

# Particles and Cirrus Clouds (PAZI): Overview of results 2000 - 2003

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*Keywords:* aerosols, cirrus clouds, contrails, combustion, chemi-ions, soot, freezing, humidity, climate.

**ABSTRACT:** PAZI is a national research project supported by the German Secretary of Education and Research (BMBF) through the Helmholtz-Gesellschaft Deutscher Forschungszentren (HGF). Research in PAZI is performed in concert with the projects SiA, INCA, PartEmis, and PARTS funded by the European Commission. PAZI investigates the interaction of aerosols with cirrus clouds, with an emphasis on aviation-produced aerosols and contrails, and their impact on atmospheric composition, radiation, clouds, and climate. This overview summarizes important results obtained during the first phase and highlights the following issues. Measurements and models addressing the formation and evolution of black carbon (BC) particles in burners and jet engines; physico-chemical characterization of aircraft-produced BC particles; measured freezing properties of liquid and BC particles; calculated global atmospheric distribution of BC from various sources; observed differences in cirrus properties between clean and polluted air masses; correlations between air traffic and cirrus cloud cover deduced from satellite observations; process studies of aerosol-cirrus interactions; parameterization of cirrus cloud formation; representation of ice supersaturation and cirrus clouds in a climate model and possible aviation impact on global cirrus properties.

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## 1 THE PAZI PROJECT

The project studied the formation of soot particles and other aerosols in jet engines and in the atmosphere, their influence on the formation of the ice phase in contrails and cirrus clouds, and their impact on air composition, cirrus, and climate. It provided a first estimate of the contribution of aviation to changes of the upper tropospheric aerosol and ice particle budget.

To achieve these goals, the participating teams employed highly innovative and complementary methods: combustion test facility, aerosol/cloud chamber, ground-based lidar, research aircraft, satellite sensors, process and climate models. One cornerstone of the project was the close collaboration with projects funded externally by the European Commission and other agencies. Partners from industry such as Lufthansa, MTU, and Rolls-Royce Germany also contributed to PAZI.

Work in PAZI was organized in four main workpackages, the results of which are presented in Section 2 and summarized in Section 3. The workpackages contained several subprojects, each of which was represented by a responsible scientist. Collaboration within and between the subprojects and workpackages proved to be very fruitful, resulting in a large number of joint publications summarized in Section 4 (as of fall 2003).

## 2 WORKPACKAGE RESULTS

It is not possible to cover every single result of a large-scale project in the limited space available in workshop proceedings. What follows is a brief overview of some of the highlights of PAZI and its associated EU projects.

### 2.1 *Soot emissions and aerosol precursors*

This workpackage consists of three subprojects dealing with soot particle formation and prediction of global, fleet-averaged aerosol emission indices (Plohr et al., 2000; Döpelheuer, 2000, 2002; Kärcher et al., 2000; Kunz et al., 2001; Braun-Unkloff et al., 2001; Noll et al., 2002; Schumann et al., 2002; Böhm et al., 2003; Krüger et al., 2003; Wahl and Aigner, 2003). Measurement campaigns were carried out in laboratory flames, behind real jet engines and segments of combustors (hot end simulator), and in DLRs novel high pressure combustor test facility Stuttgart (HBK-S). The various techniques employed allowed the teams to characterize exhaust gas and aerosol chemical speciation as well as size distribution and state of mixing of exhaust aerosols.

The laminar coflame burner fueled with various non-methane hydrocarbons was used to measure temperature and soot volume mixing ratios. With this device, basic soot formation and oxidation processes were systematically studied as a function of operating pressure and fuel stoichiometry. With the data base set up over three years, a kinetic soot model describing soot formation was considerably improved. The HBK-S contains a subscale combustion chamber that operates with high mass fluxes, fully turbulent under high pressures, and with kerosene. It was shown that this device produces soot particles with size distributions similar to those measured in flight.

The combustor measurements carried out within PartEmis confirmed the weak dependence of soot emissions on the fuel sulfur content known from airborne campaigns, but more clearly revealed that exhaust soot contains significant amounts of organic material, besides fully oxidized sulfur. This enhances the hygroscopicity of soot particles and may probably influence their ice-nucleating behavior. Surprisingly, a new electrically charged soot mode at very small sizes (<10 nm in diameter) has been detected in mass spectrometric data of chemi-ions and carbon-containing particles and, independently, in measurements of the size distribution. The role of these ultrafine soot particles in ice formation is unclear and will remain under scrutiny.

An empirical model to estimate mass and number emission indices, size distributions, and total surface area concentrations of soot particles emitted by aircraft engines in flight conditions has been developed. This model requires only a few basic parameters as input, namely pressure and temperature at the combustor inlet and soot mass concentration at the combustor exit, which are correlated with the above soot properties. Using this correlation model in conjunction with the DLR fuel usage inventory from 1992, fleet-averaged global spatial and temporal profiles of soot emission quantities have been generated for use in the ECHAM general circulation model (see Section 2.4).

## 2.2 *Ice formation in the atmosphere*

This workpackage integrates four subprojects dealing with the measurement of freezing processes in atmospheric aerosols in cirrus conditions (Szakáll et al., 2001; Gierens et al., 2002; Ovarlez et al., 2002; Möhler et al., 2003; Kärcher and Haag, 2003; Haag et al., 2003a/b; Ström et al., 2003; Kärcher, 2003). A key tool to study ice nucleation in PAZI - with many instruments simultaneously determining thermodynamic conditions, gas concentrations, and the state of aerosol and ice particles - is the aerosol-cloud chamber AIDA in Karlsruhe. Data from the Microwave Limb Sounder (MLS) were analyzed to provide a global view of ice supersaturated regions. Ice supersaturation was measured on the cloud scale with aircraft in relatively clean and relatively polluted air masses during the INCA project.

Measurements in the AIDA very closely mimic real atmospheric conditions prevailing in cirrus or polar stratospheric ice clouds in terms of temperature, relative humidity, and cooling rate conditions. The ability of the numerical simulations carried out with DLRs microphysical cirrus model APSC (Advanced Particle Simulation Code) to provide detailed explanations of the observed formation of ice in supercooled aqueous sulfuric acid particles greatly increased confidence in attempts to model this process under real atmospheric conditions, provided that accurate temperature and humidity measurements are available. It was found that the threshold values of relative humidity for homogeneous freezing from recent laboratory studies, however, may not exactly hold when applied to atmospheric conditions, as enhanced supersaturations and non-equilibrium compositions of the largest liquid particles caused by rapid cooling and low temperatures can lead to somewhat higher values than determined assuming equilibrium conditions.

Besides supercooled liquid particles, heterogeneous ice formation triggered by pure and coated soot and mineral dust particles was also observed in the AIDA. The measurements indicate that these ice nuclei may initiate the formation of the ice phase at relative humidities over ice some 25% below the homogeneous thresholds (>145%). Interpretation of these data sets is ongoing, and more coordinated measurements and detailed comparisons are needed to understand existing differences between available laboratory data sets for soot and dust aerosols.

The fact that supersaturation of ice in the upper troposphere and tropopause region is a global phenomenon was corroborated by analyses of MLS data. For example, at 215 hPa, ice supersaturation is seen not only in the tropical and polar regions, but is also common at midlatitudes, in particular along the storm tracks. Naturally, these satellite data do not capture local variability. High supersaturations up to the homogeneous freezing thresholds have been measured in situ near but outside of and inside cirrus clouds during INCA. It was found that the onset of freezing over Prestwick occurred at relative humidities over ice significantly lower (at ~130%) than required for homogeneous freezing, suggesting that the northern hemisphere background aerosol over Prestwick exhibited signatures of pollution in terms of cirrus nucleation, in contrast to the data taken over Punta Arenas in the southern hemisphere. The ice formation mechanisms at work there likely involved a limited number of efficient heterogeneous ice nuclei. The most likely cause for the observed difference in the data sets are differences in chemical and morphological particle properties.

## 2.3 *Measurements of aerosols and cirrus clouds*

This workpackage encompasses three subprojects (Rother et al., 2001; Baehr et al., 2002; Gayet et al., 2002; Kärcher, 2002; González et al., 2002; Marquart and Mayer, 2002; Immler and Schrems, 2002a/b, 2003; Immler, 2003; Kärcher and Ström, 2003; Mannstein and Schumann, 2003; Minikin et al., 2003; Meyer et al., 2002, 2003; Seifert et al., 2003a/b/c). The main objective was to characterize aerosols and cirrus clouds in the atmosphere using a wide range of instruments: the large set of meteorological, chemical, optical, and microphysical probes employed in the INCA campaigns, Aerodyne's aerosol mass spectrometer operated by the University of Mainz, AWIs Mobile Aerosol Raman Lidar (MARL), DLRs aerosol and water vapor lidars, and remote sensing data from METEOSAT and ATSR-2 (Along Track Scanning Radiometer).

Vertical profiles of aerosol properties were taken at northern and southern hemisphere midlatitudes and in the tropics. The midlatitude data sets show surprisingly high concentrations of non-volatile particles at the respective tropopause, probably indicating that soot or dust particles reach these altitudes via complex transport processes in the lower(most) stratosphere. Ambient concentrations of nonvolatile condensation nuclei at 10-12 km altitude are ~35 cm<sup>-3</sup> (Prestwick) and ~12 cm<sup>-3</sup> (Punta Arenas). This demonstrates the potential for insoluble particles from natural and anthropogenic sources to influence the formation of cirrus clouds.

The counterflow virtual impactor (CVI) allowed the INCA team to accurately determine the total number density of ice crystals, down to the aerodynamic size range of a few  $\mu\text{m}$ . The derived distributions of number density show a predominant peak at values  $0.1\text{-}10\text{ cm}^{-3}$  and a broad feature extending to the detection limit of  $\sim 0.3\text{ l}^{-1}$ . The primary maximum at high concentrations is very likely caused by homogeneous freezing in vertical wind fields at times generated by rapid mesoscale temperature fluctuations and occasionally by convection. The broad feature at lower concentrations is determined by the variability in cooling rates, dynamically-induced dilution of cloudy air parcels, sedimenting ice crystals, and, in the case of the Prestwick data, heterogeneous freezing processes. It was inferred that buoyancy waves with mean wave periods of 10-20 min (horizontal length scales of several 10 km) induce mean updraft speeds of 25-35 cm/s were responsible for the generation of the high number densities of small ( $<20\text{ }\mu\text{m}$ ) ice particles in young cirrus. Such high cooling rates tend to minimize the impact of heterogeneous freezing on the total crystal number density (Section 2.2).

These results have important implications for climate studies. Climate change may bring about changes in the global distribution of updraft speeds, air mass temperatures, and aerosol properties. These changes could significantly modify the probability distribution of cirrus ice crystal concentrations. In any effort to ascribe cause to trends of cirrus properties, a careful evaluation of dynamical changes in cloud formation should be done before conclusions regarding the role of other anthropogenic factors, such as changes in aerosol composition, are made.

Mass spectrometric measurements were performed in one of the PAZI field campaigns carried out over Central Europe. The data confirm that sulfate is a prominent chemical constituent of accumulation mode aerosol there, and that aerosol nitrate was very low during these observations. The OLEX lidar detected an extremely tenuous cirrus cloud at  $52^{\circ}\text{N}/10^{\circ}\text{E}$  at an altitude of 13 km. The cloud was geometrically thin and optically invisible: it barely showed up in the backscatter signal at 523 nm wavelength, but clearly showed backscatter signal and volume depolarisation at 1064 nm. The  $\text{H}_2\text{O}$  lidar was operated simultaneously and recorded mixing ratios near 15-20 ppm. A rough analysis suggests that cirrus particles of 2-4  $\mu\text{m}$  mean radius caused the aerosol lidar signals. Taken together, the observed midlatitude subvisible cirrus cloud appears to be comparable to the ultrathin tropical tropopause clouds detected during APE/THESEO in 1999 – an interesting finding that requires further studies but demonstrates how little is actually known about this type of clouds.

The MARL recorded a large data set of cirrus cloud optical depths during INCA. Like OLEX, this lidar is sensitive to even very thin cirrus and can detect cloud with optical depths as low as several 0.0001 in the visible. The probability distributions of cirrus optical depth over Punta Arenas and Prestwick are similar, peaking around 0.2, the largest value being  $\sim 7$ . However, the distribution is skewed towards small optical depths, and about 1/3 of the measurements revealed the presence of subvisible cirrus with values  $< 0.02$ . These lidar studies thus confirm the surprisingly high frequencies of occurrence of subvisible cirrus clouds at midlatitudes reported earlier from measurements taken at the Observatoire Haute Provence, France.

The cirrus cloud cover over Europe was determined with the help of METEOSAT data in conjunction with actual air traffic movement data provided by EUROCONTROL. Typical background cirrus coverages amount to 20-25% in this area. Both data sets were correlated, revealing a linear relationship between coverage and mean air traffic density, which saturates at the highest traffic densities near 30-35% coverage. This result could be explained by persistent contrails being spread out through the action of wind shear, i.e., the generation of contrail cirrus, although indirect effects caused by freezing of soot particles without contrail formation or natural cirrus formation in the flight corridors cannot be fully ruled out. By implication, this study confirms the frequent presence and large extent of ice-supersaturated regions in the upper troposphere.

One subproject spent efforts on improving retrievals of cirrus parameters in the presence of lower level water clouds. A convincing case study was analyzed with ATSR-2 data during INCA. Yet another improvement concerned the use of ATSR-2 reflectances from the  $0.87\text{ }\mu\text{m}$  and  $1.6\text{ }\mu\text{m}$  channels to retrieve optical depth and effective particle radius in frontal cirrus over the North Sea. The APOLLO cloud classification tool was used to detect cirrus over sea and the cirrus properties were then retrieved by comparing the radiances calculated by the comprehensive radiative transfer package libRadtran with the radiances measured by the ATSR-2.

## 2.4 Modeling of aerosols and cirrus clouds

This workpackage integrates three subprojects dealing with process studies and global modeling (Ponater et al., 2002; Lin et al., 2002; Lohmann and Kärcher, 2002; Kärcher and Lohmann, 2002a/b, 2003; Lohmann et al., 2003; Gierens, 2003; Marquart et al., 2003; Hendricks et al., 2003a/b; Lauer et al., 2003). Two subprojects devoted their work to interpreting observations from INCA and MOZAIC, improving microphysical cloud models, and developing innovative parameterization schemes used in the climate model ECHAM. The global modeling subproject synthesized results from all workpackages and provided a first preliminary global assessment of soot-cirrus interaction.

Physically-based parameterizations of homogeneous freezing, heterogeneous immersion freezing, and initial growth of the pristine ice crystals were developed and validated with parcel model simulations. The DLR aerosol/cirrus model APSC and the cloud-resolving model MESOSCOPE participated in model inter-comparison exercises in the frame of the GEWEX (Global Energy and Water Cycle Experiment) Cloud System Study (GCSS) activity. Both, parameterizations and numerical solutions reveal a strong dependence of the number of ice crystals formed on the vertical velocity and the temperature. Aerosol size effects are comparatively less important, except for very efficient ice nuclei. Using the homogeneous scheme, the first interactive simulations of ice-supersaturation and cirrus were carried out in ECHAM, addressed further below.

Liquid particles may compete with heterogeneous ice nuclei during cirrus formation, as it was probably the case over Prestwick during INCA. Which particle type dominates depends on the relative concentrations, sizes, and freezing properties of the aerosol particles, the temperature and spectrum of vertical winds. The critical number densities of ice nuclei above which homogeneous freezing is suppressed were calculated analytically. Evaluating this solution at typical conditions prevailing during INCA (freezing temperature 222 K, mean updraft speed 20-30 cm/s, heterogeneous ice nucleation at 130%) reveals that less than 0.03-0.08 ice nuclei per  $\text{cm}^3$  of air must have been present during the measurements, because homogeneous freezing did take place. It was concluded from the Prestwick observations that heterogeneous ice nuclei triggered the first ice particles but was not the dominant freezing mode (see Sections 2.2 and 2.3), consistent with this estimate. Efforts to corroborate this hypothesis with detailed microphysical simulations are underway.

The implementation of the new cirrus parameterization and the simultaneous abandonment of the frequently used saturation adjustment scheme resulted in the first calculations of global relative humidity in ice-supersaturated regions. Comparisons with MOZAIC data showed that the typical, quasi-exponential shape of the distribution function could be reproduced by the model. Previously, excess humidity created during one time step was removed instantaneously and defined as cloud ice. Basic cirrus parameters such as the total number of crystals and the effective radius used in subsequent radiative calculations were prescribed. With the new parameterizations, the simulated ice water contents are lower and the total crystal concentrations are significantly higher than those computed with the standard saturation adjustment schemes, in better agreement with observations.

The improved cloud scheme enables a self-consistent treatment of cirrus, rendering new studies of cirrus cloud-triggered feedbacks in the climate system possible. It was shown that even very strong enhancements of the background sulfate aerosol mass caused by the Mount Pinatubo eruption likely caused only weak changes in cirrus properties, consistent with a recent reevaluation of ISCCP (International Satellite Cloud Climatology Project) data. However, alterations of the properties of very thin and subvisible cirrus by volcanic eruptions could not be excluded. This implies that the initially very small (5-10 nm) aviation-produced volatile aerosols, albeit present in relatively high number concentrations, do not exert any significant impact on cirrus cloud formation.

The global distribution of black carbon soot was reevaluated with the global model ECHAM. The simulations were based on updated emission inventories, including the aircraft soot emission model described in Section 2.1. It was found that several  $0.1 \text{ cm}^{-3}$  soot particles in the upper troposphere and tropopause region originate from aircraft emissions (annual mean figure); compared to the  $<0.03\text{-}0.08 \text{ cm}^{-3}$  heterogeneous ice nuclei that influenced cirrus formation over Prestwick (see above), these emissions seem to be sufficiently high to impact cirrus formation in principle, provided that a corresponding fraction of the soot particles act as efficient ice nuclei. The exact magnitude of the calculated aircraft-induced perturbations of the soot budget depends on the relative contributions of the other black carbon sources and on details of soot ageing processes in the atmosphere on time scales of several weeks, both of which are not well known.

A first attempt to calculate the global impact of aircraft soot emissions on cirrus was also undertaken at the very end of the project. For this purpose, two scenarios were defined. The default scenario calculates cirrus formation caused by homogeneous freezing only. This is contrasted with a scenario where heterogeneous freezing of aircraft-emitted soot particles is prescribed, but only in regions where the aircraft soot concentrations exceed  $0.5 \text{ cm}^{-3}$  (to compensate for errors in the simulated vertical winds that are too low compared with observations); otherwise cirrus form homogeneously as in the first case. The soot increase by aircraft emissions was maximized by assuming minimal particle losses during ageing. In treating this indirect effect of soot-cirrus interactions, the direct effect of spreading contrails (as noted in Section 2.3) has been neglected in these studies.

By comparing annually averaged results from ten years of ECHAM simulations, total ice crystal concentrations are found to be reduced by several tens of percent at northern hemisphere midlatitudes. This is explained by the fact that the assumed soot threshold concentration above which homogeneous freezing is suppressed at the prevailing model updraft speeds is frequently surpassed by aircraft emissions. In another conceivable scenario, where soot particles (irrespective of their origin) as well as mineral dust particles can serve as heterogeneous ice nuclei, aviation leads to enhancements of ice crystal concentrations. Feedbacks of the cirrus changes on cloud cover have not been considered in the two scenarios. However, the ECHAM results obtained here reveal the potential of aircraft-induced soot particles to cause significant changes in cirrus microphysical properties.

### 3 SUMMARY AND CONCLUSIONS

- Among all exhaust particles, soot is key in further studies of potential aerosol-cirrus interactions
- Kinetic algorithms improve the prediction of soot physical properties in combustion models
- Accurate measurements better define the chemical composition and the size distributions of soot particles emitted by aircraft jet engines
- Innovative measurements in an aerosol/cloud chamber suggest early freezing of soot particles in cirrus conditions
- Interpretation of freezing experiments in the chamber increases confidence in the current ability to model homogeneous freezing and growth of small ice crystals in cirrus
- In-situ studies unambiguously confirm the presence of high number densities of small ice crystals in cirrus, tied to homogeneous freezing
- Aircraft-based, size-resolved chemical analysis of aerosol particles in the accumulation mode become available, providing important information on freezing aerosols in future campaigns
- Field measurements reveal mesoscale variability in vertical velocities as the key factor controlling cirrus formation
- Lidar studies confirm surprisingly high frequencies of occurrence of midlatitude subvisual cirrus
- Retrieval algorithm allows remote sensing of cirrus cloud optical properties in the presence of low level stratus
- Satellite analysis reveals close correlation between cirrus cloud cover changes and air traffic density
- Global model studies better constrain the apportionment of black carbon soot to a large number of sources, including aircraft
- Freezing parameterization enables more realistic predictions of ice supersaturation and cirrus clouds in global models
- First preliminary global assessment of soot-cirrus interaction shows potentially significant indirect impact of aircraft soot on cirrus crystal concentrations, if the effect is maximized

Future progress in the area of aerosol-cirrus-radiation interaction depends crucially on a better understanding of

- small-scale dynamical variability;
- indirect aerosol effects on cirrus;
- cirrus radiative properties and cover;

and the representation of these effects in global atmospheric models.

The viewgraphs of the invited talk presented at the workshop are available under <http://www.pa.op.dlr.de/aac/>. Most of the material presented here has been published in the open literature and we refer to the list of references for more details. In addition, the following contributions in this volume provide further information: Wahl et al., Petzold et al. (Section 2.1); Möhler et al., Mangold et al. (Section 2.2); Immler et al., Mannstein et al. (Section 2.3); Marquart et al., Hendricks et al. (Section 2.4).

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