an impact on their IN activity. Atmospheric particles are exposed to reactive trace gases, extremes in temperature and humidity, high-energetic radiation and electricity (lightning).

We exposed pollen to NO_2, O_3 and UVA light as model stresses to determine their impact on IN activity. Furthermore, we investigated the pollen rupture and material release caused by soaking plus mechanical stress. Fungal strains from different origins were cultivated with and without occasional freezing events to determine the impact on IN activity. However, although the total protein content differed between frozen and non-frozen samples, the IN-activity remained nearly the same. This is in contrast to bacterial IN, which showed higher IN expression, when they were cultivated at lower temperatures [Gurian-Sherman and Lindow].

Additionally, we intensified our investigation on characterizing the pollen ice nuclei. Solid phase extraction with different columns proved to be a useful tool to confine the possibilities of the chemical nature of the ice nuclei. At last, dilution rows of pollen waters were prepared and investigated to determine the concentration dependency of the IN activity. The different datasets of the oil immersion and the LACIS measurements [Augustin et al.] perfectly match, if the concentration differences are considered.

References

Kieft TL: Appl. Env. Microbiol., 54, 1678-1681, 1988

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Diehl K et al.: Atmos. Res., 61, 125-133, 2002

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Comparison of ice nucleation between birch pollen and Hoggar mountain dust

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Depending on the region, biological particles and/or mineral dust can act as ice nuclei. In this study, we compare ice nucleation on a dust collected in the Hoggar mountains in the Sahara desert with ice nucleation in the presence of washing water of birch pollen using a differential scanning calorimeter (DSC). Emulsion as well as bulk measurements were performed. For the emulsion measurements with pollen 50 mg/ml concentrated washing water of birch pollen was mixed with water or aqueous solutions of e.g. ammonium sulfate. For the emulsion measurements with dust, a suspension of the Hoggar mountain dust was mixed with water or aqueous solutions of ammonium sulfate, PEG 300, glucose or malonic acid. Emulsified with a mineral oil/lanolin mixture, the water droplets had a mean diameter of around 2 μm. Emulsions consisting of pure water suspensions froze with onset temperatures of around 237 K.

For the bulk measurements washing water of birch pollen or Hoggar mountain dust was mixed with pure water and droplets with radii of about 1 mm were subjected to repeated freezing cycles.

Emulsions of washed water of birch pollen freeze with onset temperature of about 255 K and Hoggar mountain dust emulsions freeze with onset temperature of about 247 K. In contrast to this, bulk samples of birch pollen and Hoggar mountain dust freeze both at about 260 K.

The difference between the freezing temperatures observed in the emulsion measurements and the bulk measurements gives support for the notion, that only very few best sites on the surface of the birch pollen and Hoggar mountain dust are responsible for freezing at the highest observed temperatures.

Ice Nucleation By Particles Immersed In Supercooled Cloud Droplets: A Review Of Laboratory Studies

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b School of Chemistry, University of Leeds, UK.

The formation of ice particles in the Earth's atmosphere strongly affects the properties of clouds and their impact on climate. Despite the importance of ice formation in determining the properties of clouds, the Intergovernmental Panel on Climate Change (IPCC, 2007) was unable to assess the impact of atmospheric ice formation in their most recent report because our basic knowledge is insufficient. Part of the problem is the paucity of quantitative information on the ability of various atmospheric aerosol species to initiate ice formation. Here we review and assess the existing quantitative knowledge of ice nucleation by particles immersed within supercooled water droplets. We introduce aerosol species which have been identified in the past as potentially important ice nuclei and address their ice-nucleating ability when immersed in a supercooled droplet. We focus on mineral dusts, biological species (pollen, bacteria, fungal spores and plankton), carbonaceous combustion products and volcanic ash. In order to make a quantitative comparison we first introduce several ways of describing ice nucleation and then summarise the existing information according to the time-independent (singular) approximation. Using this approximation in combination with typical atmospheric loadings, we estimate the importance of ice nucleation by different aerosol types. According to these estimates we find that ice nucleation below about -15 °C is dominated by soot and mineral dusts. Above this temperature the only materials known to nucleate ice are biological, with quantitative data for other materials absent from the literature. We conclude with a summary of the challenges our community faces.

Biological ice nucleation at tropospheric cloud heights

Emiliano Stopelli[1], Franz Conen[1], Christine Alewell[1], Cindy Morris[2]
1: Dept. Umweltgeowissenschaften, Universität Basel, CH
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Biological ice nucleators (IN) are the most abundant agents to catalyse ice formation at warm temperatures (> -12 °C). Yet, the relevance of biological ice nucleation for cloud processes, such as initiating precipitation, remains ambiguous. Moreover, very little is known about abundance and nucleation spectra of these IN at tropospheric cloud altitudes. Even if warm IN concentrations tend to be lower than other IN, this does not mean that biological IN do not play a potentially important role in clouds: a few initial ice particles can multiply in number by orders of magnitude through the process of riming and ice splintering and thereby initiate precipitation. Equally unknown is the relative importance of different kinds of biological IN in this part of the atmosphere, their likely change with seasons, with weather conditions and air mass origin.

The purpose of this project is then to understand meteorological conditions and environmental factors associated to the presence of biological ice nuclei in precipitation. Rain and snow samples are collected at the High Alpine Research Station of Jungfraujoch in the Swiss Alps, 3580 m above sea level, as representative of tropospheric cloud heights. Concentration of IN is determined by a cooling bath apparatus with an innovative system of automatic recording of freezing events, developed in order to improve the detection of the phenomenon. This information is then compared with meteorological and modelling data available for Jungfraujoch Station, to obtain a better understanding of the conditions associated to warm IN abundance. A description of the results obtained from autumn 2012 and winter 2013 sampling campaigns will be provided in the presentation. Half of the samples showed the onset of freezing at -6 °C, with some samples collected during December characterized by the presence of IN active at -3.5 °C. IN abundance seems to be linked to the origin of the clouds, with southern clouds showing IN concentration median values two times higher than clouds coming from the north, both at -8 and -11 °C.

Future assays will be dedicated to determining the abundance of microbial communities, through direct counting of bacteria through microscopy. Further tests will be conducted to identify and differentiate biological particles responsible for nucleation at warm temperatures. In case of a significant microbial presence in the samples, subsequent isolation of *Pseudomonas syringae*, known as efficient ice nucleator, will be carried out.

AIDA cloud expansion chamber experiments on freezing kinetics of solution droplets

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The AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber has been built to enable investigations of atmospheric relevant aerosol processes under controlled laboratory conditions. It consists of a large 84 m³ aluminum vessel which is filled with synthetic air and the aerosols to be investigated. A broad range of instruments enables the simultaneous measurement of temperature, pressure, water content, aerosol concentration and size distribution. The starting temperature can be chosen from ambient to as low as - 90°C, while the humidity at starting conditions for cloud studies is usually set by a partial ice cover on the chamber walls. The updraft of an air parcel with its drop in temperature and pressure and the accompanying rise in relative humidity is mimicked by evacuation of the chamber. During the expansion, the temperature, pressure and water content are monitored constantly. The formation of droplets and/or ice is measured using optical particle counters (welas) and Fourier Transform Infrared Spectroscopy. Particle habit and phase can be additionally investigated by forward to backward light scattering and backscattering linear depolarization ratio.

We present a compilation of results on the freezing behavior of solution droplet aerosols gathered in a series of AIDA campaigns. Due to the ongoing discussion on their importance in cirrus and polar stratospheric cloud formation, we lay a special focus on sulfuric acid solution droplets in the temperature range of - 80°C to - 40°C. We report nucleation onset temperatures and concentrations and compare them to literature data. Also, we give an estimate for observed nucleation rates and set them into the context of available nucleation rate formulations.

Immersion Freezing of Clay Minerals and Bacterial Ice Nuclei

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The immersion mode ice nucleation efficiency of clay minerals and biological aerosols has been investigated using the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber. Both monodisperse and polydisperse populations of various clay dust samples as well as Snomax® (a proxy for bacterial ice nucleators) and hematite are examined in the temperature range between -4 ˚C and -35 ˚C. The temperature dependence of ice formation inferred by the INAS (Ice Nucleation Active Surface-Site) density is investigated and discussed as a function of cooling rate and by comparing to predicted nucleation rates (i.e., classical nucleation theory with θ-probability density function nucleation scheme). To date, we observe that maintaining constant AIDA temperature does not trigger any new ice formation during the immersion freezing experiments with clay dust samples and Snomax®, implying strong temperature dependency (and weak time dependency) within our time scales and conditions of experiments. Ice residuals collected through a newly developed pumped counter-flow virtual impactor with the 50% cut size diameter of 10 to 20 μm have also been examined by electron microscope analyses to seek the chemical and physical identity of ice nuclei in clay minerals. In addition to the AIDA results, complementary measurements with mobile ice nucleation counters are also presented.

Organic coatings, ice nucleation, and water uptake

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Water interactions with organic surfaces are of central importance to both biological and geophysical processes. In the atmosphere the hygroscopicity of primary and secondary aerosols, and the formation and lifetime of cloud droplets and ice particles can be inuenced by organic coatings. Here we report findings from molecular and light scattering experiments that investigate ice nucleation and growth properties at cirrus cloud temperatures. In particular we focus on deposition nucleation and water uptake by ice surfaces and how these processes are altered by the presence of organic surfactants. Simple organic adsorbates like methanol and butanol are observed to inuence absolute nucleation rates and water accommodation and lead to higher-than-expected required supersaturations for nucleation. The implications for atmospheric systems that include more complex volatile compounds are discussed.

Contact freezing of supercooled cloud droplets on collision with mineral dust particles: effect of particle size

Nadine Hoffmann, Denis Duft, Alexei Kiselev and Thomas Leisner

The contact freezing of supercooled cloud droplets is one of the potentially important and the least investigated heterogeneous mechanism of ice formation in the tropospheric clouds [1]. On the time scales of cloud lifetime the freezing of supercooled water droplets via contact mechanism may occur at higher temperature compared to the same IN immersed in the droplet. However, the laboratory experiments of contact freezing are very challenging due to the number of factors affecting the probability of ice formation. In our experiment we study single water droplets freely levitated in the laminar flow of mineral dust particles acting as the contact freezing nuclei. By repeating the freezing experiment sufficient number of times we are able to reproduce statistical freezing behavior of large ensembles of supercooled droplets and measure the average rate of freezing events. We show that the rate of freezing at given temperature is governed only by the rate of droplet –particle collision and by the properties of the contact ice nuclei. In this contribution we investigate the relationship between the freezing probability and the size of mineral dust particle (represented by illite) and show that their IN efficiency scales with the particle size [Fig. 1]. Based on this observation, we discuss the similarity between the freezing of supercooled water droplets in immersion and contact modes and possible mechanisms of apparent enhancement of the contact freezing efficiency.

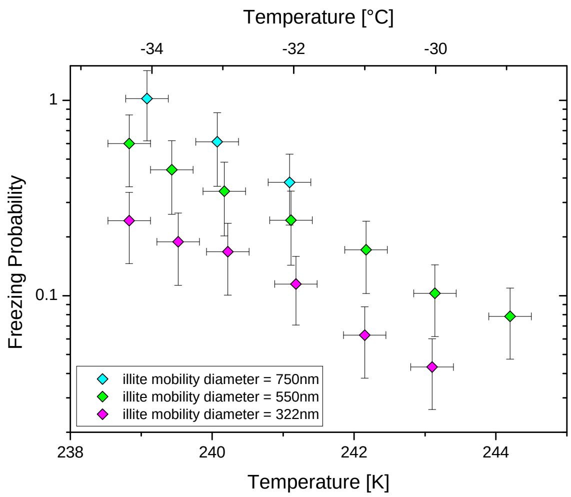

Fig. 1: Freezing probability as a function of temperature and particle size

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Ice nucleation rates of single protein complexes and single macromolecules

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Biological particles, such as some bacteria and pollen species, are known to nucleate ice at much higher temperatures than non-biological particles, e.g. mineral dust. In the case of bacteria, ice nucleation active (INA) protein complexes connected to the bacteria's outer membrane are controlling the ice nucleation. Pollen feature INA macromolecules which induce ice nucleation (Pummer et al., 2012). For the investigation of the immersion freezing behavior of these biological ice nucleating entities, we performed measurements with size-selected particles generated from a SNOMAX® solution/suspension in water, and birch pollen washing water (BW in the following), at the laminar diffusion chamber LACIS (Leipzig Aerosol Cloud Interaction Simulator, Hartmann et al., 2011). Ice fractions were measured, as function of temperature and number of INA entities, i.e., either INA protein complexes or INA macromolecules.

For SNOMAX®, which is an artificial product consisting of pseudomonas syringe bacteria, we found immersion freezing already to occur at temperatures around -7 °C. In the case of BW we observed freezing at temperatures slightly higher than -20°C. After an initial steep increase with decreasing temperature, the ice fraction stayed constant (saturation behavior) down to -35°C, for both examined materials. Based on this saturation behavior we were able to show, that the distribution of entities over the droplet ensemble follows a Poisson distribution. Using the so-called CHESS (stoCHastic modEl of similar poiSSon distributed ice nuclei, Hartmann et al., 2012) model, a combination of the Poisson distribution and a simple stochastic nucleation approach, the derivation of the ice nucleation rates for both, single INA protein complexes and single INA macromolecules are possible. The determined nucleation rates together with a reasonable number of INA entities can be used for describing biological IN agents related ice nucleation in atmospheric models. As an additional peculiarity, we observed two different INA macromolecules for Birch trees growing in central Europe and Northern Europe, with the latter initiating freezing at slightly higher temperatures.

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Immersion freezing of biological aerosol particles in the ZINC setup

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Biological aerosol particles, such as pollen and fungal spores, are constituents of the atmospheric aerosol population. Besides their health impacts, some of these particles are found to be active as ice nuclei (IN) for heterogeneous freezing and can thus initiate ice formation in clouds. This effect could lead to changes in cloud properties, thereby influencing earth's radiation budget and hence could also have an impact on our climate.
Pollen are amongst the largest aerosol particles found in the atmosphere. Since IN activity is considered to increase with particle size, these aerosol types are more likely to act as IN. In mixed-phase clouds ice nucleation in the immersion freezing mode is of major importance, because liquid cloud droplets are necessary for heterogeneous nucleation at warmer temperatures; in particular biological aerosol particles were found to have the ability to act as IN at relatively warm temperatures [1]. In the same study pollen have been investigated to act as efficient IN for freezing modes where the liquid phase was involved.
In this study ice nucleation experiments with the Zurich Ice Nucleation Chamber (ZINC, [2]) and its immersion freezing extension IMCA (Immersion Mode Cooling Chamber, [3]) has been performed to investigate the IN ability of different pollen species in the immersion freezing mode. It has previously been reported [4, 5] that also water, in which pollen had been emulsified and subsequently removed by filtration, may contain IN. We also tested these so called pollen washing waters. For this purpose the washing water was nebulized by an atomizer and the produced droplets were dried and size selected for ZINC/IMCA measurements. First results show good agreement with previous measurements of droplet residues produced from pollen washing waters in terms of their IN-activity [4].
To gain further insights into the IN properties of pollen in the atmosphere, we develop a field instrument to study immersion freezing outside the laboratory. The presented data provide important information for the adaption of IMCA for the field, which will be coupled to the Portable Ice Nucleation Chamber (PINC, [6]). Results from subsequent field measurements could give a better idea of the importance for processes in mixed-phase clouds initiated by biological aerosol particles.

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Is there a lower size limit for mineral dust ice nuclei in the immersion mode?

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There is observational evidence that atmospheric aerosol particles which are able to trigger ice nucleation are larger than approximately 100nm (e.g. Fletcher, 1959). On the other hand observations of IN active macromolecules which have been proposed to be responsible for the enhanced ice formation in the washing water of pollen indicate no such size limit (Augustin et al., 2012).

To investigate the size dependence of mineral dust species on their ability to serve as ice nuclei, the size dependent frozen fraction of droplets containing single immersed mineral dust is investigated with the IMCA/ZINC experimental setup (Lüönd et. al., 2010). Special care was taken to generate monodisperse particles in the lower size range, by using a two stage size selection setup including a differential mobility analyser and a centrifugal particle mass analyser.

Experimentally we did not find a discontinuous decrease in the ice nucleation ability of 100nm kaolinite particles. If the available surface area itself catalyses ice nucleation or rather specific active sites which are more likely to be present on a larger surface, is addressed by comparing the experimental findings to model predictions following the classical nucleation theory and the active site approach.

Measurements with different dust species corroborate the general findings from kaolinite. They are useful for modelling studies that investigate the influence of dust emissions from different sources, which are transported over varying distances and therefore exhibit different size distributions. Additionally, the experimental results support the applicability of the ice nucleation active surface site density (INAS) approach (e.g. Murray et al., 2012) to compare different measurements.

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Thermodynamics and Kinetics of the low-temperature HCl/H_2O Phase Diagram Revisited

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Thin ice films, representative of PSCs, are grown in a high vacuum multidiagnostic stirred flow reactor by deposition of pure thin ice film (1 to several μm) on a Si optical support and subsequently doped with known amounts of HCl. Steady state and pulsed valve experiments are performed to measure the rate of evaporation R_{ev} and the accommodation coefficient α of HCl and H_2O on selected ice substrates using residual gas MS. FTIR spectroscopy in transmission is used to monitor the condensed phase and led to the identification of three ice substrates of interest: amorphous HCl/H_2O (amHCl), crystalline HCl hexahydrate (HCl•6H_2O, HH) and pure H_2O ice.

The following results have been obtained (taking results on pure ice as a benchmark) that should help define the nucleation and growth conditions of solid HCl/H_2O ices: **(i)** the required H_2O supersaturation for pure ice growth (I_h) on the SiO_2 surface strongly increases with decreasing temperature with an apparent activation energy of 21 kJ/Mol. The conditions for nucleating the (metastable) crystalline HH phase ($T \leq 173$ K) probably rule out the occurrence of HH in the terrestrial atmosphere under UT/LS conditions: the stable atmospheric HCl/H_2O phase seems to be amorphous; **(ii)** α(HCl) on amHCl decreases with temperature and is lower by a factor of ~2 at T = 205 K with α(H_2O) on amHCl being lower by ~20% at T = 205 K. R_{ev}(HCl) is significantly lower by a factor of 100 compared to R_{ev} (H_2O) which is equal to that of pure ice; **(iii)** α(HCl) on HH decreases with temperature being lower by a factor ~5. α(H_2O) on HH is slightly lower by a factor of ~1.5 at $T \leq 193$ K. On HH R_{ev}(HCl) is lower by a factor of ~100 than R_{ev} (H_2O) and comes close to that for amHCl; **(iv)** the equilibrium vapour pressure P_{eq}(HCl) on both HH and amHCl are identical and lower by a factor of ~40 compared to P_{eq}(H_2O) on both HH and amHCl being equal to pure ice; **(v)** we propose a modification of the HCl/H_2O phase diagram by extending the existence area of HH to lower temperatures towards crystalline HCl•3H_2O based on IR spectroscopic evidence (see Figure 1). However, this does not change the conclusion that amorphous HCl/H_2O is the stable phase in the terrestrial atmosphere.

HCl and H_2O mass balances are excellent and within ± 10%, both accommodation and evaporation rates are thermochemically closed: the measured values of P_{eq} are equal to $R_{ev}/\alpha\omega$, where ω is the gas-surface collision frequency with the sample surface (Si window).

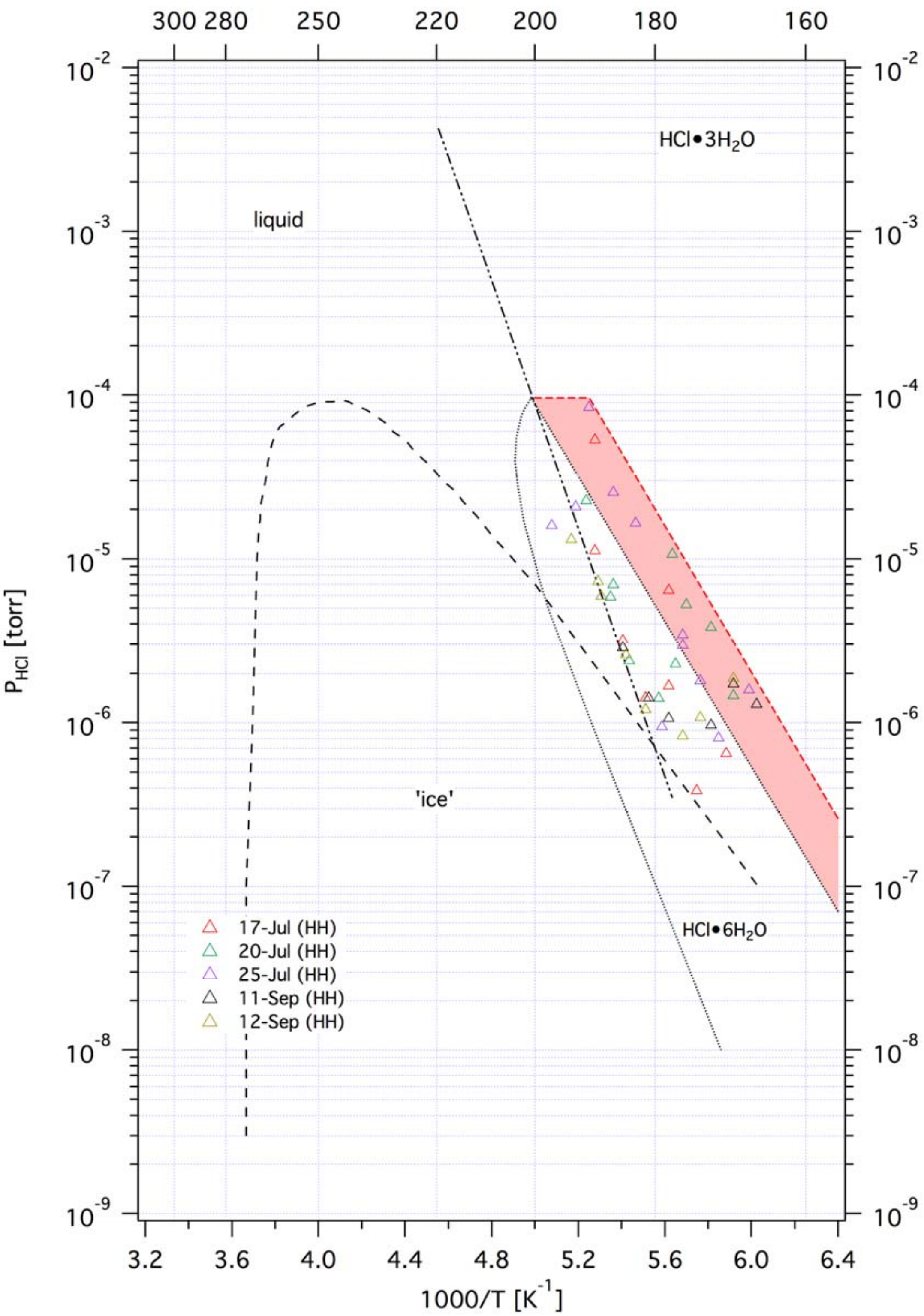

Figure 1: HCl/H_2O phase diagram for crystalline (metastable) HH of approximate thickness of 1 μm. The proposed extended area is coloured in red. The corresponding phase diagram for amHCl will be shown in the presentation.

The production and characteristics of ice nuclei from biomass burning in the US

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The production rates and chemical characteristics of heterogeneous ice nuclei (IN) from diverse sources remain largely unknown. Understanding these characteristics is necessary in determining the direct and indirect impacts of aerosols on clouds and the climate. IN emitted from biomass burning are of interest owing to their apparent potential contribution to the global IN reservoir and an anticipated increase in global wildfire frequency that may enhance the role of this source of IN relative to others. Here, we aim to gain insight concerning IN produced from biomass burning through laboratory studies and field measurements of two types of biomass burning: prescribed burning and wildfires.

IN number concentrations at various temperatures were measured with the CSU continuous flow diffusion chamber operated in the condensation/immersion freezing nucleation regime during four large prescribed burns in southwest Georgia and two large wildfires in northern Colorado, USA. Residual IN were captured as activated ice crystals for offline analysis and categorized via transmission electron microscopy based on elemental composition and morphology. Aerosol mass concentrations, total particle number concentrations, aerosol size distribution, and aerosol bulk composition were also measured, as well as carbon monoxide concentrations, used as an indicator of in-plume sampling. Fuel burned during the prescribed burns was a mixture of wiregrass and longleaf pine underbrush, while the wildfire fuels mostly consisted of ponderosa pine underbrush and timber. Specialized measurements were also made in the laboratory incorporating a single particle soot photometer to further investigate the contribution of refractory black carbon to IN produced from combusted wiregrass.

The temporal dependence of IN concentrations at various activation temperatures, relationships between IN and the number concentrations of larger-diameter particles, IN elemental categorizations, and the role of soot particles will be presented. These data provide supporting evidence that biomass burning is a source of IN, and also point to clear differences between the IN produced from prescribed burns and wildfires, likely attributed to differences in fuel type and/or combustion efficiency. The suitability of a previously-developed parameterization for prediction of IN concentrations in smoke aerosol will also be discussed.

Temperature dependence of the relation between ice nuclei (IN) and primary biological aerosol particles (PBAPs) at a forested site in Colorado

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It has been suggested that primary biological aerosol particles (PBAPs) such as bacteria and fungal spores can act as efficient ice nuclei (IN) even at temperatures warmer than -20°C. Using a continuous flow diffusion chamber (CFDC) combined with an aerosol concentrator, we investigated the number concentrations of IN active under mixed-phase could conditions at temperatures ranging from about -34°C to -9°C at a forested site during the BEACHON-RoMBAS (Bio-hydro-atmosphere interactions of Energy, Aerosols, Carbon, H_2O, Organics, and Nitrogen – Rocky Mountain Biogenic Aerosol Study) campaign in July-August 2011. Here we show that although the IN populations at the BEACHON site are related to the number concentrations of ambient fluorescent biological aerosol particles (FBAPs) measured using an ultraviolet aerodynamic particle sizer (UV-APS), their relationship clearly changes depending on temperature. Our results indicate strong increases in both IN and FBAP populations can be induced during rain events. However, the increases in the IN number concentrations in response to the FBAP number concentrations are stronger at warmer temperatures, suggesting that PBAPs make a more significant contribution to the IN populations at warmer temperatures. We specifically confirm the existence of fungal spore-like IN active at these warmer temperatures. Finally, we use these data to design a parameterization to quantify IN number concentrations, in dependence on temperature and FBAP number concentrations.

The relationship of cirrus ice water content, crystal number, and size to vertical velocity: First results from the MACPEX field campaign

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The role of ice clouds in the radiation budget of the atmosphere is difficult to determine owing to the number and variability of the microphysical, and thus radiative, properties involved, as well as the current lack of understanding of how and why some of those variations occur. The microphysical properties of an individual cirrus cloud, such as the ice crystal number, size, and water content are vital components for determining that role. However, without the proper understanding, these properties continue to be challenging to describe and parameterize.

In this study we will present observations obtained during the 2011 Midlatitude Airborne Cirrus Properties Experiment (MACPEX) that was based in Houston, Texas. Specifically we will compare the microphysical properties of the cirrus observed during MACPEX with the extensive cirrus in situ data sets of Schiller et al. (2008), Krämer et al. (2009), and Luebke et al. (2012). A first look into the data shows that MACPEX is different from the earlier cirrus data sets: higher IWCs were observed together with higher ice crystal numbers at temperatures around 215K, while at around 225K average IWCs were found, but were accompanied by low ice crystal numbers. Further investigation of the meteorological situation and of the role of vertical velocity, freezing mechanism (heterogeneous/homogeneous ice nucleation), and sedimentation of ice crystals will be included to explain the MACPEX observations.

A mid-latitude cirrus lidar climatology with thin cirrus clouds in the lowermost stratosphere

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Lidar observations (light detection and ranging) of cirrus clouds over Jülich, Western Germany, are performed and analyzed focusing on obtaining a representative cirrus climatology in terms of cirrus thickness, top and base heights, mid temperature, optical thickness and ice water content (IWC). The lidar instrument measures optical properties (i.e. backscatter signals, extinction coefficient and depolarization) of ice particles at a wavelength of 355 nm from the ground. The observed cirrus clouds are evaluated with a temporal averaged extinction profile and corrected for multiple scattering. The optical depth is used to classify radiative properties. Mean and median values of cirrus optical depth are found to be 0.28 and 0.12 (range: 0.002 – 3), respectively. 143 cirrus observations are analyzed together with additional meteorological data under macrophysical, radiative, and microphysical aspects. By comparing these cirrus properties to three other mid-latitude lidar climatologies (Sassen and Comstock, 2001; Immler and Schrems, 2002; Goldfarb et al., 2001), a rather good agreement is found and the Jülich lidar climatology is assessed to be representative.

Most observed cirrus base heights are around 6 to 10.5 km and top heights around 12 km, implying a frequent cirrus generation due to synoptic weather pattern (e.g. frontal systems). Thus, the cirrus clouds are relatively thick with a vertical extent of about 2.25 km and occur mostly directly around the thermal tropopause. However, around 4 % of the cirrus clouds in the lidar climatology occurred above the tropopause. This indicates cirrus cloud occurrence in the lowermost stratosphere (LMS). The gradient in water vapor at the transition between troposphere and the LMS is strongly negative. Even small contributions of moist tropospheric air masses from the tropics due to quasi-horizontal transport into the LMS can increase the water vapor concentration significantly. This enhanced water vapor values could initiate the formation of thin cirrus clouds in the LMS up to polar latitudes. Further investigations could improve the understanding of mixing processes in the UT/LS. ECMWF data will be used to show the origin of the air masses which contain cirrus clouds above the tropopause as implied by the lidar observations. This will be done by using the static stability as marker for stratospheric and tropospheric air masses.

Ice and liquid partitioning in mid-latitude and artic mixed-phase clouds: how common is the real mixed-phase state

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The influence of mixed-phase clouds on the radiation budget of the earth is largely unknown. One of the key parameters to determine mixed-phase cloud radiative properties however is the fraction of ice particles and liquid droplets in these clouds. The separate detection of liquid droplets and ice crystals especially in the small cloud particle size range below 50 µm remains challenging though.

Here, we present airborne NIXE-CAPS mixed-phase cloud particle measurements observed in mid-latitude and Arctic low-level mixed-phase clouds during the COALESC field campaign in 2011 and the Arctic field campaign VERDI in 2012. NIXE-CAPS (Novel Ice EXpEriment - Cloud and Aerosol Particle Spectrometer, manufactured by DMT) is a cloud particle spectrometer which measures the cloud particle number, size as well as their phase for each cloud particle in the diameter range 0.6 to 945 µm.

The common understanding in mixed-phase cloud research is that liquid droplets and ice crystals in the same cloud volume are rather sparse, but instead either liquid droplets or ice crystals are present. However, recently published model studies (e.g. Korolev, A. & Field, P., The effect of dynamics on mixed-phase clouds: Theoretical considerations. J. Atmos. Sci. 65, 66–86, 2008) indicate that a cloud state containing both liquid droplets and ice crystals can be kept up by turbulence. Indeed, our particle by particle analyses of the observed mixed-phase clouds during COALESC and VERDI indicate that the real mixed-phase state is rather common in the atmosphere. The spatial distribution of the mixed-phase ice fraction and the size of the droplets and ice crystals however vary substantially from case to case. The latter parameters seem to be influenced not only by concentration of ice nuclei but also - to a large degree - by cloud dynamics.

New species of ice nucleating fungi in soil and air

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Primary biological aerosol particles (PBAP) are ubiquitous in the atmosphere (*1*). Several types of PBAP have been identified as ice nuclei (IN) that can initiate the formation of ice at relatively high temperatures (*2, 3*). The best-known biological IN are common plant-associated bacteria. The IN activity of these bacteria is due to a surface protein on the outer cell membrane that catalyses ice formation, for which the corresponding gene has been identified and detected by DNA analysis (*2*). Fungal spores or hyphae can also act as IN, but the biological structures responsible for their IN activity have not yet been elucidated. Furthermore, the abundance, diversity, sources, seasonality, properties, and effects of fungal IN in the atmosphere have neither been characterized nor quantified. Recent studies have shown that airborne fungi are highly diverse (*1*), and that atmospheric transport leads to efficient exchange of species among different ecosystems (*4, 5*). The results presented in Fröhlich-Nowoisky *et al.* 2012 (*6*) clearly demonstrate the presence of geographic boundaries in the global distribution of microbial taxa in air, and indicate that regional differences may be important for the effects of microorganisms on climate and public health.
Thus, the objective of this study is the identification and quantification of ice nuclei-active fungi in and above ecosystems, and the unraveling of IN-active structures in fungi. Results obtained from the analysis of various soil and air samples and the presence of new fungal ice active species will be revealed.

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4. Burrows, S.M., *et al.* (2009a) *Atmos. Chem. Phys*., 9, (23), 9281-9297
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Microbial ice-nucleators in cloud water at the puy de Dôme (France)

Muriel Joly[1,2,3,4], Eléonore Attard[1,2], Martine Sancelme[1,2], Laurent Deguillaume[3,4], Cindy E. Morris[5], Anne-Marie Delort[1,2] and Pierre Amato[1,2]

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[5] INRA, UR 407 Plant Pathology Research Unit, 84140 Montfavet, France.

Ice nucleation active (INA) biological particles, in particular microorganisms, were studied in cloud water. Twelve cloud samples were collected over a period of 16 months from the puy de Dôme summit (1465 m, France) using sterile cloud droplet impactors. Samples were characterized through biological (cultures, cell counts) and physico-chemical measurements (pH, ion concentrations, carbon content…), and biological ice nuclei were investigated by droplet-freezing assays from -3°C to -13°C. The concentration of total INA particles within this temperature range typically varied from ~1 to ~100 per mL of cloud water; the concentrations of biological IN were several orders of magnitude higher than the values previously reported for precipitations: at -12°C, at least 76% of the IN were biological in origin, *i.e.* they were inactivated by heating at 95°C, and at temperatures above -8°C only biological material could induce ice. By culture, 44 *Pseudomonas*-like strains of bacteria were isolated from cloud water samples; 16% of them were found INA at the temperature of -8°C and they were identified as *Pseudomonas syringae*, *Xanthomonas* sp. and *Pseudoxanthomonas* sp.. Two strains induced freezing at as warm as -2°C, positioning them among the most active ice nucleators described so far. We estimated that, in average, 0.18% and more than 1% of the bacterial cells present in clouds (~10^4 mL^{-1}) are INA at the temperatures of -8°C and -12°C, respectively. These data will contribute to improve the parameterization of tmodels of atmospheric processes for evaluating the impact of biological ice nuclei on clouds.

TOWARDS THE IDENTIFICATION OF INDIVIDUAL BIOLOGICAL PARTICLES AS ICE NUCLEI USING SINGLE PARTICLE MASS SPECTROMETRY

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2Karlsruhe Institute of Technology, Institute for Meteorology and Climate, Germany
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4Vienna University of Technology, Institute of Materials Chemistry, Austria

Aerosols of biological origin such as bacteria, spores and pollen are gaining increasing attention in the research field of cloud microphysics. Their ability to act as heterogeneous ice nuclei has been shown in various laboratory studies; however, their atmospheric relevance in aerosol-cloud interactions is debatable. This is partly due to the lack of understanding of the actual processes driving water and ice nucleation on biological particles (e.g. Möhler et al., 2008), but also the challenging and unambiguous identification of these particles in the field.

This study focuses on mass spectrometric measurements of the chemical composition of laboratory generated bacterial cells and their residues, pollen, pollen fragments, and pollen washing water containing fragments of pollen and/or substances washed of from pollen that are expected to play a major role in the ice nucleating process involving pollen (Pummer et al., 2012; Augustin et al., 2012). For analysis, a TSI Aerosol Time-of-Flight Mass Spectrometer (ATOFMS) was used – a type of laser ablation instrument that is regularly applied for the in-situ detection and identification of ice crystal residuals (e.g. Pratt et al., 2009). The investigated biological particles are the bacterial species Pseudomonas syringae and Pseudomonas fluorescence, as well as birch and pine pollen and their washing waters, and Lycophyte spores. Except the Lycophyte spores, all species have been shown to be ice nucleation active.

“Fingerprints” usable for the identification of bacteria, spores and pollen by aerosol mass spectrometry are discussed and compared with mass spectra from real dust and soil samples that might have been exposed to or are internally or externally mixed with such biological material. Moreover, the findings are referenced to ambient spectra collected by identical instruments in the field and in the lab, and also to spectra referred to as “bioaerosols” in the literature (e.g. Fergenson et al., 2004). The results will be put into perspective for the identification of biological particles as IN.

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Introduction of a simplified version of the Soccer ball model

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Department of Physics, Leibniz Institute for Tropospheric Research, Leipzig, Germany

Heterogeneous ice nucleation is a primary pathway to form ice in the atmosphere and has a large influence on both, weather and climate. Since more than 60 years, many efforts have been made to describe this process theoretically. In this context, two contrary hypotheses have been developed: In the first case heterogeneous ice nucleation has been described as being stochastic (i.e. time dependent) in direct analogy with homogeneous nucleation, and in the second case as being singular, with ice nuclei (IN) initiating freezing at deterministic temperatures. Experimental results obtained in various research groups could be explained either with the stochastic or the singular description.

In a recent study, the Soccer ball model was introduced which bridges these two hypotheses providing a phenomenological explanation for the seemingly contradictory experimental results obtained. The model takes into account multiple nucleation sites on individual particles characterized by different contact angles θ.

In the original Soccer ball model, the contact angle distribution, which is a normal distribution, is discretized between 0 and π and through uniformly distributed random numbers each nucleation site is associated with a specific contact angle. The model is therefore fairly time consuming and hardly usable in atmospheric modelling applications.

Herewith, a new version of the Soccer ball model will be introduced. The model parameters are still the average number of surface sites n_{site} per particle, the mean contact angle μ_θ and the width of the contact angle distribution σ_θ. But in the new version the mean ice nucleation behaviour of the particle population is calculated without the use of random numbers. It will be shown that for equal parameter settings both methods lead to similar results whereas the new formulation is simpler and computationally more efficient without loss of accuracy.

Furthermore a sensitivity study concerning the dependence of the heterogeneous nucleation process on temperature and time will be performed for parameter settings which are based on experimental data (e.g., Arizona Test Dust and Birch pollen washing water) determined with the Leipzig Aerosol Cloud Interaction Simulator (LACIS, Hartmann et al., 2011).

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A new time-dependent singular model for ice nucleation

R. J. Herbert, B. J. Murray, and S. Dobbie

School of Earth and Environment, University of Leeds, United Kingdom

The presence of ice in the atmosphere has the potential to extensively alter the evolution of cloud hydrometeor species and have a significant radiative impact through the indirect effect. On the molecular scale a freezing event within a supercooled droplet is an inherently stochastic process but the complexity required to accurately model this process has led to the use of a simpler deterministic approximation that omits the time dependence, called the singular model. It is known that aerosol particles possess a wide range of ice nucleating abilities resulting from the presence of multiple components, each of which can be characterised through its activity and time-dependent nature. Studies on atmospherically relevant components show some are strongly time-dependent. This could affect the accuracy of experimental techniques that use residence times too short to capture this time-dependent nature, and lead to an under prediction when modelling ice formation following the deterministic approach. In order to improve this, the characterisation of atmospheric IN is required along with a framework that can facilitate the range of cooling rates and residence times used in freezing experiments.

In this study the Multiple Component Stochastic Model (MCSM) was used in an idealised model to examine how the distribution of nucleation sites and the temperature dependence of the nucleation rate ($^{d\ln J}/_{dT}$) affects the time dependent nature of IN. We show that, regardless of the site distribution and mean activity, it is solely the temperature dependence ($^{d\ln J}/_{dT}$) that controls the time dependent behaviour. This simple relationship can be used to determine the change in nucleation rate for a change in cooling rate, and has been applied to the deterministic approximation to provide a computationally efficient freezing model that correctly represents both the distribution of nucleating abilities and time-dependent nature of atmospheric IN. Finally, we show that this freezing model can be extended to describe ice nucleation in experiments where droplets are subjected to a constant temperature for a varying duration of time, such as isothermal and CFDC experiments. As a result this framework provides the ice nucleation community with a method for quantifying the time dependent behaviour of immersion mode ice nucleation, as well as a method for comparing datasets without bias from experimental residence times and cooling rates.

A new freezing parameterization for mixed-phase clouds in ECHAM GCM

Luisa Ickes(1), Ulrike Lohmann(1) and Corinna Hoose(2)
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(2) Institute for Meteorology and Climate Research, KIT, Karlsruhe, Germany

Mixed-phase clouds are the dominant cloud type in the Arctic and crucial for the Arctic climate and its seasonality by having a profound impact on the radiation balance and thus on the sea ice coverage [1, 2]. The formation and evolution of these clouds is highly dependent on their microphysical processes. A modest change in ice nuclei (IN) concentrations can influence the lifetime of mixed-phase clouds. However, the interaction of IN with Arctic clouds is not very well represented in many (global) models, which could be related to inadequate parameterizations of ice nuclei, heterogeneous freezing processes and the cloud processing of aerosols.

In this study the freezing processes in mixed-phase clouds and their role for Arctic climate are analyzed using the global climate model ECHAM with a two-moment cloud microphysics scheme [3] coupled to the aerosol module HAM [4]. Therefore a new freezing parameterization scheme based on Classical Nucleation Theory (CNT) [5] is introduced into ECHAM. It will be evaluated against two different Arctic case studies in comparison to an empirical freezing parameterization. For evaluation the data of the ARM Mixed-Phase Arctic Cloud Experiment and observations (MPACE) and the Indirect and Semi-Direct Aerosol Campaign (ISDAC) is used. In this study we will investigate if the choice of a parameterization scheme can influence the representation of Arctic mixed-phase clouds in ECHAM.

References

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Impact of different nucleation types on dynamically induced cirrus inhomogeneities

M. Voigt and P. Spichtinger

Spatial inhomogeneities in cirrus clouds have a great impact on their radiative properties and therefore on Earth's energy budget. The formation of inhomogeneities depends strongly on the environmental conditions and the dynamical situation.

In this study idealized numerical simulations with a 2D cloud resolving version of the EULAG model are carried out in order to identify the key processes which lead to inhomogeneities. The model includes a two moment bulk microphysical scheme for cold clouds. Homogeneous and heterogeneous nucleation, deposition and sedimentation are the processes represented by the scheme.

As environmental conditions we prescribed an ice supersaturated layer in a region of strong wind shear and stable stratification. The impact of the dynamical conditions is tested by varying the Richardson number. The relative importance of dynamic and thermodynamic processes is investigated by allowing nucleation. Simulations without nucleation are compared with simulations with only homogeneous and simulations with both homogeneous and heterogeneous nucleation. The different length scales involved in the formation of the inhomogeneities are identified by spectral analysis.

Buffet

Saturday 6th April 2013, 17:15 (Dinner)

Starter:

- Beef broth with vegetable chunks and pancake strips
- Carrot-Coco-Ginger soup with cream topping and parsnip

Main Dish:

- Pork roll with root vegetable and *Spätzle* (pasta)
- Turkey Saltimbocca with rosemary potatoes and baby tomatoes
- Spinach dumpling with goat cheese in saffron sauce with grated *Emmentaler* (cheese)
- Fresh salads from the Buffet

Sunday 7th April 2013, 12:40 (Lunch)

Starter:

- Beef broth with vegetable chunks and *Kaspressknödel* (dumpling)
- Tomato-Cream soup with cream topping, basil, and *Grana* (cheese)

Main Dish:

- Cooked special meat of beef with fried potatoes, chive sauce and apple horseradish
- Roasted turkey fillet on mushroom ragout with *Treccine* (pasta)
- Pumpkin-Brie-lasagna with spicy cranberry- and cream sauce
- Fresh salads from the Buffet

Accommodation

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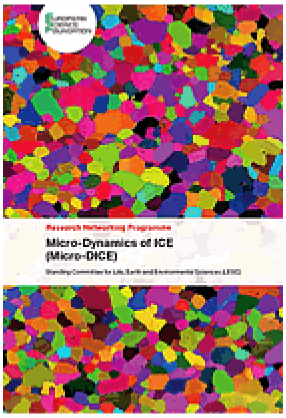

Micro-Dynamics of Ice (Micro-DICE)
microdice.eu

Pfeiffer-Vacuum
www.pfeiffer-vacuum.com

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Clouds

Fleeting Worlds
22 March 2013 - 01 July 2013

From 1800 landscape painting experienced an impressive heyday. Within this genre, artists paid increasing attention to the motif of clouds. These strange, elusive formations consisting of water, air and light appear as conveyors of different emotions and messages. Bushy clouds in a sunny sky contribute significantly to the positive atmosphere of a landscape and seem to be an almost indispensible feature in idyllic depictions of nature. A sky traversed by dark rain and thunder clouds, on the other hand, is perceived as threatening, while a band of clouds bathed in the glow of the red evening light sets a melancholy mood. Bizarre cloud formations, in turn, can be interpreted as enigmatic signs, as mysterious messages and warnings of imminent danger. A sense of foreboding is also conveyed by masses of clouds that appear out of control, occasioned either by natural disasters or by man as a result of technical intervention, such as exhaust fumes and atomic explosions.

The exhibition seeks to shed light on these different aspects of cloud depictions with a great variety of select examples of European and American painting and photography from 1800 to today. The presentation features works by Caspar David Friedrich, Carl Gustav Carus, William Turner, Claude Monet, Alfred Sisley, Paul Cézanne, Vincent van Gogh, John Constable, Ferdinand Hodler, Gustav Klimt, Egon Schiele, Edvard Munch, Emil Nolde, René Magritte, Ansel Adams, Alfred Stieglitz, Edward Steichen, Gerhard Richter, Anselm Kiefer, Paul Wolff, Olivier Masmonteil, Dietrich Wegner, Studio ++ to name but the most internationally famous representatives.

Leopold Museum at the MuseumsQuartier, U2 MuseumsQuartier, U3 Volkstheater
The Museum is open daily: 10am to 6pm, Thursdays: 10am to 9pm, Closed on Tuesdays.

Annex 2: Full list of speakers and participants

ESF - Participation List - Management

ESF ACTIVITY

Unit(s)	: LESC
Activity Title	: Micro-Dynamics of Ice

PROJECT

Science Meeting	: Workshop
Title of Science Meeting	: Atmospheric Ice Nucleation
Location	: Vienna, Austria, Europe
Date of Science Meeting	: 06/04/2013 - 07/04/2013

Add a Convenor

Convenor(s) 5

Name	City, Country	Type		
Professor Hinrich Dr. Grothe	Vienna, (AT)	Convenor	Edit	
Dr. Anne Kasper-Giebl	Vienna, (AT)	Convenor	Edit	DELETE
Dr. Martina Krämer	Jülich, (DE)	Convenor	Edit	DELETE
Mr. Bernhard Pummer	Vienna, (AT)	Convenor	Edit	DELETE
Professor Peter Spichtinger	Mainz, (DE)	Convenor	Edit	DELETE

Add a Speaker

Speakers 24

Name	City, Country	Type		
Mr. Piere Amato	Aubiere, (FR)	Speaker	Edit	DELETE
Ms. Stefanie Augustin	Leipzig, (DE)	Speaker	Edit	DELETE
Dr. Janine Fröhlich	Mainz, (DE)	Speaker	Edit	DELETE
Mr. Ross Herbert	Leeds, (UK)	Speaker	Edit	DELETE
Dr. Naruki Hiranuma	Eggenstein-Leopoldshafen, (DE)	Speaker	Edit	DELETE
Miss Nadine Hoffmann	Eggenstein-Leopoldshafen, (DE)	Speaker	Edit	DELETE
Dr. Kristina Höhler	Eggenstein-Leopoldshafen, (DE)	Speaker	Edit	DELETE
Mr. Riccardo Iannarelli	Ennetbaden, (CH)	Speaker	Edit	DELETE
Ms. Luisa Ickes	Zürich, (CH)	Speaker	Edit	DELETE
Mr. Lukas Kaufmann	Zürich, (CH)	Speaker	Edit	DELETE
Ms. Monika Kohn	Zürich, (CH)	Speaker	Edit	DELETE
Miss Anna Luebke	Boulder, (US)	Speaker	Edit	DELETE
Ms. Christina McCluskey	Fort Collins, (US)	Speaker	Edit	DELETE
Dr. Jessica Meyer	Jülich, (DE)	Speaker	Edit	DELETE
Dr. Dennis Niedermeier	Leipzig, (DE)	Speaker	Edit	DELETE
Mr. Bernhard Pummer	Vienna, (AT)	Speaker	Edit	DELETE
Dr. Christian Rolf	Jülich, (DE)	Speaker	Edit	DELETE
Dr. Berko Sierau	Zürich, (CH)	Speaker	Edit	DELETE
Mr. Emiliano Stopelli	Basel, (CH)	Speaker	Edit	DELETE
Dr. Daniel Sullivan	Leeds, (UK)	Speaker	Edit	DELETE
Dr. Erik Thomson	Gothenburg, (SE)	Speaker	Edit	DELETE
Dr. Yukata Tobo	Fort Collins, (US)	Speaker	Edit	DELETE
Mr. Matthias Voigt	Mainz, (DE)	Speaker	Edit	DELETE
Dr. André Welti	Zürich, (CH)	Speaker	Edit	DELETE

Add a Participant

Participants 14

Name	City, Country	Type		
Miss Yvonne Boose	Zürich, (CH)	Participant	Edit	DELETE
Dr. Julia Burkart	Vienna, (AT)	Participant	Edit	DELETE
Dr. Klaus Martin Gierens	Weißling, Oberpfaffenhofen, (DE)	Participant	Edit	DELETE

Mr. Florian Handle	Vienna, (AT)	Participant	Edit	DELETE
Mr. Piotr Kupiszewski	Villigen, (CH)	Participant	Edit	DELETE
Dr. Johannes Ofner	Vienna, (AT)	Participant	Edit	DELETE
Dr. Vaughan Phillips	Leeds, (UK)	Participant	Edit	DELETE
Dr. Philipp Reutter	Mainz, (DE)	Participant	Edit	DELETE
Dr. Michael Rossi	Villigen, (CH)	Participant	Edit	DELETE
Mr. Maher Sahyoun	Copenhagen N, (DK)	Participant	Edit	DELETE
Professor Otto Schrems	Bremerhaven, (DE)	Participant	Edit	DELETE
Mr. Fabian Weiss	Vienna, (AT)	Participant	Edit	DELETE
Dr. Heike Wex	Leipzig, (DE)	Participant	Edit	DELETE
Mr. Tobias Zolles	Vienna, (AT)	Participant	Edit	DELETE

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>> End >>

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