4th Workshop – Microphysics of ice clouds

Vienna- Austria

16th and 17th of April 2016

Book of Abstracts

TECHNISCHE UNIVERSITÄT WIEN
Vienna University of Technology

Preface

Dear Workshop Participant,

It is my pleasure to welcome you to Vienna for our 4th workshop 'Microphysics of Ice Clouds'. The aim is to bring together different areas of atmospheric ice research: field measurements, laboratory studies and modelling. The joint topic is the nucleation of ice in clouds and ice cloud properties. We will focus on methods and open questions concerning the microstructure and dynamics of the ice formation processes as well as the resulting ice clouds in the atmosphere discussing experimental and theoretical methods including chemistry and microphysics.

This workshop will be held on the weekend before the start of the EGU General Assembly 2016 in the Vienna Austria Centre. Additionally, the organizers of this workshop are also offering a session at the EGU conference, which is called “Atmospheric Ice Particles” AS3.3 (on Wednesday in room F2 at 8:30; this session has been ongoing for six years). The workshop and the conference session have the same topic but will not have any overlap concerning presentations. The main aim of the workshop is to provide an open, deep discussion of relevant topics, i.e. ice nucleation and ice cloud properties, which are difficult to discuss in a typical conference setting. The workshop emphasizes opportunities for early career professionals to present their recent research, some of them got a travel support by the EGU.

Again, we have 'breakout discussions'. Speakers from each of the sessions will be the 'team leaders' at small breakout discussion groups. They will be charged with leading the discussion of identifying research needs, and reporting back to the group at lunch on the last day of the meeting.

Vienna 16th April 2016

Hinrich Grothe

Organizers

Local Organizers

Prof. Dr. Hinrich Grothe
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Location

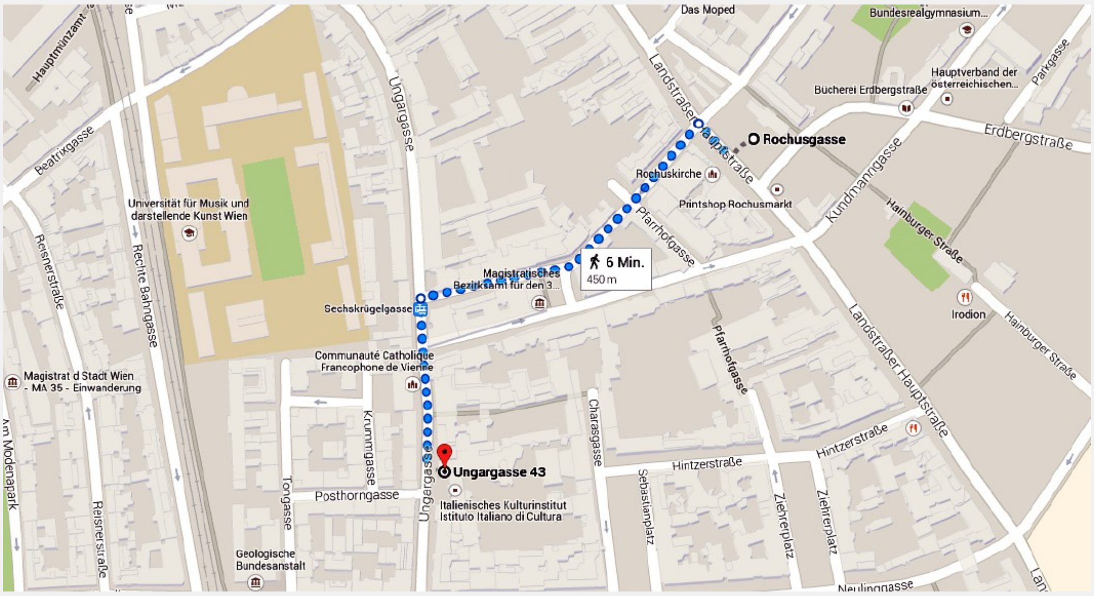
Vienna University of Technology

The workshop is held in the "Italian Culture Institute - Istituto Italiano di Cultura" at Ungargasse 43, 1030 Vienna, which is easily accessible by public transport.

Istituto Italiano di Cultura

Map:

Rochusgasse to Ungargasse



Transportation

Sessions

Saturday 16th April			
12:00 - 13:00	Registration and Buffet		
13:00 - 13:10	Opening of the workshop	Hinrich Grothe (Vienna University of Technology)	
		Ernst Kanitz (Italian Culture Institute, Vienna)	
Mixed Session			
13:10 - 13:20	1st Talk	David Schmale	Ice-nucleating strains of Pseudomonas syringae from a large freshwater lake in Virginia, USA
13:20 - 13:25	Discussion		
13:25 - 13:35	2nd Talk	Daniel O'Sullivan	The adsorption of fungal ice-nucleating proteins on mineral dusts: a terrestrial reservoir of atmospheric ice-nucleating particles
13:35 - 13:40	Discussion		
13:40 - 13:50	3rd Talk	Larissa Lacher	Ice nucleating particles on the High Altitude Research Station Jungfraujoch: 5 years of measurements
13:50 - 13:55	Discussion		
13:55 - 14:05	4rd Talk	Tani Satyanarayana	Utilizing radar data in support of the hail mitigation operations in Styria, Austria
14:05 - 14:10	Discussion		
14:10 - 14: 35	Break Out Discussion (Coffee &Cakes)		
Atmospheric Measurements I			
14:35 - 14:45	1st Talk	Yvonne Boose	Ice nucleating particle properties in the Saharan air layer
14:45 - 14:50	Discussion		
14:50 - 15:00	2nd Talk	Jessie Creamean	The relationships between insoluble precipitation residues, clouds, and precipitation over California's Sierra Nevada during winter storms
15:00 - 15:05	Discussion		
15:05 - 15:15	3rd Talk	Monika Kohn	Measuring the effect of pollen grains and their ice active macro-molecules by observations of the INP concentration during the birch pollen season
15:15 - 15:20	Discussion		
15:20 - 15:30	4th Talk	Craig Powers	An unmanned surface vehicle (USV) to study the air/water interface in aquatic environments
15:30 - 15:35	Discussion		
15:35 - 15:50	General Discussion		
15:50 - 16:15	Coffee Break		

Atmospheric Measurements II			
16:15 - 16:25	1st Talk	Emiliano Stopelli	Exploring the sources of variability in ice nucleating particles of biological origin
16:25 - 16:30	Discussion		
16:30 - 16:40	2nd Talk	Daniel Weber	Ice nucleating particles measured during the field intercomparison FIN-3 by the vacuum diffusion chamber FRIDGE
16:40 - 16:45	Discussion		
16:45 - 16:55	3rd Talk	Bernadett Weinzierl	Modification of mineral dust during long-range transport and associated impact on predicted ice nuclei concentration - insights from SALTRACE
16:55 - 17:00	Discussion		
17:00 - 17:25	Break Out Discussion (Coffee &Cakes)		

Laboratory Measurements I			
17:25 - 17:35	1st Talk	Sandy James	A laboratory study of the nucleation kinetics of nitric acid hydrates under stratospheric conditions
17:35 - 17:40	Discussion		
17:40 - 17:50	2nd Talk	Philipp Baloh	Metastable nitric acid trihydrate in ice clouds
17:50 - 17:55	Discussion		
17:55 - 18:05	3rd Talk	Peter Alpert	Analysis of isothermal and cooling-rate-dependent immersion freezing by a unifying stochastic ice nucleation model
18:05 - 18:10	Discussion		
18:10 - 18:20	4th Talk	Bertrand Chazallon	Air-plane soot surrogates and their ice nucleation activity: investigations in deposition mode
18:20 - 18:25	Discussion		
18:25 - 18:40	General Discussion		
18:40 - open end	Dinner & After-Dinner Workshop (optional)		

Sunday 17th April			
Laboratory Measurements II			
08:30 - 08:40	1st Talk	Mark Holden	Understanding ice nucleation by alkali feldspars
08:40 - 08:45	Discussion		
08:45 - 08:55	2nd Talk	Anand Kumar	Understanding ice nucleation characteristics of selective mineral dusts suspended in solution
08:55 - 09:00	Discussion		
09:00 - 09:10	3rd Talk	Thomas Häusler	Cellulose and their characteristic ice nucleation activity- freezing on a chip
09:10 - 09:15	Discussion		
09:15 – 09:25	4th Talk	Anna Kunert	TINA: A new high-performance droplet freezing assay for the analysis of ice nuclei with complex composition
09:25 - 09:30	Discussion		
09:30 - 09:55	Break Out Discussion (Coffee & Cakes)		
Laboratory Measurements III			
09:55 - 10:05	1st Talk	Szakall Miklos	Immersion and contact freezing experiments utilizing contact-free droplet levitation techniques
10:05 - 10:10	Discussion		
10:10 - 10:20	2nd Talk	Thea Schiebel	INKA - An instrument for investigations of atmospheric ice nucleation processes
10:20 - 10:25	Discussion		
10:25 - 10:35	3rd Talk	Jan Frederik Scheel	Identification & characterization of fungal ice nucleation proteins
10:35 - 10:40	Discussion		
10:40 - 10:50	4th Talk	Bernhard Pummer	How cultivation conditions affect the ice nucleation activity of Sarocladium (Acremonium) implicatum
10:50 - 10:55	Discussion		
10:55 - 11:20	Coffee Break		

Computational Modelling			
11:20 - 11:30	1st Talk	Chih-Che Chueh	A numerical study on the time-varying attitudes and aerodynamics of freely falling conical Graupel particles
11:30 - 11:35	Discussion		
11:35 - 11:45	2nd Talk	Ivan Coluzza	The role of water in selecting stable proteins in extreme thermodynamic conditions
11:45 - 11:50	Discussion		
11:50 - 12:00	3rd Talk	Michael Schauperl	Explaining the recognition process of ice binding proteins
12:00 - 12:05	Discussion		
12:05 - 12:15	4th Talk	Sylvia Sullivan	Adjoint sensitivites and attributions to understand model output ice crystal number variability
12:15 - 12:25	Closing Remarks- Hinrich Grothe		
12:25 - 14:00	Lunch Buffet		
14:00	End of the Workshop		

Ice-nucleating strains of Pseudomonas syringae from a large freshwater lake in Virginia, USA

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The bacterium *Pseudomonas syringae* can be found in a variety of terrestrial and aquatic environments. Some strains of the bacterium are plant pathogens, and some express an ice nucleation protein (hereafter referred to as Ice+) allowing them to catalyze the freezing of water at warmer temperatures. Little is known about aquatic sources of *P. syringae*. We collected samples of freshwater from four different locations in Claytor Lake, Virginia, USA over five days between November 2015 and February 2016. Samples were concentrated and plated on TSA and KBC to estimate the number of culturable bacteria and *Pseudomonas* in freshwater, respectively. A droplet freezing ice nucleation assay was used to screen colonies for the Ice+ phenotype, and those determined to be Ice+ were identified to the level of species based on cts and 16s sequences. Colonies of *Pseudomonas* were recovered from all of the lake samples, and the concentration varied among samples collected from the same location on different days. Between 6% and 15% of the *Pseudomonas* colonies were Ice+. Most of the colonies cultured on KBC were identified as *P. syringae.* Preliminary phylogenetic analysis of cts sequences from strains of *P. syringae* showed a surprising diversity of phylogenetic subgroups present in the lake. Our work shows that freshwater lakes can be a significant source of Ice+ *P. syringae*. Future worked is needed to understand the mechanics of aerosolization of *P. syringae* and other microbial ice nucleators from aquatic environments.

The adsorption of fungal ice-nucleating proteins on mineral dusts: a terrestrial reservoir of atmospheric ice-nucleating particles

Daniel O'Sullivan[1], Benjamin J. Murray[1], James Ross[2,3], and Michael E. Webb[2]

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Abstract: The occurrence of ice-nucleating particles (INPs) in our atmosphere has a profound impact on the properties and lifetime of supercooled clouds. However, the identities, sources and abundances of airborne particles capable of efficiently nucleating ice at relatively low supercoolings ($T >$ -15 °C) remain enigmatic. Recently, several studies have suggested that unidentified biogenic residues in soil dusts are likely to be an important source of these efficient atmospheric INPs. While it has been shown that cell-free proteins produced by common soil-borne fungi are exceptional INPs, whether these fungi are a source of ice-nucleating biogenic residues in soils has yet to be shown. In particular, it is unclear whether upon adsorption to soil mineral particles, the activity of fungal ice-nucleating proteins is retained or is reduced, as commonly observed for other soil enzymes. Here we show that proteins from a common soil fungus (*Fusarium avenaceum*) do in fact preferentially bind to and impart their ice-nucleating properties to the common clay mineral kaolinite. The ice-nucleating activity of the proteinaceous INPs is found to be unaffected by adsorption to the clay, and once bound the proteins do not readily desorb, retaining much of their activity even after multiple washings with pure water. The atmospheric implications of the finding that nanoscale fungal INPs can effectively determine the nucleating abilities of lofted soil dusts are discussed.

Ice nucleating particles on the High Altitude Research Station Jungfraujoch: 5 years of measurements

Larissa Lacher[1], Ulrike Lohmann[1] and Zamin A. Kanji[1]

[1]*Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland*

Clouds containing ice play an important role in the Earth's system, but fundamental knowledge on their formation and further development is still missing. Only a tiny fraction of ambient aerosol particles are ice nucleating particles (INP) and continuous measurements in an environment relevant for ice and mixed-phase clouds are rare. We perform INP measurements on a regular basis at the High Altitude Research Station Jungfraujoch. The site provides free tropospheric conditions with seasonal influence from boundary layer air and is regularly affected by Sahara dust events. By measuring INP concentrations during different seasons of the year we address the question of an annual cycle of INP concentrations in the free troposphere and quantify the influence of Sahara dust events and convectively lifted air. The measurements are performed with the Horizontal Ice Nucleation Chamber (HINC) at 242K in the water- sub- and supersaturated regime, which is relevant for the formation of ice and mixed-phase clouds. Results from field campaigns in winter, spring and summer are compared and show that INP concentrations range from almost zero to several hundred per liter. Below water saturation, INP concentrations are generally an order of magnitude lower than above water saturation, and don't show a seasonal variation. At water-supersaturated conditions, seasonal occurrence of Sahara dust and boundary layer injections influence the INP concentrations and lead on average to higher values. Variations in free tropospheric conditions are mainly due to transported Sahara dust, leading to peak INP concentrations during Sahara dust events. Furthermore, a significant increase in INP concentrations was measured during an event with influence from marine air. It also appeals that when ambient temperatures at Jungfraujoch are low, INP concentrations are also lower suggesting scavenging of INPs being transported to the measurement site. With the continuation of these measurements we have a longer time series and therefore a better understanding of the evolution of INP concentrations on Jungfraujoch.

Utilizing radar data in support of the hail mitigation operations in Styria, Austria

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Graz University of Technology, Institute of Microwave and Photonic Engineering, Graz, Austria (satyanarayana.tani@tugraz.at)

Hail storm damage is a major concern to the farmers in the province of Styria, Austria. Each year severe hail storms are causing damages to crops, resulting in losses of millions of euros. Hail mitigation operations are carried out in the province of Styria, to reduce the risk of hail damage. Radar serve for the identification of hail clouds and the aircraft's injected the seeding material into the clouds. Radar data offer high spatial (1 km x 1 km x 1 km) and temporal resolutions (5 minutes), resulting in very promising option for hail mitigation operations and evaluation. In order to provide the guidelines to the pilots, here we offer a portable radar PC, which integrates the air craft's location, cloud movement and radar reflectivity data. For the evaluation purpose, the HAILSYS software tool was developed by integrating single polarization C-band weather radar data, aircraft trajectory, radiosonde freezing level data, hail events and crop damage information from the ground. The radar data have been examined, the evaluation of hail mitigation operation will be presented with the help of some case studies.

Ice nucleating particle properties in the Saharan air layer

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Mineral dust is amongst the most common ice nucleating particle (INP) types in the atmosphere. Nevertheless, no field study exists to date investigating the ice nucleation properties of atmospheric desert dust close to its globally largest emission source, the Sahara. The presented study fills this gap.

In August 2013 and 2014 measurements of INP concentrations, aerosol particle size distributions, bulk and single particle chemistry and fluorescence were conducted at the Izaña Observatory located at 2373m above sea level on Tenerife, west of the African shore. During summer, the observatory is frequently exposed to high dust loads which are transported within the Saharan Air Layer. INP concentrations at temperatures between -40 and -15 °C and RH_i = 100 to 150 % were measured. We investigated how changes in the chemical composition and biological material affect the INP properties of desert dust. Airborne dust samples were collected with a cyclone for additional offline analysis in the laboratory under similar conditions as in the field.

Results from the field measurements show that the background aerosol at Izaña was dominated by carbonaceous particles, which were hardly ice-active under the investigated conditions. When Saharan dust was present, INP concentrations increased by up to two orders of magnitude. Long-range transported biomass burning aerosol was not ice-active under the investigated conditions. The autofluorescence analysis of the ice crystal residuals showed that up to 25 % of the INPs contained biological material during Saharan dust events, compared to only 5 % of the ambient particles. Furthermore, condensation mode INP concentrations showed a positive correlation with the ammonium sulfate to dust mass ratio below a certain threshold value. This indicates that atmospheric aging processes involving ammonium can increase the ice nucleation ability of mineral dust.

The relationships between insoluble precipitation residues, clouds, and precipitation over California's Sierra Nevada during winter storms

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Ice formation in orographic mixed-phase clouds can enhance precipitation and depends on the type of aerosols that serve as ice nucleating particles (INPs). The resulting precipitation from these clouds is a viable source of water, especially for regions such as the California Sierra Nevada. Thus, a better understanding of the sources of INPs that impact orographic clouds is important for assessing water availability in California. This study presents a multi-site, multi-year (2009 – 2012) analysis of single-particle insoluble residues in precipitation samples that likely influenced cloud ice and precipitation formation. Dust and biological particles represented the dominant fraction of the residues. Cloud glaciation, determined using GOES satellite observations, not only depended on high cloud tops and low temperatures, but also on the composition of the dust and biological residues. The greatest prevalence of ice-phase clouds and INP concentrations occurred in conjunction with biologically-rich residues and mineral dust rich in calcium, followed by iron and aluminosilicates. We show that the dust and biological particles served as efficient INPs, thus these residues are what likely influenced ice formation in clouds above the sites and subsequent precipitation quantities reaching the surface during events with similar meteorology. The goal of this study is to use precipitation chemistry and immersion INP information to gain a better understanding of the potential sources of INPs in the Sierra Nevada, where cloud-aerosol-precipitation interactions are poorly understood and where mixed-phase orographic clouds represent a key element in the generation of precipitation and thus the water supply in California.

Measuring the effect of pollen grains and their ice active macro-molecules by observations of the INP concentration during the birch pollen season

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Pollen are amongst the largest aerosol particles found in the atmosphere but are found in low concentrations in comparison to other aerosol species. Amongst other bioaerosol particles such as fungal spores and bacteria, pollen can nucleate ice at small supercooling and are therefore of interest for atmospheric ice formation, despite their low concentrations.

Birch pollen can induce freezing at temperatures as warm as 264 K in the immersion mode [1]. Laboratory measurements have shown that ice active macro-molecules (INMs) can be easily washed off from pollen grains and can be released in high numbers from a single grain [2,3]. The release of INMs directly into the atmosphere may contribute significantly to ice formation processes as they can be distributed well in the atmosphere without pollen grains as a carrier due to their small size.

In this study, we present immersion freezing ice nucleating particle (INP) concentrations from two drop-freezing techniques which were conducted during the birch pollen season in Zurich. Due to differences in the aerosol particle sizes sampled between the two methods, our measurements allow for deduction of the size of the aerosol particles that contributed to ice nucleation. Measurements of meteorological conditions, PM10 mass and physical aerosol properties further support our discussion. Over the course of 6 weeks, INP concentrations were observed in a temperature range between 256-269 K. We observed that particles larger than 10 μm may have contributed to ice nucleation at the peak of the pollen season, which indicates pollen grains (typically much larger than 10 μm) acted as potential INPs. The presence of INMs in the atmosphere was tested by measuring pollen and INP concentrations in parallel. Due to a release of large numbers of INMs from a single pollen grain, a substantial increase in INP concentrations was expected. From measured INP concentrations in samples only containing aerosol particle sizes smaller than 10 μm, we did not find indications that INMs were present in the atmosphere during the pollen season or else, contributed significantly to the INP concentration measured. Instead, environmental conditions such as relative humidity were found to effect the INP concentration for some periods investigated and are discussed.

References:

[1] K. Diehl *et al.* (2002) *Atmos. Res.*, **61**, 125-133.

[2] S. Augustin *et al.* (2012) *Atmos. Chem. Phys. Discuss.*, **12**, 32911-32943.

[3] B. G. Pummer *et al.* (2012) *Atmos. Chem. Phys*., **12**, 2541-2550.

An unmanned surface vehicle (USV) to study the air/water interface in aquatic environments

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The air/water interface (AWI) is a biologically rich and active ecological zone in aquatic environments. Little is known about the mechanics of the AWI, and how the AWI contributes aerosols that could impact weather. We developed and implemented an unmanned surface vehicle (USV) to study the AWI. In one set of experiments on a small pond in Blacksburg, VA USA, the USV was equipped with a distributed temperature sensing (DTS) system to measure temperature profiles from about 1 m below the surface of the water to about 1 m above the surface. This integrated USV DTS system resolved a temperature differential of about 6C at the AWI spanning a distance of about 8 cm with a vertical resolution of 1 mm. In other experiments, the USV was equipped with impingers and real-time particle counters positioned at multiple heights above the water surface. A field campaign was conducted over a large freshwater lake in Baton Rouge, Louisiana, USA in October, 2015 to coordinate sampling of microbial ice nucleators in and near the water with the USV, and 60 m above the water with a small drone. The USV is currently being using to resolve aerosolization phenomena at the AWI in natural aquatic systems that have the potential to impact weather. Such technology is poised to unleash a burgeoning field of aero-aquatic-ecology.

Exploring the sources of variability in ice nucleating particles of biological origin

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

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Ice nucleation is a key step for the formation of precipitation on Earth. Ice nucleating particles (INPs) of biological origin catalyse the freezing of supercooled cloud droplets at temperatures warmer than -12 °C. In order to understand the effective role of these INPs in conditioning precipitation, it is of primary importance to describe and predict their variability in the atmosphere.

Variability in time- Between 2012 and 2014, 14 sampling campaigns in precipitating clouds were conducted at the High Altitude Research Station Jungfraujoch, in the Swiss Alps, at 3580 m a.s.l. A total of 106 freshly fallen snow samples were analysed immediately on site for the concentration of INPs active at -8 °C (INPs-8) by immersion freezing. Values of INPs-8 ranged from 0.21 to 434·ml-1. Multiple linear regression models were built to describe and predict the variation in the abundance of INPs-8. These models indicate that a coincidence of strong atmospheric turbulence and little prior precipitation from a cloud coincides with large concentrations of INPs-8, a set of conditions which can be easily met for instance along the passage of a front. To obtain more information on the presence of INPs-8 of biological origin a subset of precipitation samples was progressively filtered and heated. The abundance of bacterial cells and the presence of culturable Pseudomonas syringae were studied as well. Whilst each sample presents a specific distribution of the sizes of INPs-8, almost all ice nucleating activity is lost after heating at 80 °C, and a significant part of INPs-8 is sensitive to warming at 40 °C. Just a minor fraction of the INPs-8 is potentially due to intact bacterial cells or culturable P. syringae, indicating that the majority of INPs-8 measured in environmental samples is made of heat-sensitive molecules of biological origin which can either be singularly airborne or aggregated on soil and mineral particles.Variability in space- To start studying the variability of INPs-8 over a spatial scale and testing the results obtained at Jungfraujoch, in October 2015 the project NICE (Nucleators of Ice at monte CimonE) was carried out, in the frame of the European Union funding for the Project ACTRIS. Airborne particles were collected on quartz fibre PM10 filters over a week at Monte Cimone (IT, 2165 m a.s.l.) and the concentrations of INPs-8·m-3 calculated. These results are currently being compared with the analyses of the PM10 filters collected for the same week at Jungfraujoch and Puy de Dôme (FR, 1465 m a.s.l.). Results on this spatial variability of airborne INPs-8 will be presented and constitute a first attempt to set up a network of observatories where measurements on INPs-8 can be carried out regularly.

Ice nucleating particles measured during the field intercomparison FIN-3 by the vacuum diffusion chamber FRIDGE

Daniel Weber, Jann Schrod, Heinz Bingemer & Joachim Curtius

Institut für Atmosphäre und Umwelt Fachbereich Geowissenschaften/Geographie, Johann Wolfgang Goethe-Universität, Frankfurt/Main

The Frankfurt Ice Nucleation Deposition freezing Experiment (FRIDGE) is an offline INP-counter, which first collects aerosol particles via electrostatic precipitation on silicon-wafers and exposes them to typical cloud conditions in a vacuum diffusion chamber afterwards to detect nucleation and growth of ice on INP.

FRIDGE was a part of Fin-3, a field campaign at the Desert Research Institutes Storm Peak Laboratory in Colorado, USA in September 2015. Storm Peak Laboratory (3210 m asl) is within the boundary layer in the daytime and within the free troposphere at night. For 44 aerosol samples INP-concentrations at different temperatures and humidity were determined. We observed different INP-concentrations in both atmospheric layers, as well as a high day-to-day variability. Furthermore, the effect of previous snowfall was studied.

Modification of mineral dust during long-range transport and associated impact on predicted ice nuclei concentration - insights from SALTRACE

Weinzierl, B. (1, 2), D. Sauer (2), A. Walser (3,2), M. Dollner (1), F. Chouza (2), O. Reitebuch (2), S. Groß (2), V. Freudenthaler (3), K. Kandler (4), A. Ansmann (5)

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Every year huge amounts of mineral dust are transported from Africa over Atlantic Ocean into the Caribbean affecting air quality, health, weather and climate thousands of kilometers downwind of the dust source regions. Despite this importance, there has been little research on the long-range transport of mineral dust and many questions such as the change of the dust size distribution during transport, the role of wet and dry removal mechanisms, and the complex interaction between mineral dust and clouds are still open.

To investigate the long-range transport of mineral dust from African into the Caribbean, the Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE: http://www.pa.op.dlr.de/saltrace) was conducted in June/July 2013. SALTRACE was designed as a closure experiment combining ground-based lidar, in-situ and sun photometer instruments deployed on Cape Verde, Barbados and Puerto Rico, with airborne aerosol and wind lidar measurements of the DLR research aircraft Falcon, satellite observations and model simulations.

The DLR research aircraft Falcon spent more than 110 flight hours studying mineral dust from several dust outbreaks under a variety of atmospheric conditions between Senegal, Cape Verde, the Caribbean, and Florida. During SALTRACE, a comprehensive data set on chemical, microphysical and optical properties of aged mineral dust was gathered. Although changes in the size distribution were observed during transport, one interesting finding during SALTRACE was the detection of large 10-20 µm particles in the Caribbean even after more than 4000 km of transport.

Here we use data from airborne in-situ aerosol measurements with the DLR Falcon research aircraft during the Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE: http://www.pa.op.dlr.de/saltrace) to study the modification of the mineral dust layers between Africa and the Caribbean. We will present vertical profiles of dust properties including cloud condensation nuclei number concentration. In addition, we will use a parameterization predict the number concentration of potential ice nuclei in the presence of mineral dust.

A laboratory study of the nucleation kinetics of nitric acid hydrates under stratospheric conditions

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Measurements of the kinetics of crystallisation of ternary H_2O-H_2SO_4-HNO_3 mixtures to produce nitric acid hydrate phases, as occurs in the lower stratosphere, have been a long-standing challenge for investigators in the laboratory. Understanding polar stratospheric chlorine chemistry and thereby ozone depletion is increasingly limited by descriptions of nucleation processes. Meteoric smoke particles have been considered in the past as heterogeneous nuclei, however recent studies suggest that these particles will largely dissolve, leaving mainly silica and alumina as solid inclusions.

In this study the nucleation kinetics of nitric acid hydrate phases have been measured in microliter droplets at polar stratospheric cloud (PSC) temperatures, using a droplet freezing assay. A clear heterogeneous effect was observed when silica particles were added. A parameterisation based on the number of droplets activated per nuclei surface area (n_s) has been developed and compared to global model data.

Nucleation experiments on identical droplets have been performed in an X-Ray Diffractometer (XRD) to determine the nature of the phase which formed. β-Nitric Acid Trihydrate (NAT) was observed alongside a mixture of Nitric Acid Dihydrate (NAD) phases. It is not possible to determine whether NAT nucleates directly or is formed by a phase transition from NAD (likely requiring the presence of a mediating liquid phase). Regardless, these results demonstrate the possibility of forming NAT on laboratory timescales. In the polar stratosphere, sulfuric acid (present at several weight percent of the liquid under equilibrium conditions) could provide such a liquid phase.

This study therefor provides insight into previous discrepancies between phases formed in the laboratory and those observed in the atmosphere. It also provides a basis for future studies into atmospheric nucleation of solid PSCs.

Metastable Nitric Acid Trihydrate in Ice Clouds

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The composition of high altitude ice clouds is still a matter of intense discussion. The constituents in question are ice and nitric acid hydrates. The identification and formation mechanisms, however, are still unknown but are essential to understand atmospheric processing such as the seasonal ozone depletion in the lower polar stratosphere or the radiation balance of planet Earth. We found conclusive evidence for a long-predicted phase, which has been named alpha nitric acid trihydrate (alpha-NAT). This phase has been characterized by a combination of X-ray and neutron diffraction experiments allowing a convincing structure solution. Additionally, vibrational spectra (infrared and inelastic neutron scattering) were recorded and compared with theoretical calculations. A strong affinity between water ice and alpha-NAT has been found, which explains the experimental spectra and the phase transition kinetics essential for identification in the atmosphere. On the basis of our results, we propose a new three-step mechanism for NAT-formation in high altitude ice clouds.

Analysis of isothermal and cooling-rate-dependent immersion freezing by a unifying stochastic ice nucleation model

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Immersion freezing is an important ice nucleation pathway involved in the formation of cirrus and mixed-phase clouds. Since ice nucleation cannot be directly observed in the atmosphere, laboratory immersion freezing experiments are necessary to determine the range in temperature and relative humidity at which ice nucleation occurs. In some experimental studies, it is assumed that droplets containing ice nucleating particles (INPs) all have the same INP surface area (ISA), but is this assumption valid or does it affect experimental analysis and interpretation of the immersion freezing process? Descriptions of ice active sites and variability of contact angles have been successfully formulated to describe ice nucleation experimental data in previous research; however, we consider the ability of a stochastic freezing model founded on classical nucleation theory to reproduce previous results and to explain experimental uncertainties and data scatter considering variable ISA. Experimental results derived from a variety of approaches are reproduced by our model including droplets on a cold-stage exposed to air or surrounded by an oil matrix, wind and acoustically levitated droplets, droplets in a continuous-flow diffusion chamber (CFDC), the Leipzig aerosol cloud interaction simulator (LACIS) and the aerosol interaction and dynamics in the atmosphere (AIDA) cloud chamber. Observed time-dependent isothermal frozen fractions exhibiting non-exponential behavior can be readily explained by this model if varying ISA are used. An apparent cooling-rate dependence of the heterogeneous ice nucleation rate coefficient, J_{het}, can be explained by assuming identical ISA in each droplet. When accounting for ISA variability, the cooling-rate dependence of J_{het} vanishes as expected from classical nucleation theory. A quantitative uncertainty analysis due to measureable parameters, such as the number of droplets used and variable ISA, will be discussed with implications for future experimental design and analysis.

Air-plane soot surrogates and their ice nucleation activity: investigations in deposition mode

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Emissions of solid-state particles (soot) from engine exhausts due to incomplete fuel combustion is considered to influence ice and liquid water cloud droplet activation [1]. The activity of these aerosols would originate from their ability to promote ice formation above pure water homogeneous freezing point (~235 K). When considering deposition mode, i.e., when an ice embryo forms directly by water vapor condensation on a surface, it has been suggested that no liquid water is involved in the process because the relative humidity at which ice nucleation is generally observed is below the water saturation line [2]. Further, the ice nuclei (aerosols) are then considered to have active sites that promote heterogeneous ice nucleation at relative humidity below the homogeneous ice nucleation line (i.e. at RHi (relative to ice) below ~ 140-150%). There is no clear evidence of heterogeneous ice nucleation on soot aerosols for T > 235 K, and soot particles are reported to be generally worse ice nuclei than mineral dust because they activate nucleation at higher ice-supersaturations for deposition nucleation or at lower temperatures for immersion freezing than what is usually expected for homogeneous nucleation [2]. In fact, there are still numerous opened questions as to whether and how soot surfaces' physico-chemical properties (structure, morphology and chemical composition) can influence their nucleation ability.

Therefore, systematic investigations of soot aerosol nucleation activity via one specific nucleation mode, here deposition nucleation, combined with thorough structural and compositional analyzes are needed in order to establish any association between the particles' activity and their physico-chemical properties. In addition, since the morphology of the ice crystals can influence their radiative properties [3], it is of paramount importance to investigate their morphology as they grow over both soot or pristine substrates at different temperatures and humidity ratios. In the present work, a CAST (Combustion Aerosol STandart) burner supplied with propane is used to produce soot samples using various experimental conditions. The nucleation activity is studied in deposition mode and is monitored using a temperature-controlled reactor in which the sample's relative humidity is precisely measured via a cryo-hygrometer. Formation of water/ice onto the particles is followed both optically and spectroscopically, using a microscope coupled to a Raman spectrometer. Vibrational signatures of hydroxyls (O-H) emerge when the particle becomes hydrated and are used to characterize ice crystals.

Acknowledgement: F.X. Ouf (IRSN) for providing soot samples

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Understanding Ice Nucleation by Alkali Feldspars

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The formation of ice in the atmosphere can affect cloud properties and have an impact on the Earth's radiative balance. In mixed-phase clouds, freezing of super-cooled water typically occurs by heterogeneous ice nucleation. The ice nucleating particles (INP) that cause this come from a variety of sources, including atmospheric mineral dust. Amongst the minerals that form these dusts, alkali feldspars have been identified as being important INP. However, whilst feldspars are known to nucleate ice well, the mechanism by which this occurs is not understood.

Using a range of analytical techniques, we have studied immersion mode ice nucleation for several alkali feldspars. By using thin sections, which are 30 μm thick polished sheets of natural feldspar with several centimetre diameters, we were able to characterise in detail the surfaces on which nucleation occurs. From this work, the microtexture of the feldspars studied has been seen to be an important factor in ice nucleating ability. This microtexture comprises exsolution lamellae, crystal twinning, grain boundaries and micropores. In the absence of microtexture, the ice nucleating ability of feldspars is substantially diminished. For samples with microtexture, a preference for nucleation on particular regions of the feldspar surface is seen. There are several types of active site on a feldspar surface, each with different activation temperatures. We conclude that the most active sites on the feldspar are rare, and that the microtexture of the feldspars is important for the ice nucleation observed at higher temperatures. This is important because it seems that ice nucleation by alkali feldspars is controlled by active sites which are related to specific features on a surface. This paves the way for understanding how these sites might be influenced by atmospheric processes, such as the action of acids, which are known to reduce the activity of feldspars.

Understanding ice nucleation characteristics of selective mineral dusts suspended in solution

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Introduction & Objectives

Freezing of liquid droplets and subsequent ice crystal growth affects optical properties of clouds and precipitation. Field measurements show that ice formation in cumulus and stratiform clouds begins at temperatures much warmer than those associated with homogeneous ice nucleation in pure water, which is ascribed to heterogeneous ice nucleation occurring on the foreign surfaces of ice nuclei (IN). Various insoluble particles such as mineral dust, soot, metallic particles, volcanic ash, or primary biological particles have been suggested as IN. Among these the suitability of mineral dusts is best established. The ice nucleation ability of mineral dust particles may be modified when secondary organic or inorganic substances are accumulating on the dust during atmospheric transport. If the coating is completely wetting the mineral dust particles, heterogeneous ice nucleation occurs in immersion mode also below 100 % RH.

A previous study by Kaufmann (PhD Thesis 2015, ETHZ) with Hoggar Mountain dust suspensions in various solutes (ammonium sulfate, PEG, malonic acid and glucose) showed reduced ice nucleation efficiency (in immersion mode) of the particles. Though it is still quite unclear of how surface modifications and coatings influence the ice nucleation activity of the components present in natural dust samples. In view of these results we run freezing experiments using a differential scanning calorimeter (DSC) with the following mineral dust particles suspended in pure water and ammonium sulfate solutions: Arizona Test Dust (ATD), microcline, and kaolinite (KGa-2, Clay Mineral Society).

Methodology

Suspensions of mineral dust samples (ATD: 2 weight%, microcline: 5% weight, KGa-2: 5% weight) are prepared in pure water with varying solute concentrations (ammonium sulfate: 0 – 10% weight). 20 vol% of this suspension plus 80 vol% of a mixture of 95 wt% mineral oil (Aldrich Chemical) and 5 wt% lanolin (Fluka Chemical) is emulsified with a rotor-stator homogenizer for 40 s at a rotation frequency of 7000 rpm. 4 – 10 mg of this mixture is pipetted in an aluminum pan (closed hermetically), placed in the DSC and subjected to three freezing cycles. The first and the third freezing cycles are executed at a cooling rate of 10 K/min to control the stability of the sample. The second freezing cycle is executed at a 1 K/min cooling rate and is used for evaluation. Freezing temperatures are obtained by evaluating the onset of the freezing signal in the DSC curve and plotted against water activity values corresponding to the solute concentration (obtained via Koop et al., (2000)).

Observations

A decrease in ice nucleation ability of the minerals (for immersion freezing) with increasing solute concentration (hence, decreasing water activity) was observed, similar as for homogeneous ice nucleation. Though the decrease was more pronounced in case of microcline and ATD as compared to kaolinite. Therefore, there seem to be specific interactions which needs to be studied further to explain the freezing behavior of minerals.

The current study could be helpful in investigating the ice nucleation behavior of individual minerals when present in conjunction with a solute, viz. ammonium sulfate, which is of high atmospheric relevance.

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Cellulose and Their Characteristic Ice Nucleation Activity- Freezing on a Chip

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The influence of clouds on the Earth's climate system is well known (IPCC, 2013). Cloud microphysics determines for example cloud lifetime and precipitation properties. Clouds are cooling the climate system by reflecting incoming solar radiation and warm its surface by trapping outgoing infrared radiation (Baker and Peter, 2008). In all these processes, aerosol particles play a crucial role by acting as cloud condensation nuclei (CCN) for liquid droplets and as an ice nucleation particle (INP) for the formation of ice particles.

Freezing processes at higher temperatures than -38°C occur heterogeneously (Pruppacher and Klett 1997). Therefore aerosol particles act like a catalyst, which reduces the energy barrier for nucleation. The nucleation mechanisms, especially the theory of functional sites are not entirely understood. It remains unclear which class of compound nucleates ice.

Here we present a unique technique to perform drop- freezing experiments in a more efficient way. A self-made freezing- chip will be presented. Measurements done to proof the efficiency of our setup as well as advantages compared with other setups will be discussed.

Furthermore we present a proxy for biological INPs, microcrystalline cellulose. Cellulose is the main component of herbal cell walls (about 50 wt%). It is a polysaccharide consisting of a linear chain of several hundred to many thousands of β(1→4) linked D-glucose units. Cellulose can contribute to the diverse spectrum of ice nucleation particles. We present results of the nucleation activity measurements of MCCs as well as the influence of concentration, preparation or chemical modification.

TINA: A New High-Performance Droplet Freezing Assay for the Analysis of Ice Nuclei with Complex Composition

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Freezing of water above homogeneous freezing is catalysed by ice nucleation active (INA) particles called ice nuclei (IN), which can be of various inorganic or biological origin. The freezing temperatures reach up to -1 °C for some biological samples and are dependent on the chemical composition of the IN. The standard method to analyse IN in solution is the droplet freezing assay (DFA) established by Gabor Vali in 1970. Several modifications and improvements were already made within the last decades, but they are still limited by either small droplet numbers, large droplet volumes or inadequate separation of the single droplets resulting in mutual interferences and therefore improper measurements.

The probability that miscellaneous IN are concentrated together in one droplet increases with the volume of the droplet, which can be described by the Poisson distribution. At a given concentration, the partition of a droplet into several smaller droplets leads to finely dispersed IN resulting in better statistics and therefore in a better resolution of the nucleation spectrum.

We designed a new Twin Ice Nucleation Assay (TINA), which represents an upgrade of the previously existing DFAs in terms of temperature range and statistics. The necessity of observing freezing events at temperatures lower than homogeneous freezing due to freezing point depression requires high-performance thermostats combined with an optimal insulation. Furthermore, we developed a cooling setup, which allows both huge and tiny temperature changes within a very short period of time. Besides that, TINA provides the analysis of more than 750 droplets per run with a small droplet volume of 5 µL. This enables a fast and more precise analysis of biological samples with complex IN composition as well as better statistics for every sample at the same time.

Immersion and contact freezing experiments utilizing contact-free droplet levitation techniques

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Experimental studies have been carried out in the Mainz vertical wind tunnel laboratory in order to characterize immersion and contact freezing for different ice nucleating particles (INP), such as for mineral dust or biological particles. Immersion freezing is investigated in our laboratory with two different experimental techniques, both attaining contact-free levitation of liquid droplets and cooling of the surrounding air down to about -25 °C. In an acoustic levitator placed in the cold room of our laboratory, drops with diameters of 2 mm are investigated. In the vertical air stream of the wind tunnel droplets with diameter of 700 micron are freely floated at their terminal velocities, simulating the flow conditions of the free atmosphere. Furthermore, the wind tunnel offers a world-wide unique platform for contact freezing experiments. Supercooled water droplets are floated in the vertical air stream at their terminal velocities and INP are injected into the tunnel air stream upstream of them. As soon as INP collides with the supercooled droplet the contact freezing is initiated.

Results of immersion freezing experiments with typical mineral dust (e.g., illite, feldspar) and biological particles (e.g., bacteria, cellulose) both in the acoustic levitator and in the wind tunnel will be presented. The study includes direct comparison between the different experimental techniques (acoustic levitator and wind tunnel), as well as between nucleation modes (immersion and contact freezing).

INKA – An Instrument for Investigations of Atmospheric Ice Nucleation Processes

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To obtain quantitative information on the ice nucleation activity of various atmospheric aerosol species, we have built a new continuous flow diffusion chamber (CFDC) called INKA (Ice Nucleation Instrument of the KArlsruhe Institute of Technology) which allows us to conduct fast relative humidity scans at a constant aerosol temperature. The CFDC design was originally developed and theoretically described by Rogers et al. (1988). Its main part - the chamber - consists of two vertically-aligned, concentric tubes with a total length of 150 cm. Together with particle-free, dry sheath air, the sampled aerosol particles flow through the annular space between these two cylinders. Both walls of this annular gap are coated with ice and can be controlled by a preset, linear temperature ramp. This enables us to continuously increase the temperature difference between the two walls in order to expose the aerosol particles in the sample flow to a continuously increasing saturation ratio while keeping the aerosol temperature constant. Since the wall temperatures can be reduced to values as low as -70 °C, ice nucleation and growth of relevance for both mixed-phase and cirrus clouds can be investigated under well controlled temperature and humidity conditions. In this contribution, we will present the setup of INKA and will show first measurements in comparison with literature data.

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Hunting the Ice Nucleation Protein from *Fusarium*

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Freezing of water at temperatures between homogeneous freezing and the melting point can only occur when catalyzed by ice nuclei (IN). Water cycling in our Earth´s atmosphere is highly dependent on mixed phase clouds and cloud formation and ice multiplication processes are thought to be dependent on temperatures between -2°C - -10°C. The most efficient IN catalyzing phase transition from liquid to solid within these temperatures are biological particles. *Fusarium* was already found to be IN active since 25 years and more important *Fusarium* is one of the most abundant soil fungi. Former studies analyzing *Fusarium* and other fungal IN indicated the fungal IN to be a protein or at least contain a proteinaceous compound. Although extensive efforts were made the precise knowledge of the nanometer scale IN from *Fusarium* and the corresponding genetic elements remain elusive.

In this project we plan to purify the cell free IN from fungal strains of the genus *Fusarium* to analyze their properties. With analytical methods we want to identify the primary structure of the protein or proteinaceous compounds involved in forming fungal IN. Fractionation by size exclusion chromatography showed the IN of *Fusarium* to be eluted in early fractions. This indicates the fungal IN to be of huge size and agrees with filtration experiments showing the fungal IN of *Fusarium* as well as other fungal IN have a molecular weight above 100kDa. With ionic exchange chromatography we were able to detect the IN by anion exchange and protein was always found in IN active fractions by SDS-Page. However, it was not enough material purified to get sufficient primary structure information by mass spec to date.

How cultivation conditions affect the ice nucleation activity of *Sarocladium* (*Acremonium*) *implicatum*

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The fungus *Sarocladium* (formerly: *Acremonium*) *implicatum* is capable of producing ice-nucleating proteins that are active at temperatures above 260 K (Pummer et al., 2015). Literature data show that cultivation conditions strongly influence the protein expression in microorganisms, which also lead to change or loss of their ice nucleation activity (e.g. Nemecek-Marshall et al., 1993, Tsumuki et al., 1995).

The connection between the ice nucleation activity of *Sarocladium implicatum* and the cultivation parameters, like environmental conditions (temperature, humidity, light), available nutrients, and culture age are revealed and interpreted.

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A NUMERICAL STUDY ON THE TIME-VARYING ATTITUDES AND AERODYNAMICS OF FREELY FALLING CONICAL GRAUPEL PARTICLES

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The flow fields and dynamic motions of conical graupel of diameters 0.5-5mm falling in air of 800 hPa and -20˚C are studied by solving the transient Navier-Stokes equations numerically for flow past the conical graupel and the body dynamics equations representing the 6-degrees-of-freedom motion that determines the position and orientation of the graupel in response to the hydrodynamic force of the flow fields. The shape of conical graupel made through a simple but practical existing mathematical equation allows us to have an uneven mass distribution, which is generally believed to have great influence on ice particles' orientations while falling when inertial force becomes increasingly dominant over other effects. The simulated motions include vertical fall, lateral translation, sailing, rotation, pendulum swing. The computed flow fields are characterized in terms of streamtrace patterns as well as the vorticity magnitude fields, and the corresponding motions of the conical graupel is physically featured by looking upon the graupel surface distributions of pressure coefficient, torques contributed by both pressure as well as viscous effects. Tumbling doesn't occur when an initial orientation of the graupel is either 20˚ or 160˚ about Y axis, and the torque contributed by the pressure effect is dominant over that contributed by the viscous effect. In addition, the case of the initial inclination angle equal to 20˚ shows an amplifying oscillation, whereas the same case except the different initial inclination angle set to 160˚ seems to indicate a damping oscillation. And some physically attractive animations obtained through our numerical results will be presented to prove that the numerical results can match experimental ones performed by other researchers before, and likely to further shed light on the formation of the freely falling ice particles influenced by their surrounding flow fields.

The role of water in selecting stable proteins in extreme thermodynamic conditions

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Natural evolution selects proteins that are functional under the environmental conditions. Here we study by simulations how water contributes to this selection process. We follow the idea of the protein design that artificially optimizes amino acid sequences aiming at a folded state under specific thermodynamic conditions. First, we introduce a novel selection protocol for sequence–optimization, efficient also at extreme thermodynamic conditions, that reproduces experimental data. Next, we show that sequences optimized at high temperature are remarkably stable in a temperature-pressure range broader than the original optimization range. We find that these super-stable proteins fold also at very low temperatures, as well as those proteins that have been optimized for low temperature. We observe that proteins with a high separation between hydrophilic surface and hydrophobic core, as those selected by the natural evolution, are super-stable. However, we discover that sequences with an extreme hydrophobic/hydrophilic separation have a stability comparable to the super-stable proteins. Hence, there is no evolution advantage in selecting proteins with perfect hydrophilic surfaces and hydrophobic cores. Our results elucidate the evolutionary pressure exercised by water on the hydropathy profile as function of the thermodynamic state point, with implications for the understanding of natural and artificial selection of proteins at extreme conditions.

Explaining the recognition process of ice binding proteins

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Pure water freezes at -37°C, because the formation of ice is kinetically hindered at higher temperatures.[1] The common known property of water, freezing at 0°C results only from impurities in the water acting as ice nuclei. Ice nuclei are substances catalysing the formation of ice. Experimental studies showed, that one of the most active class of known ice nuclei are proteins.[2] These proteins are called ice nucleation proteins and catalyse the freezing already at low minus degrees (0 to -5°C).

This phenomenon is especially important, when water is found in small droplets, as it is in the artificial snow production and the formation of clouds in the atmosphere, therefore having an huge impact on the earth climate.[3]

The mechanism of ice nucleation itself is understood poorly in comparison to its relevance. In our studies we used molecular dynamics simulation to understand the underlying principle of ice nucleation. The focus in this study was to describe the hydration properties of water in the surrounding of the protein. It was possible to show that for ice nucleation the enthalpic interaction between the water and the protein has to be low. In comparison to the entropy of the surrounding water molecules, which has to be medium or high. Based on these results, we came up with a new improved explanation for the ice nucleation process of ice nucleation proteins.

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Adjoint sensitivities and attributions to understand model output ice crystal number variability

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Understanding the cause of temporal and spatial variability of the nucleated ice crystal number, N_i , will help to explain sources of model diversity in cloud forcing and to provide robust comparisons with data. To this end, we have constructed a computationally efficient adjoint model of an ice nucleation parameterization (Barahona and Nenes, 2009) and implemented it within two global climate models and with four heterogeneous nucleation spectra. The output adjoint sensitivities of N_i to input variables like aerosol number and updraft velocity provide valuable information about ice nucleation regime and ice nucleating particle efficiencies for a given model formulation. We show these sensitivities and their differences between the NCAR Community Atmosphere Model, Version 5 and the NASA Goddard Earth Observing System Model, Version 5 and between a theoretically derived spectrum, an empirical lab-based spectrum and two field-based empirical spectra. Thereafter we define two attribution metrics and show that updraft velocity fluctuations can explain as much as 89% of the temporal variability in N_i. The importance of the updraft parameterization is robust over a range of output resolutions, time steps, and altitudes but increases when the model contains a turbulence parameterization.

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