Annual Report (Part 1: Scientific Report)

Type of grant:	**Virtual Institutes**
Project No.:	VH-VI-233
Project title:	Role of aerosol particles as condensation and ice nuclei in tropospheric clouds (Aerosol-Cloud Interactions ACI)
Leading Scientists:	Prof. Thomas Leisner, Dr. Ottmar Möhler
Reporting period:	January 2009 to December 2009

Scientific Report

a) Work progress according to work programme

During the year 2009, the Helmholtz Virtual Institute on Aerosol-Cloud Interactions (VI-ACI) achieved good progress as outlined in the work programme. The activities in the different work packages are briefly summarised below.

WP-L1: AIDA
During October 2009, a third AIDA campaign on heterogneous ice nucleation, ACI-03, was cunducted within the framework of the Virtual Institute VI-ACI. Major objectives of the experiments were to investigate the affect of coating with organic and inorganic substances on the cloud condensation nucleation (CCN) and ice nucleation (IN) properties on natural desert dust mineral particles. Eight VI-ACI partner groups were involved in theses measurements. The sensitivity and performance of serveral mobile ice nuclei counters in detecting low number concentrations of ice nuclei in ambient air were investigated in comparison to the AIDA chamber. For this purpose, additional partner groups from the Colorado State University in Fort Colins, Colorado, and the University of Manchester participated. The advancement of the TDLAS (tunable diode laser absorption spectroscopy) instruments at AIDA was continued in 2009. Furthermore, a new tunable diode laser (TDLAS) instrument (SP-APicT) was introduced, which allows accurate measurements at high water vapor concentrations in mixed-phase clouds. First experiments with the goal to determine the accommodation coefficient of water molecules on ice were carried out.

WP-L2: LACIS
During the reporting period, continuous heterogeneous ice nucleation measurements were carried out at LACIS. Additionally, an intensive measurement campaign (FROST II) was performed in March / April 2009. During this campaign, the immersion freezing of surface modified Arizona Test Dust (ATD) particles was investigated. Besides characterizing the considered particles with respect to their chemical composition by means of an aerosol mass spectrometer (Uni-Mainz) and an ATOF-MS (ETH-Zürich), hygroscopic growth and activation behaviour were investigated utilizing a H-TDMA (ICG-II) and a CCNc, respectively. Furthermore, ice fractions were determined as function of temperature utilizing three different IN-counters, namely the CFDC (CSU), PINC (ETH-Zürich), FINCH (University of Frankfurt), and LACIS. Evaluation of the collected data is in progress with several publications in proparation. Results indicate that ice formation maybe strongly influenced by surface modification.

WP-L3: ZINC
The IMCA-ZINC chamber was successfully used to study immersion freezing of size selected Kaolinite particles. In this chamber(IMCA), particles activate as CCN at warm temperatures and are then cooled down to the freezing temperature in the ZINC chamber. Freezing of

droplets is monitored with the depolarization detector IODE which measures a frozen fraction of all hydrometeors in the experiment. The data shows a clear size dependency for immersion freezing of Kaolinite where larger particles freeze at higher temperatures. The data was fitted with theoretical models which assume different probability distributions of nucleation active sites on the particles. This work has been submitted to JGR and was accepted recently (publication is expected soon). Further experiments with other mineral dusts are ongoing.

WP-L4: SAPHIR

During 2009, the ICG-2 of Forschungszentrum Jülich developed a method to generate $Ca(HCO_3)_2$ and $CaCO_3$ aerosols in the submicron size range from solutions of $Ca(HCO_3)_2$. Hygroscopic growth and CCN activation of $Ca(HCO_3)_2$ was determined for the first time. The growth factor at 95%RH was 1.03 ±0.005, thus $Ca(HCO_3)_2$ was shown to be non-hygroscopic. On the other hand the critical diameter for cloud droplet activation was determined to 130±5 nm at SS=0.2% making $Ca(HCO_3)_2$ a good CCN. By long time studies in the Jülich Large Aerosol Chamber we showed that $Ca(HCO_3)_2$ was partly converted into $CaCO_3$ at dry conditions but most of it was persistent for several 10 hours. At humid conditions (40%RH) in the presence of traces of HNO_3 or added NO_X/NO_Y most of the $Ca(HCO_3)_2$ was converted into $CaNO_3$ but still some $Ca(HCO_3)_2$ was persisting over several hours. $Ca(HCO_3)_2$ could be formed from $CaCO_3$ and CO_2 in cloud droplets which contain mineral dust cores. If $Ca(HCO_3)_2$ would persist for some time after cloud droplet evaporation also in the atmosphere, its presence could ease the CCN activation of mineral particles (Zhao et al., 2010). In a campaign in the Jülich plant chamber (JPAC), the composition, hygroscopic growth, droplet activation, and optical properties of SOA from Mediterranean trees (Lang-Yona et al., 2010) were measured. Within a diploma thesis it was achieved to setting up and testing a compact scanning cloud condensation nuclei spectrometer (Strathmann, 2009).

WP-M1: Processes

Based on the Computational Fluid Dynamics (CFD) code Fluent and the Fine Particle Model (FPM), a comprehensive model to describe the fluid flow, heat/mass transfer, and particle/droplet dynamics in LACIS has been developed. Concerning the particle/droplet dynamical processes, the model features the description of particle hygroscopic growth, droplet activation and dynamic growth, and droplet homogeneous and heterogeneous freezing. Concerning immersion freezing, ice nucleation rates based on a) the stochastic and the singular approaches, and b) a parameterization (CNT-type rate expression based on experimental data gained at LACIS concerning the immersion freezing of coated ATD particles), were implemented and tested. Results indicate that the rate expressions based on the parameterization and the singular approaches describe the observed immersion freezing behaviour with sufficient accuracy.

WP-M2: Clouds

The Tel Aviv University group expanded the cloud model simulations to include the role of pollution in cloud development using the WRF 3D model with detailed description of cloud microphysics. At this stage only the newly developed bin microphysics for warm clouds based on Tzivion et al. (1987) and Rasmussen et al. (2002) were tested. The ice nucleation parameterization of Meyers et al (1992) was used. In the near future, new parameterizations based on newer formulations will be tried.

WP-M3: Climate

Using ECHAM5-HAM, the influence of varying the coating thickness of sulfate on the aerosol indirect effect has been investigated (Lohmann and Hoose, ACP, 2009). This effect amounts to between 0.12 and 0.2 W/m^2 in the global annual mean. The validation of an ECHAM parameterization of supersaturation, based on the Koop-threshold for homogeneous freezing, with CALIOP is ongoing. Supersaturation estimates in the upper troposphere from model and observations show quite good agreement. A variation of the homogeneous freezing threshold, as suggested by new measurements made in WP-L1, results in an increase in the upper tropospheric supersaturation frequency in the extratropics of up to 10%. ECHAM4 expanded by a multimodal microphysical ice scheme (see previous project reports) was applied to assess the impact of heterogeneous ice nucleation on global cirrus properties. A new method was developed to quantify the effect of heterogeneous ice nuclei on the crystal concentration, particularly in areas showing a high natural variability of cirrus microphysical properties which complicates a statistical analysis. Application of the new method reveals that heterogeneous ice nuclei from surface sources (black carbon and mineral dust) cause a zonal mean annual average reduction of the crystal concentration in cirrus of 10-20% in the tropics and 5-10% in northern midlatitudes. The reduction is enhanced by several percent in northern midlatitudes and the tropics when black carbon particles from aviation are considered as ice nuclei as well.

b) Achieved milestones

During the year 2009, all milestones were achieved as outlined in the work program of the project proposal. A list of milestone objectives and status by the end of 2009 is attached to the annual report (file <VH-VI-233_Report-2009_MS-Status.pdf>) and can be downloaded from the project homepage (http://imk-aida.fzk.de/viaci_engl/).

c) Fulfilment of budget and work plan

A financial report and budget justification for the year 2009 was provided by the Financial Department of the KIT. There are no major changes to the budget and work plans.

d) Publications (peer-reviewed papers, conference contributions,…)

See attached file <VH-VI-233_Report-2009_Publications.pdf>.