Research Networking Programmes

Science Meeting – Scientific Report

Proposal Title: *2nd Workshop on Atmospheric Ice Nucleation*

Application Reference N°: 5373

1) Summary

2) Description of the scientific content of and discussions at the event

3) Assessment of the results and impact of the event on the future directions of the field

4) Annexes 4a) and 4b): Programme of the meeting and full list of speakers and participants

1) 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 far from being understood. Ice Clouds form by different mechanisms from water vapor 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 catalyzed. 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 structure and dynamics are one of the least understood parameters in cloud microphysics, and therefore so is the impact of those aerosols on the climate. 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 modelers to gain a better understanding of the processes involved and find new ways of measurement. Only in the last step the global impact of ice microdynamics on e.g. the water cycle or the radiation budget can be determined accurately.

The ESF research networking program 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.

2) Scientific Content of the Discussions

General questions:

Some of the discussions were occupied by very general questions, which were considered from many different points of view.

Some of which were: How many species of IN can be found in nature, especially in the atmosphere? What concentrations of IN can be found in the atmosphere and more important in clouds? How important is primary ice nucleation due to IN and can it be that this effect is ousted by secondary ice nucleation? What freezing temperature is necessary to really show an effect on ice cloud formation and which IN are capable to trigger ice nucleation at such high temperatures?

There were no defined answers found concerning these questions, but it shows that atmospheric ice nucleation is still not entirely understood. Therefore, there was a consensus that it is inherently necessary to investigate every single sort of IN, known at present, and to find out about its chemical and structural needs. We all agreed that this knowledge is the key step to understand the process of heterogeneous ice nucleation in general and thus to understand the process of heterogeneous ice nucleation in the atmosphere in particular.

Surface interactions:

A part of the talks was about mineral dust species and modified mineral dust species (mostly with biomolecules on the surface that can be found in nature). This modified dust species can be used to model dusts which are present in the atmosphere and differ in their behavior compared to pure minerals. A big topic hereby was potassium feldspar, which is the most important mineral IN. While the impact of the crystal structure has already been covered during the talks, the discussions were mostly about surface properties. Those are not always specified and hardly included in the results (in case of potassium feldspar e.g. ordered hydroxyl surface groups).

Another important topic regarding mineral dusts was observations concerning milled samples that sometimes show a different behavior than coarse dust. Here, mostly surface properties were discussed, since freshly milled minerals should show different properties (concerning lattice and IN active surface sites), which might need some time to age. This could lead to a behavior which is closer related to the coarse and natural aged dust. However, it is interesting that this behavior can be observed and it needs to be analyzed in more detail.

Biological ice nucleation:

Biological ice nucleation was a topic of high relevance in the workshop since several suspendable or even soluble ice nuclei were shown. Though their chemical and structural properties are still mostly unknown, it is an important finding that heterogeneous ice nucleation activity does not in every case need a macroscopic surface.

A discussion was induced by the talk of Tina Santl Temkiv, who focused on vesicles and membrane chunks released from bacteria in the atmosphere that might be a source of IN. Since those are much lighter than the whole bacteria, they could have a much bigger importance in higher latitudes, and therefore, in cloud formation and especially the vesicles have often not been considered.

Another point of discussion was the IN mechanism of the commercial product snomax® and how the protein changes once it is not embedded in the membrane anymore, which led to the influence of secondary structures of proteins on their ice nucleation ability.

Theoretical part:

Many fundamental questions of ice nucleation theory were discussed including thermodynamic and kinetic approaches. There was intensive discussion about influences on the nucleation rate as well as singular and stochastic descriptions.

Theoretical calculations of proteins were presented and their possible influences on ice nucleation due to induced ordering of water molecules in their hydration shells were discussed. This discussion was mainly about the mechanism itself and the role of proteins in heterogeneous ice nucleation in general.

Methods:

Since a large part of the workshop was concerned with new methods and new equipment for measuring ice nucleation, a big part of discussions applied to this topic. At present there are no standard methods established in the fields of ice nucleation (which was a point of discussion as well) and small differences in the method can lead to a significant shift in freezing temperatures, making results of different working groups hard to compare. Therefore, it is important to review these details and find ways to standardize the results inside this scientific community.

Some of these discussions were very specific as for example when Emiliano Stopelli presented their new measurement setup, which allows easy evaluation and storage of the sample after measurement, without possible contaminations, there was a discussion about possible heterogeneous ice nucleation triggered by plastic tubes. This was mostly driven by Janine Fröhlich and Bernhard Pummer (they work with a well comparable setup), which saw this behavior for PCR plates from one supplier, but did not see the same for PCR plates from another supplier. It was also stated that the position of the blanks can be of great relevance in finding these specific behaviors. Through discussions like these, experiences can be shared, helping in optimizing methods and minimizing failures. Storing was also discussed, concerning Emiliano Stopelli's method, how it could change the IN activity in regard of biological IN (e.g. bacterial growth).

In general, it was pointed out that there are no analytical methods at present to discriminate specific IN in natural a sample, which is a problem, but there are groups working on it trying to develop a method through the detection of nitrogen-rich fragments. Concerning the surface properties of potassium feldspar it was stated that XPS might be a suitable method for examining the specific surface properties. Also storage of especially the freshly milled dusts was discussed in regards of mineral dusts in general. Another point concerning methodology was the comparability of laboratory and field measurements, since laboratory measurements, if carried out in a flow tube, are laminar and do not take turbulences in accounts. Also environmental conditions like e.g. water vapor saturation are not always well comparable. Here, new outdoor equipment was presented (e.g. PINC and SPIN) as well as their results. The discussion was mostly about future features of these systems and the possibility of miniaturization. Also that these instruments produce quantitative and do not give qualitative information leaving a future option of new features for these systems once defined methods are developed.

3) 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 modelers 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.

Annex 4a: Programme of the meeting

EUROPEAN SCIENCE FOUNDATION

2nd Workshop - Atmospheric Ice Nucleation

Vienna; Austria

26th and 27th of April 2014

Book of Abstracts

TU WIEN 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 2nd workshop. The aim 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.

This workshop will be at the weekend before the start of the EGU General Assembly 2014 in the Vienna Austria Centre. Therefore, many workshop participants will come to Vienna and will not need extra travel support. Additionally, the organizers of this workshop also offer a session at the EGU conference, which is called “Atmospheric Ice Particles” AS 3.6 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 26th April 2014

Hinrich Grothe

Organizers

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Location

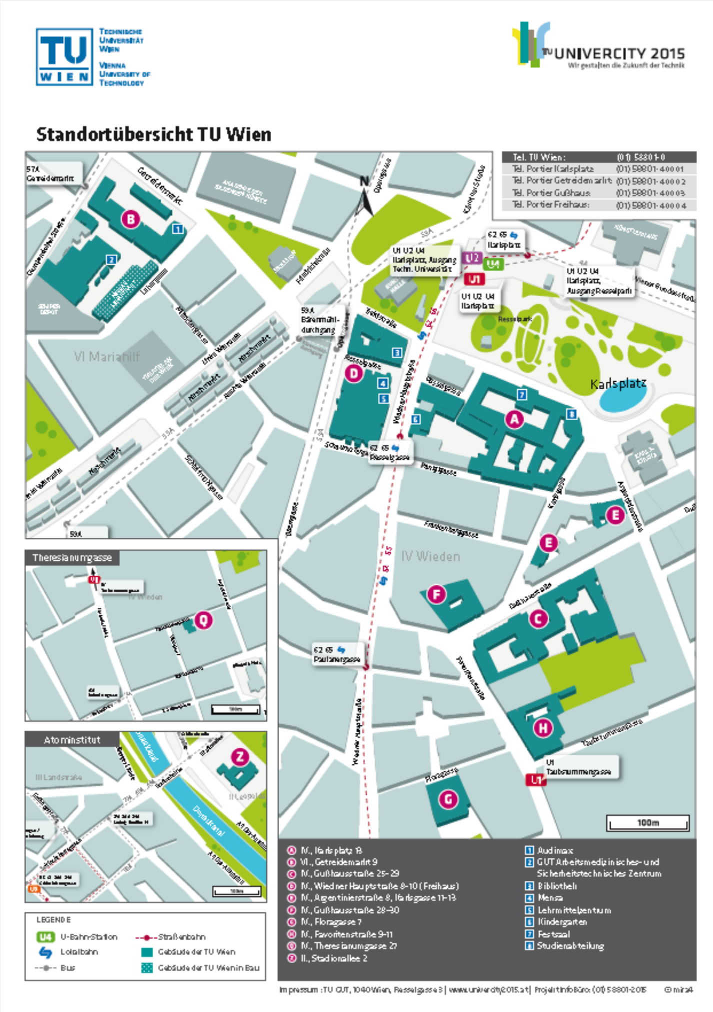
Vienna University of Technology

The workshop is held in the "Festsaal" (Ceremonial Hall) at the Vienna University of Technology, Karlsplatz 13, 1040 Vienna. This lecture hall is in the 1st floor of the main building of our university, which is situated at the Karlsplatz near Karlskirche.

Festsaal (Ceremonial Hall)

Ceremonial Hall

Maps



Transportation

Sessions

	Saturday 26th April		
12:00 - 13:00	Registration and Buffet		
13:00 - 13:10	Opening of the workshop	Hinrich Grothe	

	Laboratory Measurements I		
13:10 - 13:20	1st Talk	Bernhard Pummer	Characterization of biological ice nuclei from *Acremonium implicatum* and *Isaria farinosa*
13:20 - 13:25	Discussion		
13:25 - 13:35	2nd Talk	Janine Fröhlich	Ice nucleation activity in the widespread soil fungus *Mortierella alpina*
13:35 - 13:40	Discussion		
13:40 - 13:50	3rd Talk	Tina Santl Temkiv	Airborne *Pseudomonas sp.* and the ice nucleation activity of their cell fragments
13:50 - 13:55	Discussion		
13:55 - 14:05	4th Talk	Emiliano Stopelli	A new immersion freezing apparatus for biological ice nucleation research
14:05 - 14:10	Discussion		
14:10 - 14:20	General Discussion		
14:20 - 14:35	Coffee Break		

	Laboratory Measurements II		
14:35 - 14:45	1st Talk	Stefanie Augustin	Ice nucleation ability of mineral dust particles mixed with biological substances
14:45 - 14:50	Discussion		

14:50 - 15:00	2nd Talk	Joseph Niehaus	Laboratory measurements of contact freezing by dust and bacteria at temperatures of mixed phase clouds
15:00 - 15:05	Discussion		
15:05 - 15:15	3rd Talk	Tobias Zolles	Heterogeneous ice nucleation on mineral dust particles
15:15 - 15:20	Discussion		
15:20 - 15:30	4th Talk	André Welti	Ice nucleation properties of K-Feldspar polymorphs: How important is the crystal lattice?
15:30 - 15:35	Discussion		
15:35 - 15:45	5th Talk	Andreas Peckhaus	Immersion freezing on potassium-rich feldspar particles
15:45 - 15:50	Discussion		
15:50 - 16:00	General Discussion		

16:00 - 16:15	Coffee Break		

	Laboratory Measurements III		
16:15 - 16:25	1nd Talk	Sebastien Facq	Structures of ice particles formed during homogeneous freezing of micro-droplets of EtOH aqueous solutions
16:25 - 16:30	Discussion		
16:30 - 16:40	2nd Talk	Sandy James	Investigating the Phase Behaviour of Ternary Nitric / Sulfuric Acid Solution Droplets
16:40 - 16:45	Discussion		
16:45 - 16:55	3rd Talk	Michel Rossi	The Nucleation of crystalline Hydrate Films of HCl and HNO_3 under Laboratory Conditions
16:55 - 17:00	Discussion		

17:00 - 17:10	4th Talk	Carsten Budke	A novel optical freezing array for the examination of cooling rate dependence in heterogeneous ice nucleation
17:10 - 17:15	Discussion		
17:15 - 17:30	General Discussion		

17:30 - 17:40	Break		

	Laboratory Measurements IV		
17:40 - 17:50	1st Talk	Anatoli Bogdan	Visualization of the freezing process of aqueous organic solutions
17:50 - 17:55	Discussion		
17:55 - 18:05	2nd Talk	Thea Schmitt	Homogeneous Freezing of Water Droplets and its Dependence on Droplet Size
18:05 - 18:10	Discussion		
18:10 - 18:20	3rd Talk	Baban Nagare	Determination of Collision Efficiency from Contact freezing of Silver Iodide
18:20 - 18:25	Discussion		
18:25 - 18:35	4th Talk	Ottmar Möhler	A summary of heterogeneous ice nucleation results from the AIDA cloud simulation chamber
18:35 - 18:40	Discussion		
18:40 - 18:55	General Discussion		

18:55	Opening of the Buffet and Wine Reception

Sunday 27th April

	Atmospheric IN Measurements		
08:15 - 08:25	1st Talk	Karoliina Ignatius	First results obtained with the new portable ice nuclei counter SPIN
08:25 - 08:30	Discussion		
08:30 - 08:40	2nd Talk	Piotr Kupiszewski	Investigating the atmospheric ice phase at the high altitude Jungfraujoch site
08:40 - 08:45	Discussion		
08:45 - 08:55	3rd Talk	Yvonne Boose	In-situ detection of ice nuclei concentrations in the free troposphere in the deposition and condensation regi
08:55 - 09:00	Discussion		
09:00 - 09:10	4th Talk	Ryan Mason	Characterization of the Micro-Orifice Uniform Deposit Impactor-droplet freezing technique (MOUDI-DFT) for size-resolved quantitative measurements of ice nuclei
09:10 - 09:15	Discussion		
09:15 - 09:30	General Discussion		

09:30 - 09:45	Coffee Break		

	IN Characterization		
09:45 - 09:55	1st Talk	David G. Schmale III	Research on Airborne Ice-Nucleating Species (RAINS): A Collaborative Research Project Funded by the National Science Foundation
09:55 - 10:00	Discussion		
10:00 - 10:10	2nd Talk	David G. Schmale III	Expanding our Knowledge of Ice Nucleation Activity in the Fungal Genus Fusarium: From Biological Research Collections to Atmospheric Sampling Missions with Drones

10:10 - 10:15	Discussion		
10:15 - 10:25	3rd Talk	Andrew Heymsfield	Cloud Conditions Favoring Secondary Ice Particles in Tropical Maritime Convention
10:25 - 10:30	Discussion		
10:30 - 10:40	4th Talk	Denis Duft	Nucleation and ice growth on charged meteor smoke particles
10:40 - 10:45	Discussion		
10:45 - 10:55	General Discussion		

10:55 - 11:10	Coffee Break		

	IN Modelling		
11:10 - 11:20	1st Talk	Christoph Dellago	Studying nucleation processes with computer simulations: from freezing to cavitation
11:20 - 11:25	Discussion		
11:25 - 11:35	2nd Talk	Ivan Coluzza	Transferable coarse-grained potential for de novo protein folding and design
11:35 - 11:40	Discussion		
11:40 - 11:50	3rd Talk	Michael Schauperl	Polarizable Force Fields Improve Models of Ice Nucleation
11:50 - 11:55	Discussion		
11:55 - 12:05	4th Talk	Roland Huber	Molecular Dynamics of Ice Nucleation
12:05 - 12:10	Discussion		
12:10 - 12:20	5th Talk	Maher Sahyoun	On the usage of classical nucleation theory (CNT) in quantification of the impact of bio-aerosols on weather and climate
12:20 -	Discussion		

12:25			
12:25 - 12:35	6th Talk	Luisa Ickes	Sensitivity of the classical nucleation theory on thermodynamic and kinetic parameters
12:35 - 12:40	Discussion		
12:40 - 12:55	General Discussion		
12:55 – 13:00	Closing Remarks	Hinrich Grothe	
13:00 - 14:00	Lunch Buffet		
14:00	End of the Workshop		

Characterization of biological ice nuclei from *Acremonium implicatum* and *Isaria farinosa*

B. G. Pummer, U. Pöschl, and J. Fröhlich-Nowoisky

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Laboratory studies on fungal ice nucleation (IN) showed that only a handful of fungal strains showed activity, while most others were IN-negative [Kieft and Ruscetti 1990, Pouleur et al. 1992, Hasegawa et al. 1994, Tsumuki et al. 1995, Iannone et al. 2011, Morris et al. 2013, Haga et al. 2013, Pummer et al. 2013]. Recently, IN activity was also discovered in *Acremonium implicatum*, *Isaria farinosa* [Huffman et al. 2013] and some other soil and air fungi.

In laboratory experiments, we found out more about the active sites of these species: Like many biological IN, they can be easily washed off from the cells with water. They were further characterized by determining the impact of filtration (from 5 µm to 100 kDa), heating to 60°C, guanidinium chloride, lipase and papain on the IN activity. Therefore we applied a freezing assay of droplets [Pummer et al. 2013]. Via the Vali formula, we calculated the amount of IN per gram of mycelium, which is more than 10^5 g^{-1}.

The initial freezing temperature of *I. farinosa* (-4°C) was higher than for *A. implicatum* (-8°C). Apart from that, they have a mass of 100s of kDa and are sensitive towards heat, guanidinium chloride and papain. The results indicate that the IN are proteinaceous compounds like those of *Fusarium* spp. and lichen mycobionts, which belong to the same phylum, the *Ascomycota*.

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Ice nucleation activity in the widespread soil fungus *Mortierella alpina*

J. Fröhlich[1], Thomas C. J. Hill[2], Bernhard G. Pummer[1], Gary D. Franc[3], and Ulrich Pöschl[1]

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Soil organic matter has long been proposed as a source of atmospheric ice nuclei (IN), and biological IN can dominate the fraction active at warmer temperatures [1–3]. However, the sources of biological IN released from soils remain unclear, even though their role in triggering glaciation and precipitation has recently been supported [4,5]. Analysis of the cultivable fungi in topsoils from a range of land uses and ecotypes in south-east Wyoming found ice nucleation active (INA) fungi to be both widespread and often abundant, particularly in soils with recent inputs of readily decomposable organic matter. For example, in harvested and ploughed sugar beet and potato fields, and in the organic horizon beneath Lodgepole pine forest, their relative abundances among the cultivable fungi were 25%, 17% and 17%, respectively. Across all soils, 8% of fungal isolates were INA. All isolates initiated freezing at -5°C to -6°C and all belonged to a single species, *Mortierella alpina*, within the phylum *Fungi incertae sedis* (previously *Zygomycota*). By contrast, the handful of fungal species so far reported as INA all belong within the *Ascomycota* or *Basidiomycota* phyla[6,7]. *Mortierella alpina* is known to be saprobic (utilizing non-living organic matter), widespread in soil and has been previously found in air and rain samples. Sequencing of the ITS region and the gene for γ-linolenic elongase (apparently unique to *M. alpina*) revealed four distinct clades, each affiliated with a different soil ecological niche: cultivated with abundant organic residues, general cultivated, grassland and forest. The IN produced by *M. alpina* seem to be extracellular and easily suspendable macromolecules <300 kDa. Ice nucleating fungal mycelium will ramify topsoils and may release cell-free IN into it. If these IN survive decomposition or are adsorbed onto mineral surfaces this contribution will accumulate over time, perhaps to be transported with soil dust and influencing its ice nucleating properties.

References

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Airborne *Pseudomonas* sp. and the ice nucleation activity of their cell fragments

Santl-Temkiv T.[1,2,3], Sahyoun M.[4], Ling M.[2,4], Boesen T.[4], Hartmann S.[6], Augustin S.[6], Stratmann F.[6], Wex H.[6], Korsholm U.S.[5], Nielsen N.W.[5], Sørensen J.H.[5], Karlson U.G.[1], Finster K.[2,3]

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Biogenic IN are the only reported particles that promote ice formation at temperatures close to 0°C. Some common atmospheric bacteria, e.g. *Pseudomonas syringae*, have the unique capacity of forming ice nucleation active (INA) proteins and exposing them at their outer membrane surface. Besides intact INA cells, different bacterial residues, which carry INA proteins, can enter the atmosphere independently and may increase the atmospheric densities of bacterial INA proteins.

We analyzed cultivable bacterial communities during 14 precipitation events and we observed prevalence of strains affiliated with the genus *Pseudomonas*. Screening of isolated bacteria for their ice nucleation activity showed that ~12% carried INA genes. The analysis of backward trajectories and of radar reflectivity images indicated that airborne INA bacteria either came into the cloud through local convective transfer or were transported longer distances by air masses of continental origin. We investigated the presence of small INA bacterial fragments (<220 nm) in two precipitation events and found that they were present at densities of 482 IN per L of rain and 199 IN per L of melted snow. Bacterial fragments that are more abundant than intact cells could thus have additional impact on atmospheric processes. Two INA strains were able to produce outer membrane vesicles (with diameters of ~75nm and ~85 nm), which could be a source of INA bacterial fragments that we observed in the atmosphere. However, the immersion freezing experiments, performed in the laminar flow diffusion chamber LACIS, showed that only a negligible number (< 1.5%) of vesicles were INA. It thus remains unclear what kind of INA bacterial fragments we detected in precipitation samples. Our future plan is to prepare membrane discs carrying single INA proteins and use them as a model for the ice nucleation activity of bacterial fragments, which will help us understand the importance of these fragments for ice nucleation in the atmosphere.

A new immersion freezing apparatus for biological ice nucleation research

Emiliano Stopelli[1], Franz Conen[1], Christine Alewell[1], Cindy Morris[2]

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[2]: INRA, Plant Pathology Research, Avignon, FR

Drop-freezing is a useful technique for the analysis of biological ice nucleators, but current instruments have limitations like evaporation of the sample or its potential contamination.

LINDA (LED-based Ice Nucleation Detection Apparatus) is a new immersion freezing apparatus, where ice nucleation events are automatically recorded, thus improving the precision of the analysis. LINDA is based on the principle of decrease of red light transmission upon formation of ice into a solution. The liquid sample to be analysed (viz snow, rain or air collected with liquid impingers) is divided into small tubes containing aliquots of it and put into a cooling bath, where tubes are placed over a grid of LED lamps. Once a nucleator is activated, the whole aliquot of water in the tube freezes, provoking a decrease in light transmitted through it, which is automatically recorded by a camera placed above the grid.

This apparatus is specifically conceived for the detection of ice nucleators active at warm temperatures (≥-12°C), usually characterised by a vast majority of biological particles, such as bacteria, fungal spores, organic matter on soil.

The improvement of automatic detection in closed tubes ensures the sample to be safely maintained during and after the measurements without risks of contaminations, then leading to its conservation for further analyses. This aspect is of crucial importance to develop new approaches in biological ice nucleation research. For instance, a sample could be stored at low temperatures and its nucleating behaviour studied over time, allowing the potential enhancement of bacterial expression of ice nucleating proteins. In parallel, ice nuclei may be progressively isolated from the whole sample through successive dilution of the aliquots freezing at warm temperatures, thus reducing the interference of other background particles, minimising the loss of sample or cross contaminations during subsequent dilutions.

A brief reference to the results coming from a monitoring field campaign developed at the High Altitude Research Station of Jungfraujoch in the Swiss Alps (3580 m a.s.l.) will be also provided, as an example of field application of this apparatus.

Ice nucleation ability of mineral dust particles mixed with biological substances

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Ice nucleation in the atmosphere has been observed at temperatures higher than -20°C (e.g. Kanitz et al., 2011). In contrast, laboratory studies showed that the most abundant atmospheric ice nuclei (IN), i.e., mineral dust particles, are ice active at much lower temperatures (e.g. Murray et al., 2012). Biological particles such as bacteria or pollen nucleate ice at temperatures similar to those observed in the atmosphere. However, their numbers seem to be too small to explain atmospheric ice nucleation (Hoose et al., 2010). Recent studies suggest, that ice nucleation active (INA) macromolecules, i.e. protein complexes in the case of bacteria (e.g. Wolber et al., 1986), and most likely polysaccharides in the case of pollen (Pummer et al., 2012), which are responsible for the freezing, maintain their nucleating ability even when they are separated from their original carriers (bacterial cell or pollen grain, Hartmann et al., 2013; Augustin et al., 2012). This opens the possibility of accumulation of such INA macromolecules in e.g. soils and the resulting particles could be an internal mixture of mineral dust and INA macromolecules. If such biological IN containing soil particles are then dispersed into the atmosphere due to e.g. wind erosion or agricultural processes they could induce ice nucleation at temperatures higher then -20°C.

To explore this hypothesis, we investigated the ice nucleation behavior of mineral dust particles internally mixed with INA macromolecules. Specifically, we mixed pure mineral dust (Illit) with INA biological material (SNOMAX and birch pollen washing water) and quantified the immersion freezing behavior of the resulting particles utilizing the Leipzig Aerosol Cloud Interaction Simulator (LACIS). To characterize the mixing state of the produced aerosol we used single mass spectrometry as well as electron microscopy. We found that internally mixed particles which containing ice active biological material show the same ice nucleation behavior as the purely biological particles. That shows that INA macromolecules which are located on a mineral dust particle dominate the freezing process.

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Laboratory measurements of contact freezing by dust and bacteria at temperatures of mixed phase clouds

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Contact nucleation of ice is thought to play a significant role in the atmosphere where the freezing of water droplets remains one of the biggest uncertainties in current models of the atmosphere. Contact freezing efficiencies for various atmospherically relevant aerosols are reported for the temperature range 0 to -20 °C. The results are discussed in the context of mixed phase clouds, and we find that dry, micron sized dust aerosols can have substantive impact on warm temperature nucleation. Bacteria has the potential to be even more effective. Samples of Pseudomonas syringae and Pseudomonas fluorescens had widely varying freezing behavior. Nucleation threshholds cannot be easily predicted by the gene markers ice-positive or ice-negative as was done in past years for immersion freezing. In all cases the contact mode dominates the immersion mode freezing.

For Arizona Test Dust, feldspar, or rhyolitic ash, more than 1e3 (1e5) particles sized between 0.3um - 10.0um are required to initiate a freezing event at -20 °C (-15 °C) in the contact mode. An ice negative strain of Pseudomonas fluorescens is an order of magnitude more effective than the mineral dusts at every temperature tested. We find that an ice positive strain of Pseudomonas syringae reaches its maximum nucleating efficiency of 0.1 twelve degrees earlier than does the Pseudomonas fluorescens, similar to the behavior of ice negative and positive bacteria in the immersion mode, as discovered 40 years ago [Maki et al., 1974; Vali et al., 1976]. Surprisingly, cells of the ice positive strain (CC94) Pseudomonas syringae which did not express the ice+ gene, showed no contact freezing activity, whereas the ice- strain of Pseudomonas fluorescens did.

Heterogeneous ice nucleation on mineral dust particles

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Ice nucleation activities of mineral dust particles were investigated. The experiments were carried out using cryo-microscopy which is an oil-emulsion based method. The immersion freezing mode was addressed with this experimental setup. The studied samples were common inorganic atmospheric aerosols. Single minerals and natural samples were tested. Mineral dust particles are active ice nuclei in the immersion freezing mode up to 256K/ −17°C. The nucleation temperatures of mineral dusts are much lower than for biological ice nuclei. K-feldspar is the by far most active ice nuclei followed by other silicates. Natural samples which contain more than 5% K-feldspar were also active. The activity of K-feldspar can be attributed to its surface structure and the presence of potassium ions in the surface.

Ice nucleation on mineral dust particles takes place at certain nucleation sites. These sites are domains of molecular sites where water is stabilized in an ice-like structure. To form a good ice nucleation site, the site density of molecular sites needs to be high. More molecular sites are able to form larger domains on the surface, leading to better nucleation sites. This suggests further that the nucleation temperature of mineral dust particles scales with the surface area. The exact configuration of a molecular site is material specific and influenced by the local chemistry and structure of the dust particle surface. A favorable arrangement of the functional groups like surface hydroxyl and oxygen is proposed for the K-feldspar. Potassium ions seem to have a positive or neutral effect on the ice nucleation property of a silicate surface while cations with a higher charge density like calcium and sodium have a negative influence. K-feldspar is abundant in the environment and actually is the most important dust ice nucleus in the atmosphere. The nucleation temperatures of the K-feldspar particles are sufficient to enable further meteorological glaciation processes in high altitude clouds..

Ice nucleation properties of K-Feldspar polymorphs: How important is the crystal lattice?

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Due to the effect of pressure and temperature on the spatial disposition of atoms in the crystalline lattice during rock formation, chemically identical feldspars exist that have different lattice configurations. The feldspar polymorphs also have different physical properties such as cleavage, hardness, specific weight, melting point. In the current study we test the effect of the difference in crystal lattice on the ice nucleation efficiency of these particles.

In the past some authors suggested that it is mainly the chemical factors which are of importance for the ice nucleation efficiency of a substance (e.g. Pruppacher and Sänger, 1955). The identical composition of the feldspar polymorphs offer a natural system where the importance of the crystal structure can be investigated on chemically equivalent particles.

In a set of laboratory experiments using different feldspar polymorphs (Adular, Microcline, Orthoclase) the IMCA/ZINC experimental setup was used to measure the frozen fraction of single immersed, size selected feldspar particles. For each polymorph 200, 400 and 800nm particles were tested to gain information on the size dependency of the nucleation efficiency of the individual species.

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Immersion freezing on potassium-rich feldspar particles

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Recent laboratory measurements showed an increased ice nucleation ability of potassium-rich feldspar particles in the immersion freezing mode [1]. It was suggested that the proportion of K-feldspar in atmospherically relevant ice nuclei is related to their ice nucleation ability. The importance of K-feldspar is further supported by the field measurements, indicating that it can make a mass fraction of up to 24% in Asian and African mineral dusts [2]. In this contribution we present results of immersion freezing experiments with monodisperse droplets of aqueous suspensions of K-feldspar on a cold stage. We show that the ice nucleation activity strongly depends on i) the particle size distribution (in particular the ice nucleation properties of submicron feldspar particles) ii) the weight concentration of the particles in the aqueous suspension and thus on the total particle surface immersed into the droplets and iii) the age of the particles in an aqueous environment. Further a comparison of different K-feldspars is presented indicating that the origin and the processing methods have a significant impact on the IN activity. The mineralogical composition of feldspar samples is analyzed by means of Raman spectroscopy and a quantification of the particle surface is carried out with environmental scanning electron microscopy (ESEM). The results of freezing experiments are interpreted within the concept of ice nucleation active surface site (INAS) density, which allows a comparison with data obtained with different experimental methods (IN counters, expansion chambers, etc.).

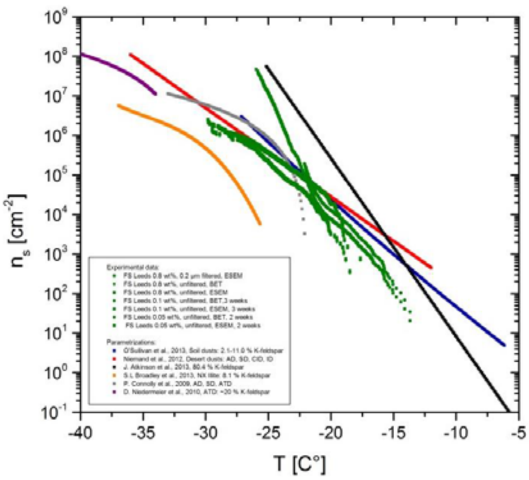

Fig. 1: INAS density for K-feldspar and mineral dusts.

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Investigating the Phase Structure of Ethanol Aqueous Solutions Micro-Droplets During Homogeneous Freezing

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In the upper troposphere, ice particles can form via homogeneous ice nucleation from submicrometric liquid particles or heterogeneous ice nucleation on solid ice nuclei. These atmospheric ice particles form cirrus clouds which play a regulation role in radiation fluxes at the top of the troposphere. Much research has been done in this regard focused on the homogeneous ice nucleation process of pure water particles. However, homogeneous ice nucleation can also occur from aqueous solutions [1].

If the influence of organics of high molecular weight on homogeneous ice nucleation from liquid particles is well-characterized [2], little is known about the effect of oxygenated volatile organic compounds. However, many light organics such as alcohols, aldehydes, ketones are ubiquitous in the upper troposphere [3, 4]. Among them, alcohols and especially ethanol, exhibits very glass transition at low temperature (T_g=97K) in such a way that the formation of amorphous particles is very unlikely [5]. It is therefore relevant to decipher what crystalline structures are formed during the homogeneous ice nucleation of micro-droplets containing ethanol and compared them with structures formed during homogeneous ice nucleation of pure water and aqueous solutions containing organics of high molecular weight.

In this study, we present in situ Raman spectroscopy and X-ray diffraction evidences of the formation of hexagonal ice or ethanol hydrate (depending on the ethanol content) during the homogeneous ice nucleation of ethanol aqueous solutions micro-droplets.

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Investigating the Phase Behaviour of Ternary Nitric / Sulfuric Acid Solution Droplets

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The literature regarding the phase behaviour of Polar Stratospheric Clouds (PSCs) shows a distinct disagreement between laboratory and field studies. Some PSCs have been shown to be composed of Nitric Acid Trihydrate (NAT) which nucleates on water ice crystals formed during cooling induced by gravity waves. However, other solid PSCs of synoptic scale have been observed in air masses unlikely to contain water ice crystals.

Laboratory studies of nucleation kinetics, which have most commonly investigated binary solutions of nitric acid and water, have tended to find that Nitric Acid Dihydrate (NAD, either the α- or β- polymorph) is at least the primary nucleating phase.

To address this dichotomy, the crystallisation behaviour of solutions of Nitric and Sulfuric acid of concentrations relevant to the stratosphere is under examination using 2 techniques;

- Raman microscopy studies of µl solution droplets suggest that, whilst binary solutions tend to form NAD, inclusion of sulfuric acid leads to rapid formation of NAT.
- To confirm this result, and to examine a more atmospherically relevant regime with greater statistical significance, XRD studies of nl droplets are currently underway.

The Nucleation of crystalline Hydrate Films of HCl and HNO_3 under Laboratory Conditions

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Thin ice films, representative of PSC's, are grown in a high vacuum multidiagnostic stirred flow reactor by deposition of a pure thin ice film (1 to several µm) on a Si optical support and subsequently doped with known amounts of HCl and HNO_3. Multidiagnostic experiments involving residual gas mass spectrometry (MS) used to monitor both the partial pressures of HCl, HNO_3 and H_2O as a function of time, thus dosing, and FTIR spectroscopy in transmission for the investigation of the condensed phase during growth have been applied. This led to the identification of three ice/HNO_3 substrates of interest: crystalline nitric acid trihydrate (NAT, HNO_3•$3H_2O$), both in their α- and β-forms, as well as metastable nitric acid dihydrate (NAD, HNO_3•$2H_2O$). Together with the amorphous HCl/H_2O (amHCl) phase, crystalline HCl hexahydrate (HCl•$6H_2O$, HH) and pure crystalline H_2O ice presented last year the nucleation data base so far obtained will be discussed regarding common features and differences of the solid phases regarding thermodynamic stability of the corresponding ice phases.

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, HNO_3/H_2O and pure H_2O ices:

(i) Pure ice and HNO_3 under NAT growing conditions are characterized by a singularity (peak) in partial pressure of H_2O and HNO_3, respectively, which may be interpreted as the required supersaturation pressure (P_{SS}) of approximately a factor of 2 to 5 over the steady-state or equilibrium vapour pressure of HNO_3. At the nucleation threshhold the condensed phase appears as "pure" ice in the case of NAT growth owing to the limited sensitivity of FTIR spectroscopy in the mid-IR. No temperature threshhold seems to operate as long as P_{SS} is attained or exceeded. Only α-NAT could be investigated because β-NAT was exclusively generated through spontaneous decay of α-NAT. NAD behaves in a similar manner as NAT regarding PSS despite the fact that it is metastable.

(ii) The nucleation conditions for HH are different in that no partial pressure singularity or supersaturation pressure requirement was ever observed. Instead, the temperature threshhold for nucleation of HH of $T \leq 173$ K is prohibitively low for terrestrial atmospheric applications. Beyond threshhold spontaneous conversion to the amorphous HCl/H_2O phase of the same average composition is observed. HH is therefore metastable at $T \geq 173$ K.

(iii) HH growth is limited by mass transport of HCl across the nascent crystalline phase at the interface. Beyond a thickness of roughly 450 formal monolayers or a dose of 3.5×10^{17} molecules of HCl crystalline growth comes to a gradual halt. Conversely, for NAT and NAD no such mass transport limitation has been observed at the same dose of HNO_3.

(iv) The required H_2O supersaturation for pure ice growth (I_h) on a SiO_2 surface strongly increases with decreasing temperature with an apparent activation energy of 21 kJ/Mol. This activation energy is approximately equal to the activation energy for (surface) diffusion of H_2O on acrystalline ice surface.

(v) The condition for nucleating the (metastable) crystalline HH phase ($T \leq 173$ K) rules out the occurrence of HH in the terrestrial atmosphere under UT/LS conditions: The stable atmospheric HCl/H_2O phase is most likely amorphous which has the advantage that heterogeneous reactions involving HCl such as the important reaction $ClONO_2 + HCl \rightarrow Cl_2(g) + HNO_3(ads.)$ occur at a higher efficiency compared to a crystalline surface.

A novel optical freezing array for the examination of cooling rate dependence in heterogeneous ice nucleation

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Homogeneous ice nucleation is a stochastic process, implying that it is not only temperature but also time dependent. For heterogeneous ice nucleation it is still under debate whether there is a significant time dependence or not. In case of minor time dependence it is probably sufficient to use a singular or slightly modified singular approach, which mainly supposes temperature dependence and just small stochastic variations.

We contribute to this discussion using a novel optical freezing array termed BINARY (Bielefeld Ice Nucleation ARraY). The setup consists of an array of microliter-sized droplets on a Peltier cooling stage. The droplets are separated from each other with a polydimethylsiloxane (PDMS) spacer to prevent a Bergeron-Findeisen process, in which the first freezing droplets grow at the expense of the remaining liquid ones due to their vapor pressure differences. An automatic detection of nucleation events is realized optically by the change in brightness during freezing.

Different types of ice nucleating agents were tested with the presented setup, e. g. pollen and clay mineral dust. Exemplarily, cooling rate dependent measurements are shown for the heterogeneous ice nucleation induced by Snomax®.

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Visualization of the freezing process of aqueous organic solutions

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Upon freezing aqueous solutions separate into ice and a residual freeze-concentrated solution (FCS)[1-10] which freezes[2,3] or vitrifies[4-7] upon further cooling. The morphology and mutual distribution of ice and FCS are essential in fields ranging from food industry[4,5] and pharmaceutics[6,7] to geophysics[8-10]. Yet the freezing process, which governs the formation of ice/FCS, remains not fully understood. In our work, using cryo-microscopy and differential scanning calorimetry we show that aqueous citric acid (CA) and sucrose freeze in two stages, fast and sluggish, and the whole freezing process yields a continuous ice framework (IF) and two FCS regions of different concentration, FCS_1 and FCS_2, which vitrify at two distinct temperatures, $T_{g1,c}>T_{g2,c}$. The FCS_1 is a maximally FCS (MFCS) and interpenetrates the IF, whereas the less concentrated FCS_2 envelops the whole IF/FCS_1. Sluggish freezing proceeds in FCS_2 until it is terminated by the FCS_2-glass transition, $T_{g2,c}$, and then recommences upon warming above a reverse glass-FCS_2 transition, $T_{g2,w}$. The recommenced sluggish freezing in FCS_2 together with a reverse glass-FCS_1 transition, $T_{g1,w}$, produce the T_{tr2}-transition[4], which has remained misunderstood for decades[4,5]. Our results and videos demonstrate how ice is formed during freezing, expelling solute from the ice lattice and producing a continuous IF immersed into FCS_1 and enveloped by FCS_2.

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Homogeneous Freezing of Water Droplets and its Dependence on Droplet Size

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The formulation and parameterisation of microphysical processes in clouds, such as phase transitions, still presents a challenge for weather and climate models. This includes the homogeneous freezing of supercooled water droplets which is an important process in deep convective systems, where almost pure water droplets may stay liquid until homogeneous freezing occurs at temperatures around 238 K. Though the homogeneous ice nucleation in supercooled water is considered to be well understood, recent laboratory experiments with typical cloud droplet sizes showed one to two orders of magnitude smaller nucleation rate coefficients than previous literature results. The discrepancies motivated us to re-analyse homogeneous droplet freezing experiments conducted during the previous years at the AIDA (Aerosol Interactions and Dynamics in the Atmosphere) chamber. This cloud chamber has a volume of 84 m3 and operates under atmospherically relevant conditions within wide ranges of temperature, pressure and humidity, whereby investigations of both tropospheric mixed-phase clouds and cirrus clouds can be realised. By controlled adiabatic expansions, the ascent of an air parcel in the troposphere can be simulated.

The new AIDA results, together with the results from single levitated droplet experiments, now allow the conclusion that freezing of droplets even with diameters as low as a few micrometers is a volume dominated process. Furthermore, a contribution of surface induced freezing can be ruled out.

The obtained volume nucleation rate coefficients from AIDA experiments are in good agreement, within error bars, with some previous literature data, including earlier AIDA results, but they do not agree with recently published lower coefficients.

This contribution will show the results from the re-analysis of AIDA homogeneous freezing experiments with pure water droplets and will discuss the comparison to the literature data.

Determination of Collision Efficiency from Contact freezing of Silver Iodide

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Collision efficiency of cloud droplets with aerosol particles is one of the topics of importance for scavenging of aerosol in the atmosphere. Various studies had been carried out by different groups but it has not been possible to measure collision efficiency for temperatures below 0 ºC. Collision efficiency is important for contact freezing in these conditions. In our laboratory, we built the Collision Nucleation CHamber (CLINCH) (Ladino et al. 2011) in which water droplet can collide with aerosol particles. In this study, we are attempting to derive empirical collision efficiency with the help of contact freezing of silver iodide particles. Silver iodide has been known for its ice nucleation ability since 1940s (Vonnegut 1947) and we are assuming each collision between silver iodide particle and water droplet will lead to freezing of water droplet in the range temperature range -28 to -36 ºC. In CLINCH, size selected 200 nm silver iodide particles collide with water droplets of 80 µm diameter. With the extension in chamber length it is possible to vary the interaction time between particles and the droplets. Our experiments are performed between -10 to -36 ºC for various concentrations of particles and different interaction times. Number of collisions between a single droplet and several aerosol particles can be calculated at the experimental conditions. The importance of phoretic forces will be investigated as proposed by Santachiara et al (2012).

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A summary of heterogeneous ice nucleation results from the AIDA cloud simulation chamber

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The formulation and parameterization of primary ice formation in tropospheric clouds is still one of the most incomplete and uncertain parts in cloud, weather and climate models. This includes homogeneous freezing of water and solution droplets, though this part is considered to be well understood and formulated for models, as well as the various modes of heterogeneous ice nucleation like deposition nucleation, condensation freezing, immersion freezing and contact freezing, which involve solid atmospheric aerosol particles as heterogeneous ice nuclei.

During the recent years, numerous laboratory experiments of heterogeneous ice nucleation have been conducted for different aerosol types, but it is still not clear which are the most relevant aerosol types and most appropriate formulations for heterogeneous ice nucleation processes to be considered in models. Some of the parameterisations rely on the classical nucleation rate theory, others describe the ice nucleation activity as function of ice-active sites on the aerosol particle surface.

This contribution will introduce the ice nucleation active site (INAS) density concept as a simple straight forward approach for the model parameterisation of heterogeneous ice nucleation of various aerosols on the basis of laboratory cloud simulation experiments with the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) Aerosol and Cloud chamber facility at the Karlsruhe Institute of Technology. A few minutes will also be spent to introduce recent findings on the pre-activation in glassy and crystalline organic aerosols.

First results obtained with the new portable ice nuclei counter SPIN

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Aerosol particles known as ice nuclei (IN) play an important role in the atmosphere in a number of processes related to cloud formation and cloud microphysics. Despite the progress within ice nucleation research during the past decade, much is still unknown. There is a particular need for data on atmospheric IN number concentrations from observations, as only a small number of these observations exist, while they would provide valuable input for atmospheric modelling (DeMott et al., 2010). In situ measurements with portable IN counters are an important way to gain knowledge in this field. The Spectrometer for Ice Nuclei (SPIN) is a new commercially available portable IN counter manufactured by Droplet Measurement Technologies, Inc. SPIN is a continuous flow diffusion chamber with parallel plate geometry following the design of the Portable Ice Nuclei Chamber (PINC) (Chou et al., 2011). The aerosol sample flows through a chamber where a supersaturation of water vapor with respect to ice is obtained by keeping two ice covered walls at different temperatures (below 0 °C). The aerosol sample is then exposed to an isothermal evaporation section for evaporation of liquid droplets before particle detection with a linear depolarisation optical particle counter (OPC). With SPIN, it is possible to investigate heterogeneous ice nucleation in both deposition nucleation and condensation/immersion freezing modes. We have tested the SPIN chamber with well characterized ice nuclei such as illite and SNOMAX™, as well as investigated the homogeneous freezing of ammonium sulphate particles. Here we present exemplary results of these studies.

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Investigating the atmospheric ice phase at the high altitude Jungfraujoch site

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One of the major sources of uncertainty in climate model projections is an insufficient understanding of aerosol-cloud interactions. Particularly poorly understood is the formation of mixed-phase clouds (MPC), where ice nuclei (IN), a small but important subset of aerosol particles, facilitate heterogeneous ice nucleation. The resulting cloud glaciation enhances precipitation formation, decreasing cloud cover and lifetime, and affects cloud radiative properties.

To investigate properties of IN in MPCs, small, freshly formed ice crystals must be separated from interstitial particles and supercooled droplets. The ice residuals (IR) contained within the crystals can then be analyzed. The new Ice Selective Inlet (ISI) has been designed to extract small ice crystals from mixed-phase clouds, simultaneously counting, sizing and imaging the hydrometeors contained in the cloud with the use of optical particle spectrometers. The core of the ISI is an evaporation unit with ice-covered inner walls, removing droplets using the Wegener-Bergeron-Findeisen process, while transmitting the ice crystals.

The ISI was deployed at the High Alpine Jungfraujoch Research Station (3580 m.a.s.l) during winter 2014 as part of the CLACE 2014 field campaign. The campaign included comprehensive measurements of cloud microphysics and aerosol properties. A host of online aerosol instrumentation was deployed downstream of the ISI to analyze the physical and chemical characteristics of IR. These included a Grimm OPC, a scanning mobility particle sizer (SMPS) and an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) for number size distribution measurements and a single particle soot photometer (SP2) and Waveband Integrated Bioaerosol Sensor (WIBS-4) for analysis of the chemical composition, with particular focus on the content of black carbon (BC) and biological particles in IR. IR were also collected using a single-stage impactor for scanning electron microscopy and scanning transmission x-ray microscopy analysis. Corresponding instrumentation sampled through a total aerosol inlet. By comparing observations from the ISI with those from the total inlet the characteristics of ice residuals relative to the total aerosol could be established. First results from these analyses will be presented.

In-situ detection of ice nuclei concentrations in the free troposphere in the deposition and condensation regime

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Several laboratory studies have shown the efficiency of mineral dust particles to act as ice nuclei (IN) at temperatures below -10°C [1]. The interest in ambient IN number concentration as an input parameter for global and regional climate models is high due to the important role of ice containing clouds in the Earth's radiative balance. However, few in-situ studies of atmospheric IN concentrations have been conducted and up to date none close to a main global dust source such as the Sahara.

In the current work we present IN concentration data from two distinct locations, namely the Jungfraujoch in Switzerland and the Izaña Observatory on Tenerife, Spain. Both locations are high altitude research stations (Jungfraujoch: 3580 m a.s.l., Izaña: 2373 m a.s.l.). During winter, the Jungfraujoch is considered representative for being in the free troposphere as is Izaña during night time if it is not located in the Saharan air layer (SAL). Three field studies have been conducted; two of them at the Jungfraujoch during January/February 2013 and 2014, and one at Izaña during August 2013. Measurements were made using the Portable Ice Nucleus Counter (PINC) [2], a continuous flow diffusion chamber.

IN concentration data in the deposition and condensation mode at various temperatures will be presented for both locations. The results from Jungfraujoch show that IN concentrations in the deposition mode were extremely low, with on average less than 1 IN/l at a temperature of T = 241 K and a relative humidity of 127 % with respect to ice (RHi). At Izaña, mean IN concentrations were usually below 10 IN/l as well for the same measurement conditions but increased up to 32 IN/l during a strong Sahara dust event. At the same temperature, IN concentrations were much higher in the condensation freezing regime, where they reached values of 1200 IN/l during the dust event.

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Characterization of the Micro-Orifice Uniform Deposit Impactor-droplet freezing technique (MOUDI-DFT) for size-resolved quantitative measurements of ice nuclei

Ryan Mason (1), Meng Si (1), Jixiao Li (2), J. Alex Huffman (2), Christina McCluskey (3), Ezra Levin (3), Victoria Irish (1), Cédric Chou (1), Thomas Hill (3), Luis Ladino (4), Jacqueline Yakobi (4), Corinne Schiller (5), Jon Abbatt (4), Paul DeMott (3), and Allan Bertram (1)

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Ice formation within a cloud system can significantly modify its lifetime and radiative forcing. Many current instruments for measuring atmospheric concentrations of ice nuclei (IN) are not capable of providing size-resolved information. Such knowledge is useful in identifying the sources of IN and predicting their transport in the atmosphere. Furthermore, those that use size-discrimination to identify IN typically exclude particles with an aerodynamic diameter greater than 2.5 μm from analysis. Several studies have indicated this may be an important size regime for IN, particularly with those activating at warmer temperatures.

The recently developed Micro-Orifice Uniform Deposit Impactor-droplet freezing technique (MOUDI-DFT) addresses these limitations through combining sample collection by a model of cascade impactor with an established immersion freezing apparatus. Here we present a characterization of the MOUDI-DFT and the development of a modified technique which address experimental uncertainties arising from sample deposit inhomogeneity and the droplet freezing method. An intercomparison with a continuous-flow diffusion chamber (CFDC) was performed.

We also show preliminary results from a campaign undertaken in a remote coastal region of western Canada. Correlations between atmospheric IN concentrations and the abundance of suspended submicron and supermicron particles, biological aerosols, carbonaceous aerosols, and prevailing meteorological conditions were investigated.

Research on Airborne Ice-Nucleating Species (RAINS): A Collaborative Research Project Funded by the National Science Foundation

David G. Schmale III[1], Brent C. Christner[2], Cindy Morris[3,4], David C. Sands[4], Boris A. Vinatzer[1], and Carolyn F. Weber[5]

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Microorganisms are abundant in the atmosphere and play important roles in cloud development, cloud chemistry, and ultimately weather. Some microorganisms produce proteins that catalyze the nucleation of ice crystals at significantly warmer temperatures than would normally be required for ice formation. Little is known about the abundance, diversity, and flux of ice-nucleating microorganisms in the atmosphere. Our collaborative research project (Research on Airborne Ice-Nucleating Species or RAINS), aims to examine the diversity and function of ice-nucleating microorganisms in the atmosphere. We are characterizing the taxonomic, genetic, and functional diversity of ice-nucleating microorganisms in rain, snow and air collected from multiple sites in the U.S. and in France. Our research will increase our knowledge of the associations of microorganisms with rain, transforming the way society perceives the relationships between microorganisms and weather. The project will also sponsor an international workshop for early career scientists to highlight recent advances and identify the outstanding scientific questions surrounding microbes at the interface of land-atmosphere feedbacks.

Expanding our Knowledge of Ice Nucleation Activity in the Fungal Genus Fusarium: From Biological Research Collections to Atmospheric Sampling Missions with Drones

David G. Schmale III[1], Cindy Morris[2], Emiliano Stopelli[2,3], and Claudia Bartoli[2,4]

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The fungal genus Fusarium contains a number of important plant and animal pathogens, mycotoxin producers, and ice nucleators. We used an LED-based Ice Nucleation Detection Apparatus (LINDA) to examine the ice nucleation activity (INA) of isolates of Fusarium from a large biological research collection (over 60 different species) and from atmospheric sampling missions with drones 100m above the ground in Blacksburg, VA USA (at least 12 species). Preliminary results indicated that seven of the isolates from the biological research collection (representing seven different species) were INA positive at temperatures between -12°C and -5°C. Isolates of three of these species (F. avenaceum, F. acuminatum, and F. tricinctum) have been reported as INA positive in previous studies. However, to our knowledge, the INA of the remaining four isolates (F. begoniae, F. langsethiae, F. armeniacum, and F. concentricum) has not been reported previously. One of the isolates from the drone collections (F156N33, F. avenaceum) was INA positive at about -6°C. This work expands our knowledge of INA in the fungal genus Fusarium, and engenders new hypotheses about the aeroecology of ice-nucleating fusaria in the upper atmosphere (100m above ground level and beyond).

Cloud Conditions Favoring Secondary Ice Particles in Tropical Maritime Convection

Andrew Heymsfield

National Center for Atmospheric Research, 3090 Center Green Drive, Boulder, CO 8030, USA

Progress in understanding the formation of ice in lower tropospheric clouds is slowed by the difficulties in characterizing the many complex interactions that lead to ice initiation and to the dynamic, non-steady state nature of the clouds. The present study characterizes the conditions where secondary ice particles, specifically identified as needle or thin columnar types, are observed in tropical maritime convection during the Ice in Clouds Experiment-Tropical (ICE-T), based out of St. Croix, V. I., and the NASA AMMA experiment (NAMMA) in 2006 sampling from Cape Verde, Africa. The properties of the cloud droplet populations relevant to the secondary ice production process, and the ice particle populations, are characterized as a function of temperature and vertical velocity. These secondary ice particles are observed primarily in regions of low liquid water content and weak vertical velocities. Two relatively unique situations are examined in detail. First, ice formation is examined by following the tops of a group of ICE-T chimney clouds as they ascend and cool from a temperature of +7°C to –8°C, examining the production of the first ice. Then, using the data from a cloud system sampled during NAMMA, we elucidate a process that promotes ice multiplication. The intention is that this study willlead both to a better understanding of how secondary ice production proceeds in natural clouds as well as to more realistic laboratory studies of the processes involved.

Nucleation and ice growth on charged meteor smoke particles

Denis Duft

Karlsruher Institut für Technologie (KIT), Institut für Meteorologie und Klimaforschung, Atmosphärische Aerosolforschung (IMK-AAF)

Meteoric smoke particles (MSP) are believed to act as heterogeneous ice nuclei under the temperature conditions in the polar mesopause region leading to the formation of polar mesospheric clouds (PMC). Up to date little is known of the microphysical properties of these nanoscale particles. Their composition is assumed to be represented by the abundance of elements found in meteors vaporized in the lower thermosphere region. In this contribution we present an experimental setup designed to study fundamental microphysical properties of such particles in the laboratory together with first measurements on ice nucleation and ice growth abilities.

Studying nucleation processes with computer simulations: from freezing to cavitation

Christoph Dellago

Faculty of Physics, University of Vienna, Vienna, Austria

Under suitable conditions, first order phase transitions such as the freezing of a liquid or the structural transformation of a solid occur via a nucleation and growth mechanism in which a nucleus of the thermodynamically preferred phase forms in the metastable phase. This process is opposed by a barrier related to the free energetic cost of the interface between the two phases, leading to nucleation times that typically exceed the characteristic times of basic molecular motions by many orders of magnitude. This wide separation of time scales complicates computer simulations of nucleation processes. In this talk, I will discuss how biased sampling in configuration space and trajectory space can be used to address this challenging problem. Using the crystallization of supercooled water and cavitation of water under negative pressure a examples, I will show how molecular simulations can be employed to determine nucleation rates and obtain microscopic information about the nucleation mechanism.

Transferable coarse-grained potential for de novo protein folding and design

Ivan Coluzza

Computational Physics, University of Vienna, Boltzmanngasse 5, A-1090 Wien

Protein folding and design are major biophysical problems, the solution of which would lead to important applications especially in medicine. Here a novel protein model capable of simultaneously provide quantitative protein design and folding is introduced. With computer simulations it is shown that, for a large set of real protein structures, the model produces designed sequences with a remarkable similarity to the corresponding natural occurring sequences. For an independent set of proteins, notoriously difficult to fold, the correct folding of both the designed and the natural sequences is also demonstrated. The folding properties are characterized by free energy calculations. which not only are consistent among natural and designed proteins, but we also show a remarkable precision when the folded structures are compared to the experimentally determined ones. Ultimately, this novel coarse-grained protein model is unique in the combination of its fundamental three features: its simplicity, its ability to produce natural like designed sequences, and its structure prediction precision. The latter demonstrated by free energy calculations. It is also remarkable that low frustration sequences can be obtained with such a simple and universal design procedure, and that the folding of natural proteins shows funnelled free energy landscapes without the need of any potentials based on the native structure.

Polarizable Force Fields Improve Models of Ice Nucleation

Michael Schauperl, Roland G. Huber, Christian Kramer, Klaus R. Liedl

Institute of General, Inorganic and Theoretical Chemistry/Theoretical Chemistry and Center of Molecular Bioscienes (CMBI), University of Innsbruck, Austria.

Ice formation plays a major role in a variety of research fields ranging from biology to physics.[1] In the last decades the importance of heterogeneous ice nucleation was emphasized by its crucial role in climate science.[2] Recent studies report a range of particles influencing the nucleation process.[3] Despite the fact that understanding ice nucleation is a key challenge for climate science, it is not fully clear how the chemical properties of the nuclei surface influence the prosperity to form ice.

To generate insight in the ice nucleation process a range of theoretical tools have been used including Molecular Dynamics simulations.[4] However, deficiencies of classic biomolecular force fields (fixed point charge, no polarization) have become increasingly evident. Therefore, new types of force fields that include terms for polarizability and anisotropic charge distribution such as AMOEBA[5] (Atomic Multipole Optimized Energetics for Biomolecular Applications) enable new possibilities for inquiry. Polarization effects, which are not covered by conventional force fields, are of major interest, as atoms are able to react to their environment[6] resulting in a force field which can be used to describe molecules in gas- and condensed liquid and solid phases.

We show how polarizable multipole force fields improve the description of phase transitions over conventional force fields. Subsequently, we examine ordering processes of water molecules around simple chemical systems and demonstrate the importance of polarization for these phenomena.

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Molecular Dynamics of Ice Nucleation

Roland G. Huber, Michael Schauperl, Klaus R. Liedl

Department of Theoretical Chemistry, Faculty of Chemistry and Pharmacy, Center for Molecular Biosciences Innsbruck (CMBI), Leopold Franzens University Innsbruck, Innrain 80/82, 6020 Innsbruck, Austria

Understanding ice nucleation is key in a variety of natural and industrial processes. The importance of ice nucleation is exemplified by its crucial role in cold adaption of plant and animal life as well as in local and global atmospheric phenomena.[1] These phenomena have significant environmental and commercial implications, as they influence weather events such as precipitation (e.g. hailstorms, snowfall) as well as broader implications for the global climate. Therefore, an in-depth understanding of the atomic-scale processes involved in ice nucleation has far-reaching implications in a wide variety of academic and commercial endeavors.[2]

Molecular dynamics simulations are a well-established tool to investigate physicochemical processes on the picosecond to millisecond time scales at atomistic resolution. Environmental conditions, e.g. pressure, temperature gradients or contaminants can be easily modeled. Observed phenomena are analyzed using statistical thermodynamics in order to infer macroscopic systems behavior.

We currently investigate ice nucleation around protein contaminants of various sizes and experimentally determined nucleation potential. Biogenic nuclei are known to form an important part of air particulate matter.[3,4] We try to determine the relative magnitude of nucleation propensity induced by either particle size or specific pre-ordering induced by directed chemical interactions of the protein contaminant on the surrounding solvation shells. We determine localized solvent entropy[5] at various points around the contaminant while we subject the system to simulated cooling/heating cycles.

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On the usage of classical nucleation theory in quantification of the impact of bio-aerosols on weather and climate, Modeling study

M. Sahyoun[1,2], N.W. Nielsen[1], J.H. Sørensen[1], K. Finster[2], U.G. Karlson[3], T. Šantl-Temkiv[4], S. Augustin[5], S. Hartmann[5], D. Niedermeier[5], F. Stratmann[5], H. Wex[5], and U.S. Korsholm[1]

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[4]{Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark},
[5]{Leibniz Institute for Tropospheric Research (TROPOS), Leipzig, Germany}

Some bacteria, e.g. Pseudomonas syringae, have previously been found efficient in nucleating ice heterogeneously at temperatures close to -2°C in laboratory tests. They are also present in the atmospheric boundary layer for a residence time on the order of several days. Therefore, ice nucleation active (INA) bacteria may be involved in the formation of ice and precipitation in mixed phase clouds, and could potentially influence weather and climate. Investigations into the impact of INA bacteria on climate have shown that emissions are too low to significantly have an impact or at least still questioned.

The goal of this study is to investigate the usability of ice nucleation rate parameterization based on classical nucleation theory (CNT) which is widely used in climate models.
Two parameterizations to describe the heterogeneous ice nucleation were compared. Both parameterizations were implemented and tested in a 1-d version of a weather model in different meteorological cases. The first parameterization is based on CNT (CH08) and is a function of temperature and the size of the IN. The second parameterization (HAR13) was derived from nucleation measurements. HAR13 is a function of temperature and it implicitly considers the number of protein complexes attached to either intact bacteria cell or membrane fragments of the cell. The sensitivity of meteorological variables to changes in the relative fraction of cloud droplets containing INA bacteria or fragments here of was investigated.

In this study, HAR13 was found to produce more cloud ice and precipitation than CH08 as the bacterial fraction increases. The ice production using HAR13 was found to be sensitive to the change of the bacterial fraction while CH08 did not show such sensitivity. In classical theory, having larger surface size leads to more active sites. This fact is true in the case of using dust as IN, but it’s not necessarily applicable in the case of bacteria. In IN bacteria, the biological microstructure analysis suggests that singular protein complexes are the locations where ice nucleation occurs. The number of protein complexes does not scale with the surface area of the cell. As a result, dealing with bacteria in a classical way similar to dust is not adequate. The number of proteins is a more important parameter than the size of IN to be considered when modelling the freezing of bacteria and it’s likely that CNT is still missing that.

Sensitivity of the classical nucleation theory on thermodynamic and kinetic parameters

Luisa Ickes(1), André Welti(1), Corinna Hoose(2) and Ulrike Lohmann(1)

(1) Institute for Atmospheric and Climate Science, ETH, Zurich, Switzerland

(2) Institute for Meteorology and Climate Research, KIT, Karlsruhe, Germany

Classical nucleation theory (CNT) can be used to parameterize ice nucleation in cloud droplets as a stochastic process based on thermodynamic and kinetic parameters. Due to the lack of a complete physical model for liquid water, several thermodynamic and kinetic parameters are not known leading to various formulations of CNT. The aim of the presented work is to review the different estimates of thermodynamic and kinetic parameters currently used in the research community and to investigate the sensitivity of the nucleation rate on the choice of parameters. By evaluating experimental data sets of droplet freezing it is possible to narrow down a reasonable range for the parameters. This helps to develop CNT-based parameterizations for freezing of droplets in regional and global climate models, which will improve the representation of cold clouds.

Buffet

Saturday 26th April 2014, 12:00 (Lunch)

Snack:

- Open sandwich and finger food (salad with strips of beef and paprika, red onion and seed oil, mozzarella with tomatoes and basil pesto or chicken liver mousse on port jelley with red pepper)

Saturday 26th April 2014, 18:55 (Dinner)

Starter:

- Beef broth with vegetable chunks and rusks
- Ruccula foam soup with cream topping and cardamon

Main Dish:

- Ragout from Falkenstein venison with root vegetables, star anise, napkin dumplings and lingonberries
- Roasted chicken breast on lentils ragout, rosemary potatoes and baby tomatoes
- Bulgur-Curry-Pan with vegetable, roasted almonds and Yoghurt-Chili-Dip
- Fresh salads from the Buffet

Dessert:

- Cake buffet with 3 different cakes (cream cake, poppy-curd cheese-raspberry- and nougat slice)

Sunday 27th April 2014, 13:00 (Lunch)

Snack:

- Open sandwich and finger food (salad with strips of beef and paprika, red onion and seed oil, mozzarella with tomatoes and basil pesto or chicken liver mousse on port jelley with red pepper)

Accommodation

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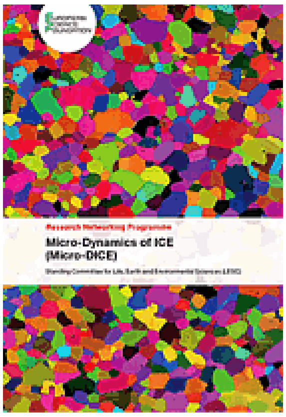

Micro-Dynamics of Ice (Micro-DICE)
microdice.eu

Vienna University of Technology
www.tuwien.ac.at

Annex 4b: 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	: 2nd Workshop on Atmospheric Ice Nucleation
Location	: Vienna
Date of Science Meeting	: 26/04/2014 - 27/04/2014

Add a Convenor

Convenor(s) 3

Name	City, Country	Type		
Professor Hinrich Grothe	Vienna, (AT)	Convenor	Edit	
Professor Regina Hitzenberger	Vienna, (AT)	Convenor	Edit	DELETE
Professor Thomas Koop	Bielefeld, (DE)	Convenor	Edit	DELETE

Add a Speaker

Speakers 28

Name	City, Country	Type		
Miss Stephanie Augustin	Leipzig, (DE)	Speaker	Edit	DELETE
Dr. Anatoli Bogdan	Innsbruck, (AT)	Speaker	Edit	DELETE
Ms. Yvonne Boose	Zürich, (CH)	Speaker	Edit	DELETE
Dr. Carsten Budke	Bielefeld, (DE)	Speaker	Edit	DELETE
Dr. Ivan Coluzza	Vienna, (AT)	Speaker	Edit	DELETE
Dr. Denis Duft	Eggenstein-Leopoldshafen, (DE)	Speaker	Edit	DELETE
Dr. Sebastien Facq	Cambridge, (UK)	Speaker	Edit	DELETE
Dr. Janine Fröhlich	Mainz, (DE)	Speaker	Edit	DELETE
Dr. Andrew Heymsfield	Boulder, (US)	Speaker	Edit	DELETE
Mr. Roland Huber	Innsbruck, (AT)	Speaker	Edit	DELETE
Miss Luisa Ickes	Zürich, (CH)	Speaker	Edit	DELETE
Ms. Karoliina Ida Aleksandra Ignatius	Leipzig, (DE)	Speaker	Edit	DELETE
Mr. Sandy James	Leeds, (UK)	Speaker	Edit	DELETE
Mr. Piotr Kupiszewski	Villingen PSI, (CH)	Speaker	Edit	DELETE
Mr. Ryan Mason	Vancouver, (CA)	Speaker	Edit	DELETE
Mr. Ottmar Möhler	Karlsruhe, (DE)	Speaker	Edit	DELETE
Mr. Baban Nagare	Zürich, (CH)	Speaker	Edit	DELETE
Mr. Joseph Niehaus	Houghton, (US)	Speaker	Edit	DELETE
Dr. Bernhard G. Pummer	Mainz, (DE)	Speaker	Edit	DELETE
Professor Michel J. Rossi	Villingen, (CH)	Speaker	Edit	DELETE
Mr. Maher Sahyoun	Copenhagen N, (DK)	Speaker	Edit	DELETE
Dr. Tina Santl Temkiv	Aarhus, (DK)	Speaker	Edit	DELETE
Mr. Michael Schauperl	Innsbruck, (AT)	Speaker	Edit	DELETE
Dr. David Schmale	Blacksburg, (US)	Speaker	Edit	DELETE
Ms. Thea Schmitt	Eggenstein-Leopoldshafen, (DE)	Speaker	Edit	DELETE
Mr. Emiliano Stopelli	Basel, (CH)	Speaker	Edit	DELETE
Dr. Andrè Welti	Zürich, (CH)	Speaker	Edit	DELETE
Mr. Tobias Zolles	Vienna, (AT)	Speaker	Edit	DELETE

Add a Participant

Participants 17

Name	City, Country	Type		
Mr. Thomas Berkemeier	Mainz, (DE)	Participant	Edit	DELETE
Mr. Henner Bieligh	Leipzig, (DE)	Participant	Edit	DELETE
Mrs. Katharina Dreischmeier	Bielefeld, (DE)	Participant	Edit	DELETE
Mrs. Laura Felgitsch	Vienna, (AT)	Participant	Edit	DELETE
Mr. Lukas Kaufmann	Zürich, (CH)	Participant	Edit	DELETE
Dr. Alexei Kiselev	Eggenstein-Leopoldshafen, (DE)	Participant	Edit	DELETE
Mr. Klaus Liedl	Innsbruck, (AT)	Participant	Edit	DELETE

Mrs. Meilee Ling	Aarhus, (DK)	Participant	Edit	DELETE
Professor Thomas Loerting	Innsbruck, (AT)	Participant	Edit	DELETE
Professor Werner Lubitz	Kritzendorf, (AT)	Participant	Edit	DELETE
Professor Ulrich Poeschl	Mainz, (DE)	Participant	Edit	DELETE
Professor Yinon Rudich	Rehovot, (IL)	Participant	Edit	DELETE
Professor Otto Schrems	Bremerhaven, (DE)	Participant	Edit	DELETE
Dr. Martin Simmel	Leipzig, (DE)	Participant	Edit	DELETE
Dr. Martin Tollinger	Innsbruck, (AT)	Participant	Edit	DELETE
Miss Romy Ullrich	Karlsruhe, (DE)	Participant	Edit	DELETE
Dr. Heike Wex	Leipzig, (DE)	Participant	Edit	DELETE

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