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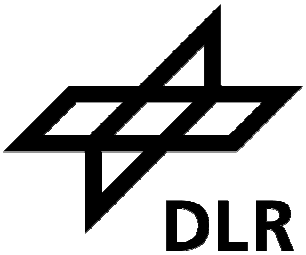

LIST OF ABSTRACTS

Oral Presentations

Session 1 : Engine emissions and plume processes

Novel Rates of OH induced Sulfur Oxidation. Implications to the Plume Chemistry of Jet Aircraft

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A number of environmental aspects of aircraft emissions such as contrail formation and impact on cirrus formation have been suggested to depend on sulfuric acid formation from the fuel sulfur content (FSC) as a result of the rapid rates of oxidation of S(IV)in the engine and/or the plume. Despite this importance the chemical kinetic date base to assess this oxidation is far from being reliable.
Novel rate coefficients for the most important and rate controlling sulfur oxidation reaction, $OH + SO_2 \rightarrow HSO_3$, over an extended range of pressure and temperature have been derived from ab initio quantum chemical/RRKM dynamical calculations. From these calculations the rate of oxidation of S(IV) to S(VI) under typical conditions of a jet aircraft plume is predicted to be considerably slower than previously accepted on the basis of interpolations of experimental data (i.e. Tremmel and Schumann,1999). This is maily due to the nature of the fall-off behaviour of the rate coefficent as well as to a slight revision of the equilibrium constant.
We have incorporated these kinetic results into a chemical-dynamical code of the jet regime of a B-747 airliner (BOAT code) and predict sulfur conversion efficiencies in this regime of less than 1%. It is shown that this efficiency depends on the OH emission factor as well as on the evolution of the OH field in the early plume. Because this field also depends on the emission factors of NO_x and organics, the impact of these factors on the sulfur conversion efficiency has been tested.
It is concluded from our work, that the well-known conversion ratio of S(IV) to S(VI) of about 1-2% as confirmed by a number of airborn experiments, can only be reproduced assuming sufficient formation of SO_3 or H_2SO_4 already inside the engine and/or the turbine. The plume effect on this ratio is less important.

Determination of Soot Mass Fraction, Soot Density and Soot Fractal Character in Flame Exhaust Gases

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The DLR – Soot Generator was used as a variable well defined soot source. The particle mean diameters of the log normal size distributions can easily be shifted between 6nm and 250nm.
This soot loaded exhaust gas is sucked through a quartz fiber filter via a computer controlled gas sampler. The soot particles are trapped on a quartz fiber filter. This special quartz filter has a sampling efficiency better than 99,9% for particles between 6nm and 250nm. The carbon load on the quartz filter is burned in an oxygen atmosphere. The resulting carbon dioxide concentration is measured with a Fourier Transform IR spectrometer (FTIR). If the gas sampling volume, the gas cell volume and the carbon dioxide concentration is known, a soot mass fraction can easily be calculated.
The corresponding size distributions, number concentrations and volume concentrations are measured with a Scanning Mobility Particle Sizer system (SMPS). Using the soot mass of experiment one and the soot volume of the SMPS measurements, a soot density was calculated. This soot density is based on the mobility diameter of the fractal soot particles. The quotient of graphite density and calculated soot density gives the fractal character of the soot.

Overview of Results from the NASA Experiment to Characterize Aircraft Volatile Aerosol and Trace Species Emissions (EXCAVATE)

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EXCAVATE was conducted at Langley Research Center in late January, 2002, and focused upon assaying the production of aerosols and aerosol precursors by a modern commercial aircraft, the Langley B757, during ground-based operation. The experiment was motivated by remaining uncertainties in the post-combustion fate of jet fuel sulfur contaminants and the need to obtain observations for evaluating the impact of terminal area aircraft operations upon local air quality. Sponsored by NASA's Atmospheric Effects of Aviation Project (AEAP) and the Ultra Effect Engine Technology (UEET) Program, EXCAVATE objectives included determining exhaust black carbon levels and gas ion densities as a function of plume age and engine power; the fraction of fuel S converted from S(IV) to S(VI) as a function of engine power and fuel S level; the concentration and speciation of volatile aerosols and gas-phase acids as functions of engine power, fuel S, and plume age. To accomplish these objectives, participants from NASA Langley, NASA Glen, the Air Force Research Laboratory, Aerodyne, and the University of Minnesota, placed fast-response instruments downstream of well characterized aerosol and gas sample inlets and acquired measurements behind both the Langley T-38 (J85-GE engine) and B757 (RB211) aircraft at sampling distances ranging from 1 to 35 meters as the engines burned fuels of various sulfur concentrations and ran their engines at settings ranging from idle to near take-off power. Preliminary observations indicate that chemion densities were very high in the exhaust of both aircraft, consistent with values that are presently being used in microphysical models of aerosol formation in exhaust plumes. Both aircraft were found to emit high concentrations of organic aerosols, particularly at low power settings and to produce black carbon concentrations that increased with engine power. Although observed aerosol size distributions and number densities were highly dependent upon the sample dilution ratio, total particle emission indices for the B757 were typically a factor of 10 higher at 25 to 35 meters than at 1 meter behind the engine. The concentration of sulfate aerosol were directly dependent upon the fuel sulfur level and increased considerably as sampling took place progressively further downstream of the exhaust plane, suggesting that sulfate particles form and undergo rapid growth within aircraft exhaust plumes. Our observations also indicate that aerosol concentrations and characteristics take several minutes to reach equilibrium values after changes in engine power. This was particularly notable when the engines were reduced from high to low power, a situation that would be found during aircraft taxi and landing cycle.

SAE E-31 Committee on Aircraft Exhaust Emission Measurements and an Aerospace Information Report on the Measurement of Non-volatile Particle Emissions

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Agencies responsible for regulating and certifying aviation operations have begun to examine methods for measuring particle emissions from aircraft engines. There is general consensus that the regulations regarding the emission of visible smoke for aircraft engines, which have been in place for decades, do not address and are not relevant to the measurement of particles responsible for health effects and environmental impacts. Working Group 3 of the ICAO Committee on Aviation Environmental Protection (CAEP) has asked the SAE E-31 committee for technical assistance in developing appropriate particulate characterization techniques for routine certification of aircraft turbine engines. The SAE E-31 committee has specified measurement techniques and protocols for aviation emission measurements for many existing regulations and the committee has accepted these requests for the specification of small particle emissions measurement. It is the intent of the E-31 committee to make use of both committee expertise and outside technical advice to develop a set of recommendations that will form the basis for an Aerospace Information Report (AIR). This AIR will be subject to evaluation and review by the regulatory agencies, industry, and the engineering community that performs aviation emissions measurements. This AIR is currently being written and reviewed and its general content will be presented. Based on the experience gained and on

improvements in measurement practice, the AIR will then be used over the course of several years to develop a set of measurement specifications described in an Aerospace Recommended Practice (ARP) on particle measurements. ARPs are the official statement of the SAE on how emissions measurements should be performed and, as such, have historically provided methodologies acceptable to the regulatory agencies both in the US and internationally.

SO3 and H2SO4 in Exhaust of an Aircraft Engine: Measurements and Implications for Fuel Sulfur Conversion to S(VI) and SO3 to H2SO4

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Gaseous sulfuric acid (GSA) formed by aircraft engines is of considerable current interest as it plays a potentially important role in forming and activating aerosol particles which become water vapour condensation nuclei. The later promote the formation of contrails and potentially even of clouds. GSA is formed via fuel sulfur oxidation to SO3, followed by SO3 reaction with water vapor leading to GSA. The most important questions in this process are: (i) which fraction of S(VI) gases present in the aircraft engine exhaust is formed already in the combustor and (ii) which fraction of S(VI) emits as SO3 molecules? The later means an incomplete conversion of S(VI) to GSA in an exhaust plume.
The presentation reports on first experimental estimation of the conversion (ε) of fuel sulfur to S(VI) = SO3 + H2SO4 and conversion (ε A) of SO3 molecules to H2SO4 in an exhaust at the exit of aircraft gas turbine combustor. Here ε = [S(VI)]/(ST), εA = [H2SO4]/[S(VI)] and ST is a total sulfur atom concentration. The major results of the presented CIMS-experiments and their interpretation with a model simulation are: (i) The efficiency ε =2.3 $^{+}/_{-}$ 1 % at exhaust age about 0.5 ms from the combustor exit; (ii) The SO3–molecules represent a major fraction of S(VI)-gases ε A<50 % and an essential SO3-conversion to H2SO4 takes place in the sampling line with a sufficiently long time of spending and where the temperature is lower than in a hot exhaust. The coincidence of ε from our work (the measurement for the sampling point in exhaust just behind the combustor exit) and ε from the measurements in an exhaust at plume age about 1 s suggests that the S(VI)–formation is inefficient in the post-combustor flow inside of aircraft engine.

Particle Emissions from Aircraft Engines - an Overview of the European Project PARTEMIS

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In the framework of the European project PartEmis („Measurement and prediction of emissions of aerosols and gaseous precursors from gas turbine engines“), the influence of operation conditions and fuel sulphur content (FSC) on the microphysical and chemical properties of particles emitted from a jet engine simulator was investigated. This engine simulator consisted of a real jet engine combustor and a so-called Hot End Simulator (HES) which simulates the pressure and temperature profiles found in a jet engine turbine section. In a first experiment, the emission properties of the combustor were studied. These data were then used as boundary conditions for a second experiment which focused on the emission properties of the combustor-HES combination. The aerosol microphysical and chemical properties investigated in this study covered number, size, and mass concentration of primary combustion aerosol particles which form in the combustion process and secondary volatile particles which form outside the combustor in the cooling plume. Furthermore, the volatile fraction of internally mixed combustion particles, particle hygroscopicity, and cloud condensation nuclei (CCN) activation potential were studied. In addition, the emission of non-methane

volatile organic compounds (NMVOCs) was monitored.
The combustor-HES combination was operated at two inlet conditions with different exhaust temperatures representative of the gas path temperatures of older and more modern jet engines. Fuels with different FSC (50, 410 (ave. found in aviation fuel) and 1270 µg g-1) were burnt. The employed aerosol measurement methods consisted of various size-selective Condensation Particle Counters, Differential Mobility Analysers including a Hygroscopicity Tandem DMA and a Volatility Tandem DMA, aerosol absorption photometers, thermodenuder methods, a Cloud Condensation Nucleus Counter, and an extensive set of chemical analytical methods. The overview will summarise the observed influences of engine operation conditions and fuel sulphur content on microphysical and chemical properties of emitted particles. Conclusions on expected effects of the exhaust aerosol on atmospheric properties will be discussed.

Emission of non-methane volatile organic compounds (NMVOCs) from a jet engine combustor and a Hot End Simulator (HES) during the PARTEMIS project

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During the PARTEMIS measurement campaigns at QinetiQ, Pyestock in January 2001 and in March 2002 the emissions of more than 100 different non-methane volatile organic compounds (NMVOCs) from a jet engine combustor and a hot end simulator (HES) were identified and quantified. The species investigated accounted for up to 91 wt % of the total NMVOCs emitted. In addition, CO_2 measurements were also performed for determining emission indices (EI).
The C_2-C_{15} aliphatic and aromatic hydrocarbons were monitored using a compact online gas chromatograph (GC) (Airmovoc 2010) with enrichment system and FID detector. The hydrocarbons (HCs) were measured with a time resolution of 10 min and detection limits in the pptV range.
Selected partially oxidised hydrocarbons such as aldehydes and organic acids were collected by using special sampling cartridges and measured off-line using HPLC with a photo-array detector. The partially oxidised HCs were measured with a time resolution of 4 min and detection limits in the ppbV range.
A fully automated online GC system (Chromato-sud) with TCD detector was used to monitor CO_2. Measurement cycle and detection limit of this system were 5 min and 10 ppmV, respectively.
Sample line effects on the measured data were investigated and are included in the error bars of the data. The NMVOC emission indices of the combustor and HES measurements (e.g. mg benzene/kg fuel burnt), which were calculated by using the CO_2 data were found to be comparable with data from the previous AEROTRACE study. During the PARTEMIS campaigns the influence of combustor power, pressure in the different stage of the HES and different fuel sulphur content (FSC) on the emission of the species studied was also investigated and will be discussed.

Modeling of Soot Precursor Formation in Laminar Premixed Flames with C1-, C2- and C6-Fuels

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Aromatics and polycyclic aromatic hydrocarbons (PAH) are of particular concern in combustion processes because of their potentially adverse health effects. They are formed in the combustion of hydrocarbon fuels (e.g. kerosene) and have been identified as key precursors of soot.
Former investigations of flame and shock tube experiments, especially at high pressures, show the importance of the PAH growth on soot particle inception and on calculated soot volume fraction. However, for high pressure conditions, due to the very thin reaction zone it is not possible to measure PAH profiles within and close to the very thin reaction zone. Therefore, PAH profiles measured in recent experiments with laminar premixed flames at atmospheric pressure only have been used as data base for simulation. In these slightly sooting flames of aliphatic (methane, ethane and ethene) and aromatic hydrocarbons (benzene) a large variety of PAH (besides aliphatic compounds) has been measured by gas chromatography / mass

spectrometry ranging from two- to five-fused aromatic rings.
For comparing calculations a recent comprehensive gas phase mechanism from literature and an improved gas phase mechanism established by the authors were used. It contains detailed chemistry for PAH growth as combinative growth steps of aromatic species and reactions to form species up to C30. Measured and predicted PAH profiles were compared and the main routes for PAH growth were identified by reaction pathway analysis. These different reactions pathways for the formation of PAH, the dominant species for soot precursor formation, were examined numerically to obtain a deeper understanding of the soot inception process.
Additionally for some of the experimental results soot volume fractions have been simulated by using a soot model, which combines a gas phase mechanism and models for particle inception, coagulation, surface growth by gaseous species and soot particle oxidation.
As in all current soot models particle inception is modeled exclusively by coagulation of different large mass PAH. The influence of different PAH on the onset and the amount of soot was investigated systematically. Only a few PAH have to be considered for simulation of the inception regime.

Stable Carbon Isotope Signatures of Aircraft Particles

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The stable carbon isotope ratio ($^{13}C/^{12}C$) of aircraft engine emissions of particles and polycyclic aromatic hydrocarbons was measured using gas-chromatography isotope ratio mass spectrometry. Samples were taken from the combustor exit and at a simulated engine exit (the PARTEMIS hot-end siulator). The stable C isotope ratio on the particles and PAHs was shown to be consistently different from the parent fuel ratio, and exhibited a ratio that was different to other common combustion sources. The signal was shown to be fuel independent (using kerosene, petrol and diesel) and engine condition dependent. There was a small consistent variation from behind the combustion zone to combustor and engine exits. PAHs were measured in both gas and particle phases and no variation in C isotope ratio was observed. These preliminary observations indicate that this technique, in conjunction with more advanced isotope-ratio techniques (e.g. using deuterium) may enable a better source reconciliation of aviation particles over other sources.

Modelling of volatile particles during PartEmis

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In the frame of the European project PartEmis, volatile particles produced in the sampling system of a combustor test rig has been modelled. These particles, although formed in situations which differ from those prevailing during flight conditions, remain of major interest as their growth is highly connected to the amount of sulphuric acid available in the exhaust. In this matter, attention has been paid to the sulphur conversion factor required to fit the modelled results to the measurements of volatile particles, in the 4 to 7 nm size range. The specific sampling system features (temperature, pressure, gas and particle losses to the wall etc.) have been modelled as well as particle formation, using a coagulation based model commonly used for in flight aircraft modelling. Our first results indicate that for the highest fuel sulphur content used (1270 µg of sulphur per kg of fuel), a sulphur conversion of about 2.2 % to 3 % at the exit of the combustor is necessary to reproduce the measurements. This average value is close to the experimental values, if we take into account uncertainties associated to measurements. In addition, our results has been found to be in good agreement with experiments carried out to study growth factors of soot particles at high relative humidities. As a conclusion, the modelling of particle formation in the sulphuric acid-water binary system has provided an indirect way to estimate an upper limit of sulphur conversion, one of the key parameters needed to understand particle formation and evolution in an aircraft plume.

Growing and Dispersion of Particles in a Turbulent Exhaust Plume

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Emissions of water vapor, sulfur dioxide and particles (soot, metallic particles…) by jet engine is known to induce the formation of new particles , i;e; aerosols and contrails. These particles may have an impact on cloudiness and may affect the Earth's radiative budget balance. In order to better understand their formation, preliminary studies on the dispersion of particles (soot and aerosols) in the exhaust jet and on their modification by plume processing are necessary.
This work is focused on the numerical simulation of dynamics and growth of spherical particles in the near-field of an aircraft wake.
Three-dimensional temporal evolution of a gas hot round jet is provided by performing large eddy simulation (LES), in order to assess velocity, temperature and condensable species (water vapor) turbulent fields. The LES technique is associated with subgrid scale models accounting for the flow compressibility.
A condensation and transport model has been developed using the Lagrangian particle tracking approach. In the context of a dilute solid phase in the carrier fluid, the particle collision and the influence of particles on the flow are neglected. Fukuta & Walter's model provides the temporal evolution of a spherical particle radius as a function of the condensable vapor supersaturation. Particle formation and growth inside the wake flow occur via heterogeneous nucleation involving active particles, initially concentrated in the jet.
First results have shown that the particle growth could be significant for sufficiently small initial sizes of particles. For the present particular set of parameters, the particle growth does not increase significantly the inertia of initially small particles. For two different initial particle sizes, the water vapor condensation process gives rise to maximum final particle radii of the same order. In the case of initially large condensation nuclei size, particles have significant inertia and concentrate at the periphery of the large-scale structures developing in the turbulent plume.
The study is focused on the coupled effects of particle growth and dynamics, which are not considered in classical particle-laden turbulent plume models. As most of previous studies correspond to incompressible flow, the influence of the gas flow compressibility on the particle dispersion will be also investigated.

The Effect of Plume Processes on Aircraft Impact

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In the present study, we investigate the chemistry of expanding aircraft exhaust plumes for a range of conditions (latitude, altitude, season, plume expansion rate, time of day of emissions, aircraft type, composition of background air mass). The effect of plume chemistry on 2-D model calculations of the impact of subsonic aircraft on ozone is discussed.
An expanding Gaussian plume model is used to represent the plume dynamics and the chemical solver incorporates a comprehensive tropospheric reaction set. Before each plume simulation, a five day background spinup is done, during which O3 and HNO3 are held constant and NOx is reset every noon. Species are initialised using available climatologies. The effect of composition of the background atmosphere was tested by comparing calculations for different air masses identified in the SONEX measurement campaign. Plume processing is reported using both emission conversion factors
[Kraabol et al., 2000] and relative emission changes [Petry et al., 1998].
Conversion of NOx to NOy in the plume was found to be a strong function of altitude, latitude, season and composition of the background air mass. Conversion is determined largely by O3 and HOx in the background air mass. HOx is responsible for conversion in the daytime, while ozone affects the night-time conversion to N2O5 and, ultimately, to HNO3 via heterogeneous chemistry. Ozone also contributes to HOx production and so affects both daytime and night-time conversion rates but, because there are other sources of HOx, the effect on night-time chemistry is greater. Because night-time chemistry is less subject to non-linearities in the chemistry than daytime chemistry, the magnitudes of relative emission changes are smaller for air masses influenced strongly by night-time chemistry.
Modifying subsonic aircraft emissions in the CSIRO 2-D chemical transport model using relative emission

changes from the plume model resulted in changes to the calculated aircraft impact on ozone of less than 3%. The effect of using emission conversion factors instead of relative emission changes in the 2-D model is also discussed.

Aviation fuels - Where are we going and why?

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Gas turbine fuels for aviation are taken for granted. Their main purpose is to store energy until it is released in the combustion chamber. However, this is only part of the fuels use. It is also used as a coolant and lubricant. Some of the fuel characteristics that have been associated with aircraft emissions, such as sulphur and aromatic content, can also affect the usability of the fuel.
Sasol have been producing a semi-synthetic aviation fuel from coal and using it in commercial aircraft since 1999. Production processes such as Fisher Tropsch methods and gas to liquid techniques can generate kerosene's that are free of aromatics and virtually sulphur free, but at what cost to the engine?
In May this year a workshop, organised by AERONET 2, was held looking at future clean fuels for aviation. The objective of the workshop was to review the status quo, potential future fuel developments, both evolutionary and revolutionary, and their possible consequences on emissions and engine technology. This presentation will report on the findings of the workshop and give a layman's guide to the use of fuel in today's gas turbines.

Session 2: Transport and impact on chemical composition

NOy in the UT/LS: A Source Attribution Study Utilising MOZAIC Measurements.

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MOZAIC (Measurement of OZone, water vapour, carbon monoxide and nitrogen oxides by Airbus In-service airCraft) measurements are combined with output from the Cambridge 3D global model (TOMCAT) in order to study NOy chemistry in the UT/LS. This region is subject to both biogenic and anthropogenic emissions, the impact of which are highly variable due to the strong gradients in chemical concentration across the tropopause. The contributions of different emission sources within TOMCAT, including in-situ aircraft emissions, to both ozone and its production rate will be presented.

The TRADEOFF project: Goals and achievements

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TRADEOFF is an EU funded project in which 10 European research groups participate. The main goal of the project is to calculate current and future impact of a rapidly growing fleet of aircraft (composition, climate), to reduce the uncertainties in the calculated long term impact of aircraft emissions, and to identify options to reduce future impact of aircraft emissions. The focus have been on processes and impact in the UTLS (upper troposphere/lower stratosphere) region, where predicted rapid growth in air traffic and in aircraft emissions could significantly perturb atmospheric composition (e.g. NOx and ozone concentrations, the formation and distribution of contrails and cirrus clouds) and contribute to forcing of climate. The TRADEOFF

project covers a wide range of research topics: Testing and improvement of model performance through extensive model/model and model/measurement comparisons; studies of chemical and dynamical processes affecting composition and changes in the in the UTLS region; updated scenarios for aircraft emissions; estimates of the impact of NOx emissions on ozone and methane lifetime for the current and future (2050) atmospheres; analysis of satellite observations of contrails and cirrus clouds in corridors with high air traffic, and estimates of their radiative forcing. Furthermore, specific modelling studies have been performed to quantify possible tradeoffs in air traffic to reduce the climate impact. These include, traffic at specific times during the day, changes in cruising altitudes and in routings. Some limited studies of the impact of a potential future fleet of supersonic aircraft flying in the lower stratosphere were also performed. Results from the TRADEOFF project presenting new estimates of the impact of NOx on ozone and CH4, including sensitivity estimates, based on updated emission inventories and improved model tools will be presented. Likewise, results from the analysis of contrail and cirrus cloud studies will be presented. Finally, estimates of the radiative forcing from aircraft, based on the TRADEOFF studies, will be presented.

On the quality of chemistry-transport simulations in the upper troposphere/lower stratosphere region

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In the framework of the EU funded project TRADEOFF the quality of the performance of five European chemistry transport (CTM) and two general circulation models (GCM) has been evaluated through a detailed comparison with observations. The study helps to better assess the models‘ capability of estimating the impact of aircraft emissions upon the chemical composition of the atmosphere. We placed a special emphasis on the upper troposphere/lower stratosphere (UT/LS) region where most air traffic emissions occur and where changes in the greenhouse gas ozone have the largest impact on climate change. For this purpose an extensive database of insitu observations of major compounds related to O3 photochemistry was established for the 4-year period 1995 to 1998, covering observations from the most relevant commercial and scientific aircraft measurement campaigns as well as O3 soundings. We will present a one-by-one comparison of simulated and observed trace gas concentrations and we will focus on the UT/LS region which is particularly difficult to simulate for several reasons: A high vertical model resolution is required to accurately represent the steep tracer gradients across the tropopause. Convective processes, which are difficult to simulate, strongly affect the photochemistry of the UT region by rapidly lifting upwards pollutants emitted at the surface, and lightning associated with convection provides an important source of nitrogen oxides to this region. Production of OH radicals appears to depend not only on O3 and H2O levels but also on carbonyls and peroxides whose concentrations are again strongly linked to vertical transport. Finally, due to the long lifetimes of many compounds in the UT/LS region small inaccuracies in the advection scheme may have a significant effect on their concentration levels. The abilities and limits of the participating CTMs and GCMs will be discussed in light of these issues.

Lightning NOx emissions and the impact on the effect of aircraft emissions - Results from the EU-project TRADEOFF

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The major upper troposphere NOx emissions are lightning (5 TgN per year) and aircraft (0.7 TgN per year) emissions. The main lightning NOx source is located at low latitudes and therefore separated from the main aircraft emission region at mid latitudes. However, transport of upper troposphere tropical air masses to

lower latitudes mixes air masses of tropical and mid-latitude characteristics, i.e. with lightning and aircraft NOx characteristics. Simulations with the global climate-chemistry model E39/C are presented, which demonstrate these effects and which enables the possibility to quantify these effects. The impact of aircraft emissions upon the chemical composition of the atmosphere (especially ozone) has uncertainties not at least because the lightning NOx source is only poorly known in terms of total strength and spatial distribution. This will be highlighted and quantified based on simulations with varying vertical distributions of the lightning NOx source.

Impact of Present-Day and Future Subsonic Aircraft Emissions on Tropospheric Ozone and Associated Radiative Forcing of Climate

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Subsonic aircraft release significant quantities of chemical compounds into the upper troposphere and lower stratosphere. Nitrogen oxides NO_x (= NO + NO_2) are of particular importance, since they have the potential to modify the ozone concentration near the tropopause and hence perturb the radiative forcing on the climate system. In addition to that, changes in NO_x associated with aircraft emissions will also increase the hydroxyl radical OH leading to a reduced methane residence time in the atmosphere.
Within the EU-project TRADEOFF, state-of-the-art global atmospheric chemistry models have been used to investigate the present-day and future (2050) changes in atmospheric composition. In this paper, we present and discuss the results from this study and summarize the changes in NO_x, O_3, and OH, generated by several chemical transport models. These chemical perturbations are then used as input to radiative transfer model in order to quantify the ozone and methane forcings on climate. The methane forcing partly offsets the positive forcing associated with ozone on the global scale. However, geographical and temporal distributions of the two effects are significantly different. These features will be illustrated.

Impact of aircraft NOx emissions: Effects of changing the flight altitude.

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Within the framework of the European TRADEOFF project, model studies have been performed to investigate the altitude dependence of the impact of NOx emissions from aircraft on the chemical composition of the atmosphere. Here we present results from two chemical transport models (the Oslo CTM-2 and the Cambridge TOMCAT model) and one coupled climate-chemistry model (the DLR E39C model). The model simulations focus on present (year 2000) conditions and use different global aircraft emission scenarios, which were developed for TRADEOFF: 1) base case assuming normal cruise altitudes, 2) aircraft cruising 2000 ft higher, and 3) aircraft cruising 6000 ft lower. As current cruise altitudes are determined by fuel efficiency considerations, both an increase and a decrease in flight altitude will result in enhanced fuel consumption and higher NOx emissions. By considering additional scenarios where the total NOx emission is normalized to the base case, we separate the effect of changing cruise altitudes and concomitant increases in fuel consumption.
Lower emission heights of NOx result in lower residence times of aircraft NOx in the atmosphere since wash-out processes are more likely to occur at lower altitudes. Therefore, the aircraft-induced increase in ozone is clearly smaller than in the base case. The enhanced fuel consumption compensates for this only to a minor degree. By contrast, higher cruise altitudes lead to an increase in aircraft impact, as more emissions occur in the stratosphere where pollutants have a significantly longer residence time.

CTM Simulation of Tropopause Ozone: Lessons from TRACE-P

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The ability to calculate aviation's impact on the atmosphere, whether through gases or particles, relies on accurate simulation of the transport and mixing processes near the extra-tropical tropopause where most emissions occur. The recent TRACE-P campaign combined extensive airborne measurements (including ozone Lidar and sondes) with new high-resolution, EC-forecast meteorological datasets to drive the chemistry-transport models. While we have identified some systematic biases of the CTM, these high-resolution simulations (180 x 180 x 1 km) do a remarkable job of simulating the observed temporal and geographic patterns observed, including the correlations of ozone and carbon monoxide in the tropopause region. Such CTM validation tests, although admittedly for only one region and season, support the tracer transport in this new model; and we compare our new results for the idealized case of aviation exhaust accumulation with previous studies.

Improved mass fluxes in a global chemistry-transport model: implications for upper tropospheric chemistry

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Calculating the impact of aircraft emissions is a particularly difficult task, since the largest fraction of these emissions occur in the upper-tropospheric and lowermost-stratospheric (UTLS) region. Global chemistry-transport models have great difficulties to simulate trace gas concentrations in this region that is characterised by strong cross-tropopause concentration gradients and mixing between the stratosphere and the troposphere. Yet it is near the tropopause where radiative forcing is very sensitive to the greenhouse gas perturbations.

In the framework of the EU-project TRADEOFF we tested a new method for calculating the mass fluxes from ECMWF data. This new method solves the mass-imbalances between the mass transport and the surface pressure tendencies that exist in most global chemistry-transport models. We calculated the mean age of air with this new method and compared it with the observed mean age of air. The improved mass fluxes lead to a better agreement with the observations, although the air in the extra-tropical stratosphere remains too young. In addition we performed a calculation based on ERA40 data. However these results were in poor agreement with the observed mean age of air, hinting towards problems with this data set, as already acknowledged by ECMWF.

Next we investigated the implications of the improved mass fluxes against a large set of observations in the UTLS-region that was compiled within the TRADEOFF project. Besides comparison with NOx we compared model results with ozone and CO observations since the calculation of these trace gas concentrations are especially sensitive to errors in the mass fluxes. Finally, we analysed the implications for the OH budget by comparing model results with the very complete set of the SONEX observations.

Activities of NASA's Global Modeling Initiative (GMI) in the Assessment of Subsonic Aircraft Impact

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The Global Modeling Initiative has now integrated both a tropospheric and stratospheric version of their three-dimensional Chemical Transport Model. The stratospheric version has been used in the past to assess the potential impact of a future fleet of High Speed Civil Transport (HSCT). We utilize the tropospheric version of the model to assess the impact of subsonic aircraft on the chemical composition of the atmosphere. The tropospheric version utilizes prescribed ozone fluxes from the stratosphere by adopting a fixed stratospheric ozone source, constraining the total magnitude of cross tropopause flux. The spatial distribution and seasonality of this flux is determined by the adopted meteorological fields. We use three different sets of meteorological fields, derived from: a) the Goddard Institute for Space Studies GCM, version 2"; b) the Middle Atmosphere Community Climate Model, version 3 (MACCM3); c) assimilated fields from the NASA/GSFC Data Assimilation Office (DAO) for 1997 conditions. Model results for all three fields have been tested by comparison to a suite of tropospheric measurements, including ground-based ozone and CO observations, ozone sonde data, and aircraft data for different species. NOx emissions from subsonic aircraft have been calculated for 1995 conditions. We have used the model to simulate the impact of doubling these emissions on the tropospheric chemical composition. Since model components for all simulations are the same, except those directly related to meteorological fields, the simulations will estimate the variability in the assessment due to differences in meteorological inputs. We will also discuss future plans for GMI aircraft assessment

Parametric Study of Potential Effects of Aircraft Emissions on Stratospheric Ozone

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There has been much consideration over the last decade of the potential impacts on the environment of projected fleets of passenger jets with cruise altitudes in the lower stratosphere. In addition to the fleets of the supersonic High Speed Civil Transport (HSCT) aircraft studied in the 1990s, other aircraft have been under consideration that would also fly extensively in the lower stratosphere, such as the sonic cruiser and supersonic business jets (SSBJs). Existing ozone-impact studies have not fully analyzed the potential extent of possible flight and emissions criteria for such aircraft. In addition, recent improvements to understanding of atmospheric chemical and physical processes would also affect earlier analyses. Scenarios used in many of these parametric studies were developed by Baughcum (2002) as part of a set of "generic" scenarios for assessment calculations to understand the atmospheric sensitivity to cruise altitude, EI(NO_x), and fleet fuel use for possible higher flying subsonic or supersonic aircraft.

In this study, we employ our recently updated state-of-the-art zonally-averaged model of atmospheric chemistry and physics in a series of parametric studies to examine potential effects of emissions from hypothetical fleets of stratospheric-flying aircraft on stratospheric ozone. The new studies examine how the modeling of aircraft effects has changed since the 1999 IPCC assessment (Aviation and the Global Atmosphere). The new studies are also aimed at better understanding of the full envelop of effects on stratospheric ozone from potential aircraft emissions. In addition, a series of aircraft emissions tracer studies are done with both our two-dimensional model and the MOZART three-dimensional model towards defining how different the effects on ozone would be if the newest version of MOZART with a complete representation of stratospheric processes was applied to these analyses.

Stratospheric Ozone Sensitivity to Aircraft Cruise Altitudes and NO_x Emissions

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The stratospheric ozone impact of higher flying aircraft is sensitive to the cruise altitude, NO_x emission levels, and fleet fuel use at cruise altitude. In this study, we present the results obtained using the CSIRO 2-D CTM for a range of parametric aircraft emission scenarios. Reaction rate constants and photolysis cross sections were from the JPL-2000 recommendations. The parametric scenarios were based on aircraft emissions for long range (greater than 2500 nautical mile) missions projected to the year 2020. Cruise altitudes were varied in 2 km increments over the 13 to 21 km altitude range and NO_x levels were varied for NO_x emission indices [EI(NO_x)] of 5, 10, and 20 grams(NO2)/kilogram fuel use.
The column ozone impact was found to depend strongly on cruise altitude, with very little impact for emissions at 13-15 km. Ozone response was found to depend linearly on NO_x levels. The sensitivity of the results to the recommended rate constant compilations (e.g., JPL2000 vs JPL97 or JPL2003) will be discussed.

Investigating the Global Atmosphere by Using Commercial Aircraft: CARIBIC and MOZAIC

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Although compared to the massive plumes of pollution and natural emissions that emanate from the continents, aircraft emissions form a modest contribution, it is in the nature of us sufficiently rapidly booking progress in fully understanding the chemistry and physics of the earth atmosphere, that improved observation capacities are put in place. Principally, no other platform than that commercial aircraft movements do offer, can better trace the emissions from aircraft and the subsequent transport, mixing and chemical transformation although there obviously are limitations as to the actual analytical payloads commercial aircraft can routinely transport.
The German Lufthansa supports various innovative science programs that are designed to gain a more quantitative understanding of a host of atmospheric processes. In the MOZAIC project a modest range of analyzers is flown on a larger number of aircraft. By this, the largest set of in situ measurements of ozone and water vapor has become available to the international community of atmospheric chemists.
In CARIBIC a large range of analyzers and air samplers are carried once to twice each month by a single aircraft, which will be one of Lufthansa's new A340-600ers. The CARIBIC-LUFTHANSA approach offers a powerful package, including a DOAS system, NO and NO_y analyzers, a PTRMS system, several aerosol analyzers, an air sample collection systems for 28 air samples an aerosol collections system, a high precision CO_2 analyzer, a mercury analyzer, ultra fast sensors for CO and O_3, systems for total and gaseous water, etc. The new CARIBIC measurement container can now be well considered as a powerful, compact, automated, flying-laboratory that will be regularly deployed on long distance flights.
The first phase of CARIBIC from 1997 until 2002 with LTU has already yielded a broad range of interesting studies, and several examples of massive pollution plume studies, but also the detection of nitrogen oxides from aircraft, will be presented.

The Importance of Aviation for Tourism: Status and Trends

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Leisure-related travel is an important factor in global travel, accounting for about 50% of all travel in the industrialized countries. On average, daily mobility in industrialized countries is in the order of 40 pkm per day, out of this roughly 20 pkm traveled for leisure. Car travel accounts for 70-75%, air travel for 15-20%, and other means of transport for 5-10% of the total distances traveled. Global energy use associated with leisure-related transport may have amounted to 13,200 PJ in 2001, out of this almost 18% (2,360 PJ) for air travel. It deserves mention that the industrialized countries, which constitute only 15% of the world's population, account for 82% of the global leisure-related transport. These figures highlight the importance of air travel for tourism and the highly skewed distribution of leisure-related travel between industrialized and other countries. In the future, air travel is likely to increase substantially, both as a result of changing leisure conceptions in industrialized countries and the increasing participation of people from developing countries in air travel. There is, however, some uncertainty about the medium-term role of the tourists' risk perception in travel behavior, i.e. the importance of globally spreading diseases such as SARS, terror attacks, and war.

The SCENIC project: presentation and first results.

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The SCENIC Project aims to study the atmospheric impact of possible future fleets of supersonic aircraft. In a unique partnership between relevant European industry representatives and atmospheric scientists, the project will use the most realistic aviation emissions scenarios to date within state-of-the-art numerical models of the atmosphere.
The scientific objective is to address the following important questions: How large are the impacts of a mixed fleet containing high-speed supersonic passenger aircraft on atmospheric composition and climate likely to be? How can we reduce the possible environmental impacts generated by a supersonic fleet?
The first part of the presentation will consist of an overview of the SCENIC-project. The second part will focus on the first results obtained.

A 3D model intercomparison of the effects of future supersonic aircraft on the chemical composition of the stratosphere.

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Gas and aerosol emissions from future supersonic aircraft may affect the chemical composition of the stratosphere in a significant way. The net effect on the global ozone distribution is not easy to assess with photochemical models, due to the complex interactions of different catalytic cycles for ozone destruction in the stratosphere, with partial opposing effects at different altitudes. The large scale accumulation of H2O and NOy from high-flying aircraft is one key point to be assessed with the models. In addition, ozone and water vapour absorb planetary radiation in the middle atmosphere, so that changes in their distribution may feedback on stratospheric dynamics. Here we focus on pure photochemical effects and compare the results of three completely independent three-dimensional chemical-transport models (CTM) (University of L'Aquila, University of Cambridge, University of Oslo), as part of the EC-sponsored TRADEOFF project. It should be noted that the 1999 IPCC assessment was largely based on the results of zonally averaged or two-dimensional models. The University of L'Aquila CTM is run in interactive mode with a microphysics code for aerosol formation and growth, in order to calculate the aircraft forced changes of surface area density (SAD) of sulphuric acid aerosols. This SAD perturbation is then provided off-line to the other two models, in order to

assess the sensitivity of the three CTMs to both NO_x and SO_x emissions. We will first validate the aerosol results in the stratosphere (extinction and surface area density) and then discuss similarities and diffences of the three CTMs in terms of aircraft forced changes of H2O, NOy and O3.

Session 3: Particles and clouds

Particles and Cirrus Clouds (PAZI) - Overview of Results 2000-2003

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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 INCA, PARTEMIS, and PARTS funded by the European Commission in the Fifth Framework Programme. PAZI investigates the interaction of aerosol particles with cirrus clouds, with a strong emphasis on aviation-produced aerosols and contrails, and their impact on atmospheric composition, radiation, clouds, and climate. This overview summarizes several important research results obtained during the first phase of the project (2000-2003).

In particular, the following issues will be highlighted. Measurements and models addressing the formation and evolution of black carbon (BC) particles in burners and jet engines; physico-chemical characterisation 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 addressing aerosol-cirrus interactions; parameterization of cirrus cloud formation; representation of ice supersaturation and cirrus clouds in a climate model and possible climatic impact.

Open research questions, strategic goals, and the organisation of the follow-on project PAZI-2, planned for the period 2004-2008, are briefly described.

Upper tropospheric aerosol formation inside and outside aircraft wakes: new findings from mass spectrometric measurements of gaseous and ionic aerosol precursors and very small aerosols.

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This paper reviews recent mass spectrometric measurements of upper tropospheric (UT) gaseous and ionic aerosol precursors and very small aerosol particles made inside and outside of aircraft wakes. The measured parameters include: a) condensable gases particularly sulfuric acid and their precursor SO_2 b) positive and negative cluster ions c) electrically charged small positive and negative soot particles d) electrically charged positive and negative vary small atmospheric volatile aerosol particles which have diameters <3nm and which therefore cannot be measured by condensation particle counters.

The new measurements suggest that formation and growth of new volatile aerosol particles take place inside aircraft wakes and outside aircraft wakes. Both types of particles, the ones formed by an aircraft and the ones formed in the background UT, experience most of their condensational growth due to the uptake of condensable gases which are photochemically formed in the background UT and whose precursor gases stem from ground level sources some of which are due to man-made activities.

Single Particle Black Carbon Measurements in the UT/LS

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Very few measurements of black carbon (BC) aerosols have been made in the upper troposphere and lower stratosphere (UT/LS), e.g., the limited measurements of Pueschel et al. (1992) and Blake and Kato (1995) are the basis for almost all global estimates of BC loading in this region. In the winter of 2003 a new instrument, the single particle soot photometer (SP2), made measurements of the light absorbing component of single particles from the NASA DC-8.
Measurements were made with the SP2 on seven flights during the SAGE III Ozone Loss and Validation Experiment (SOLVE II) from January 24 - February 6, 2003. The majority of these flights were flown north and west of Kiruna, Sweden (67.8N, 20.3E) at altitudes above 10 Km. The tropopause was usually between 9 and 10 Km, so the measurements were in stratospheric air during most of each flight. The focus of SOLVE II was to study ozone loss in the polar vortex, so the majority of the time of each flight was spent in some region of the vortex. Spiral descents in the vortex were made on several flights so that vertical profiles could be made of the particle and gas species.
The vertical profiles of BC fraction show that the fraction of particles that contain BC vary from 4% to 16% of the total number of particles counted. The fraction maximizes near the tropopause but is relatively constant between 6 and 9 Km. The good correlation between CO and the BC fraction and mass suggests combustion as the likely source of the BC (Baumgardner et al. ,2002). The magnitude of the CO suggests vertical transport from surface combustion sources. Further evaluation with back trajectory analysis and comparison with other tracers is needed to identify the likely sources. Pueschel et al. (1992) estimated that BC represented 0.03% of the total aerosol number concentration in the UT/LS. The results from the present study show BC fractions of 5-20%, i.e. concentrations that are 100-400 times larger than estimated from previous studies. These results indicate that global BC loading may be much higher than previously assumed and further analysis is needed to understand the environmental impact of these new findings.

Ice-nucleating ability of soot particles in UT/LS

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Aircraft–generated soot aerosols are assumed to be the most likely candidates for heterogeneous ice formation of contrails and cirrus clouds. Morphology, microstructure and water adsorbability of laboratory made kerosene soot being an aircraft soot surrogate were studied to establish the correlation between the morphology porosity and the ice nucleation ability of the soot surface.
Quasi-Elastic Neutron Scattering (QENS) and neutron diffraction (ND) have been used to highlight the dynamics and structure of water/ice confined in the porous soot network. The routine of these experiments roughly followed the evolution of temperature (T) and relative humidity (RH) in the plume and down to UT/LS conditions. QENS spectra show a continuous water freezing below the water bulk melting temperature. Decrease of the translational and rotational diffusion coefficients with temperature is attributed to the nucleation of supercooled water in the highly constrained regions of the micro and supermicropores. This effect causes a depression of the homogeneous nucleation point and therefore maintains some water in a highly supercooled state below 204K. ND spectra show a mixture of amorphous ice probably located in the soot pores coexisting with ice Ih at the soot surface.
In the youngest plume, the water molecules adsorb on the primary active centers filling the soot micropores (~ 0.5 nm) where they remain strongly localized. When the plume cools down and RH ~ 70-80 %, the soot supermicropores ~ 2 nm become completely filled. Finally, capillary condensation occurs in the soot mesopores 2 nm as well as multilayer growth on the external surface. At the saturation plume conditions ~ 30% of the water adsorbed on soot transform into ice probably in the soot mesopores 2 nm and on the external surface. But the ice nucleation/growth process is completely suppressed in the soot micro and supermicropores because near 35 % of the water remains liquid under these conditions.
Upon evaporation of the aircraft contrail, new ice forming nuclei containing the soot particles appear in the UT. At T $\cong$ 220K, the existence of 75% ice component inside the soot pores will increase the soot potential to

act as secondary ice nuclei for cirrus clouds. Moreover, ~ 15% of the water confined in the soot supermicropores may remain liquid down to LS temperature ≅ 200K.

Experimental investigation of homogeneous and heterogeneous freezing processes at simulated UTLS conditions

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Ice nucleation (IN) in the UTLS region can occur either by homogeneous freezing of solution droplets below about 240 K, or be heterogeneously induced by so-called ice nuclei. At these low temperatures, homogeneous IN requires high ice supersaturations of up to 60%, which are frequently observed in the upper troposphere. High updraft velocities favour cirrus formation at the homogeneous freezing threshold. At lower updrafts, however, heterogeneous ice nuclei, e.g . aircraft emitted soot particles, may selectively be activated at lower supersaturation. The pristine ice crystals grow by water uptake, thus eventually limiting the maximum supersaturation to values below the homogeneous freezing threshold. This mechanism could explain optically thin cirrus layers with ice particles of low number concentration and large size. The large coolable and evacuable aerosol chamber AIDA (Aerosol Interaction and Dynamics in the Atmosphere) of Forschungszemtrum Karlsruhe is used as a moderate expansion cloud chamber to study processes of ice formation at simulated cirrus conditions like temperature, cooling rate, and ice supersaturation. The freezing onset is detected by measuring the intensity and depolarisation of forward- and back-scattered laser radiation, highly sensitive to the formation of a-spherical ice particles. Until freezing onset, relative humidity is calculated from total water concentration measured with the FISH (fast in situ stratospheric hygrometer) instrument. The ice particle number concentration and size is measured with an optical particle counter. The growing and evaporating ice cyrystals are also characterized by in situ FTIR extinction spectra. In this paper we briefly discuss recent process studies of the formation, growth, and optical properties of ice crystals in relevant aerosol systems, e.g. sulphuric acid droplets and soot particles coated with sulphuric acid and ammonium sulphate layers.

Detailled Modelling of Cirrus Cloud - an intercomparison of different approaches for homogeneous nucleation.

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We developed a cirrus model with detailed microphysics including homogeneous nucleation, deposition on ice crystal and riming of supercooled droplets on ice crystals. The model treats the particle distribution as two-dimensional, which allows to keep the information on the aerosol particles which act as CCN and determine the solute concentration of the droplet.
To get accurately the number of crystals, homogeneous nucleation must be determined very carefully during the short period of cirrus formation. The formation period is limited by the consumption of water vapour by the strong depositional growth of the new small crystals. Therefore, these two processes should be treated closely. There are several approaches for the treatment of homogeneous nucleation. A comparison of those scheme have been published in Lin (JAS, 2002). A major problem in this intercomparison for homogeneous nucleation is that the models involved also use different description on ice particles. Thus this intercomparison does not allow a conclusion on which process „ nucleation or deposition „ is responsible for the different results. That‘s why we‘ve implemented into our model three different schemes of nucleation for supercooled acid sulphuric solution droplets: the classical approach of Tabazadeh (GRL, 2000), the effective temperature model of DeMott (JGR, 1997), and the laboratory data set of Koop (Nature, 2000). With our detailed cirrus model for crystal growth we compared the three different parameterisation of nucleation, and tested the effect of solute concentration on the formation of cirrus clouds.
We‘ve also implemented a riming scheme based on the numerical solution of Bott (JAS, 2000) for two-dimensional particle distributions. We‘ve seen that in pure cirrus conditions riming is negligible to explain ice particle size and also to explain the amount of residual aerosol mass.

Overview of contrail and cirrus cloud measurements from the WB-57 aircraft in the CRYSTAL-FACE mission

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The WB57F aircraft component of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment (CRYSTAL-FACE) mission provided 27 gas and particle sampling instruments for in situ characterization of chemical and microphysical properties of cirrus clouds in the upper troposphere and lower stratosphere. The aircraft collected over 65 hours of science data during the mission, with a substantial fraction of the time spent sampling cirrus clouds. The aircraft also obtained valuable chemical data in contrails, near the tropopause and in the stratosphere up to altitudes of 18 km. The WB57F data set certainly represents the largest and most comprehensive in situ examination of cirrus cloud properties in the 12 km to 15 km region of the atmosphere. This presentation will overview the aircraft payload, flight profiles, and preliminary data. Among the observations to be discussed will be the behavior of nitric acid on cirrus and contrail particles, water vapor concentrations and ice water content in contrails, water isotope variations in and out of clouds, and particle composition of contrail and cirrus cloud particle nuclei.

Simulation of Contrail Coverage over the USA Missed During the Air Traffic Shutdown

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Following the tragic events of 11 September 2001, commercial and personal air traffic was halted for at least 36 hours with resumption of more normal flight activity by 15 September 2001. During the air traffic shutdown, the contrail coverage over the United States of America (USA) decreased dramatically with only a few military jets producing contrails. This lack of contrails over the USA was even noticed by astronauts. Analyses of weather data during the shutdown period indicate an anomaly in the diurnal range of surface air temperature that was attributed to the lack of contrails. Such an anomaly would result from the lack of radiative forcing by contrails and would indicate not only that contrails affect climate but can affect the daily weather. Better quantification of the radiative forcing that would have occurred during normal air traffic during the shutdown requires a simulation of the effects of the missing air traffic. An analysis of satellite data and hourly rapid update cycle (RUC) profiles of temperature and humidity are used to tune a simple model of contrail formation, spreading, and dissipation for several days during September 2001. Normal air traffic is then „flown through the model“ using the conditions observed and analyzed during the shutdown period. Satellite analyses of cloud cover are used to specify areas where natural cloud cover would mask or negate the simulated contrails. The resulting simulated contrails are then used to estimate the radiative forcing as a function of the time of day and compared with the distribution of diurnal temperature range anomalies.

CONUS Contrail Frequency Estimated from RUC and Flight Track Data

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Contrails can affect the global atmospheric radiation budget by increasing planetary albedo and reducing infrared emission. The total amount of the global radiative forcing depends on several poorly known factors including the global mean contrail optical depth, cloud microphysics and the frequency of contrail occurrence. Current theoretical estimates of global contrail coverage are tuned to early estimates of linear contrail coverage determined visually from infrared satellite imagery. The estimates differ based on the parameterization used to diagnose contrails and the numerical weather analyses employed to determine the ambient conditions. Development of reliable methods for diagnosing persistent contrails and their physical

and radiative properties from numerical weather analyses is essential for predicting future contrail climate impacts.
A new estimate of contrail frequency and coverage over the continental United States (CONUS) is developed using hourly meteorological analyses from the Rapid Update Cycle (RUC) numerical weather prediction model and commercial air traffic data from FlyteTrax. The potential persistent contrail frequency over the CONUS is computed directly from RUC analyses using a modified form of the classical Schmidt-Appleman criteria for persistent contrail formation. The potential contrail frequency is adjusted to account for the occurrence of thick cloudiness in possible regions of persistent contrail formation. The air traffic density data is then combined with the potential contrail frequency to estimate the expected contrail coverage. This estimate is compared with previous estimates of contrail coverage, and from a direct satellite estimate of contrail coverage based on an empirical contrail detection algorithm.

Contrail Properties Derived From UARS MLS Measurements

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The goal of this study is to evaluate the fraction of the air traffic in the ice-supersaturated areas (ISA) and to derive contrail coverage using the best available measurements. In contrast to previous studies, which primarily used ECMWF assimilated data, we utilize multi-year Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) measurements of relative humidity above ice (RHI) at altitudes 8-14 km. Applying the MLS RHI measurements during the 1991-1997 period and scheduled air traffic scenarios, the fraction of air traffic flown through ISA was evaluated. Estimates of globally and annually averaged contrail coverage were also calculated.
The sensitivity of our results to aircraft propulsion efficiency and to variations in cruise altitude were evaluated. Uncertainties of our analysis and possible future work were discussed.

Observations of contrails and cirrus over Europe

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Condensation trails (contrails) are now a common feature at the mid latitude skies. Young contrails can be easily identified by their linear appearance, but during the ageing both, the macroscopic structure and the microscopic composition, approach that of natural thin cirrus clouds. Optically thin cirrus clouds and also contrails are known to have a positive climate impact: they are warming the earth/atmosphere system. Up until now only the linear contrails have been considered in studies concerning the climate impact of air traffic. In this work a simplified theory on cirrus coverage due to the spreading of contrails is verified by satellite observations. As expected by this model a significant increase of cirrus coverage of 3% due to air traffic over Europe was found. This is ten times more then the coverage by linear contrails.

Updated perturbations on cirrus and contrail cirrus

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This study presents results from the special workpackage of the TRADEOFF project on clouds and contrails. The work is focused on the detection of contrail cirrus from satellite images and the determination of air traffic contribution. Changes in cirrus coverage and their association to aviation activities are examined and analyzed at congested air corridors of the northern middle latitudes. The analysis is based on the latest version of the International Satellite Cloud Climatology Project D2 data set and covers the period 1984-2001. Over regions of the northern middle latitudes with increased air traffic density from 1992 to 2000, the effect of large-scale modes of natural climate variability such as ENSO, QBO and NAO fluctuations, were first removed from the cloud data set in order to calculate the long-term changes of observed cirrus cloudiness. The results show significant positive trends of cirrus coverage between 1984 and 2001, over the heavy air traffic locations of the northern hemisphere during the summertime. These changes could be related to the change in highflying air traffic density from 1992 to 2000. It is shown that along the latitudinal belt centered at the North Atlantic air corridor, the longitudinal distribution of long-term changes in cirrus cloudiness between 1984 and 2001 is strongly correlated to the longitudinal distribution of changes in fuel consumption from 1992 to 2000 (R=+0.73), providing an independent test of possible impact of aviation on contrail cirrus formation. During winter, the higher inter-annual natural variability in wintertime atmospheric synoptic systems may mask the possible anthropogenic effect on cirrus clouds. These results are compared with other studies and different periods of records and it appears that, as evidenced in this and in earlier studies, there exists general agreement on the anthropogenic effect on high cloud trends.

Potential alteration of ice clouds by aircraft soot

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It has been proposed that aircraft can affect cloudiness by changing the number of aerosols available for ice nucleation at flight altitudes. The impact of aircraft on cloudiness depends on the competition between surface sources of ice nuclei and those from aircraft which depends on both the number of nuclei and the mode of nucleation. We have developed a parameterization that accounts for the effects of aerosol number concentration as well as two different modes of nucleation in the upper troposphere. The parameterization treats homogeneous nucleation by sulfate and immersion nucleation by soot. It has been used to study the effects of sulfate aerosols from surface sources on the initial number concentration of ice particles formed in the upper troposphere. Here, we use concentrations from the IMPACT and GMI aerosol models to examine the relative importance of soot, acting as immersion nuclei, from both surface and aircraft sources in altering the effects of sulfate from homogeneous ice nucleation. The relative effects of sulfate and soot are compared along with a commonly used deposition nucleation parameterization by Meyers et al (1992).

Potential impact of aviation-induced black carbon on cirrus clouds: Global model studies with the ECHAM GCM

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Recently potential impacts of aviation-induced particles on cirrus occurrence frequency and cirrus optical properties have been discussed. Aircraft exhaust particles, especially black carbon (BC) and sulfate aerosols, may perturb the aerosol populations in the upper troposphere and lowermost stratosphere (UTLS) and may act as ice nuclei via homogeneous or heterogeneous ice nucleation occurring at sufficient supersaturations. Recent studies suggest that the impact of aircraft sulfur emissions on cirrus properties via homogeneous freezing of sulfate aerosols is probably small. Hence, the question has been addressed whether aircraft-generated BC particles serving as heterogeneous ice nuclei (IN) may have a significant impact on cirrus cloudiness and cirrus microphysical properties.
In the present study, global simulations on the potential impact of aircraft-generated BC particles on cirrus clouds via heterogeneous nucleation have been perfomed. The general circulation model ECHAM4 is applied including predictions of major aerosol species and cloud condensate. In a first step, the global impact of aircraft activity on the availability of potential heterogeneous IN, such as mineral dust or BC particles, in the UTLS was quantified. The results suggest a significant large-scale contribution of aviation to the heterogeneous IN number concentration. This indicates a potential for aviation-induced BC to impact cirrus cloud formation. Hence, in a second step, potential impacts of aircraft BC particles on cirrus clouds have been simulated. Sensitivity experiments have been performed considering various scenarios of ice nucleating efficiencies of different types of potential heterogeneous IN. The presentation will highlight the potential impacts of BC from aircraft on cirrus properties simulated for the different scenarios. Uncertainties associated with the model predictions will be discussed.

Future Development of Contrail Cover, Optical Depth and Radiative Forcing: Impacts of Increasing Air Traffic and Climate Change

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The future development of linear-shaped contrails is investigated considering changes in air traffic, aircraft technology as well as climate change by means of a contrail parameterization developed for the ECHAM general circulation model. Time slice simulations performed during the EU-project TRADEOFF show an increase in global annual mean contrail cover from 0.06% in 1992 to 0.14% in 2015 and to 0.22% in 2050. In the northern extratropics, the enhancement of contrail cover is mainly determined by the growth of aviation. In the tropics, contrail cover is, additionally, highly affected by climate change. In order to quantify the effect of systematic errors in the model climate on contrail cover, we additionally perform offline diagnostic studies. These studies suggest an underestimation of global contrail cover in the ECHAM simulations by a factor of about 0.8-0.9.
The effect of the bias in the model climate is strongest in tropical latitudes.
The temporal development of the simulated contrail radiative forcing is most closely related to total contrail cover, although the mean optical depth is found to increase in a warmer climate. Our best estimate is an increase of global annual mean radiative forcing from 3.5 mWm^-2 in 1992 to 9.4 mWm^-2 in 2015 and to 14.8 mWm^-2 in 2050. Uncertainties in contrail radiative forcing mainly arise from uncertainties in microphysical and optical properties such as particle shape, particle size, and optical depth.

A studie of contrails in a general circulation model

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The IFSHAM model is used for a study of contrails in a general circulation model. The IFSHAM model is a model based on the dynamical core from the IFS model and the physical parameterization package from the ECHAM model.
The contrail parameterization scheme of Ponater et. al. (JGR 2002) has been implemented in this model in order to be able to study contrail formation. The scheme is based on thermodynamic theory and the principles of the models cloud scheme.
Sensitivity experiments have been performed in order to study the impact of model resolution and the results are compared with the few other model studies that exist and with observed data. Results for global contrail cover, radiative forcing and contrail optical properties are shown.
In order to investigate the importance for contrail formation of the systematic errors of the model, experiments have been performed where the model is nudged towards observed data. Using this technique the systematic errors are substantially reduced and the impact on contrail formation is discussed.

Session 4: Mitigation

On the potential of the cryoplane option to reduce aircraft climate impact

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One technological option to reduce the climate impact of air traffic is a switch to alternative fuels like liquid hydrogen. We have investigated the quantitative potential of such a change, evaluating a scenario that incorporates, both, the expected increase of air traffic between 1990 and 2050, and a technology transition between 2015 and 2050.
The study covers the effects of reduced CO2 emissions, reduced NO_x emissions, and a different contrail radiative impact to be expected from changes in coverage and optical properties. Dedicated experiments with a microphysical process model as well as with a sophisticated climate model have been run to identify key numbers for the specific impact of cryoplane contrails on the climate. A linear response model has then been used to describe the global climate impact. We find a typical value of about 25% radiative forcing reduction from aircraft emissions for the 2050 time slice, if cryoplane were introduced. Best estimates range between 16% and 29%, depending on the speed of the technology transition. Due to inherent scientific uncertainties this range widens to between 14% and 40%.
Some further sources of uncertainty like cirrus cloud changes or possible CO2 emissions from the liquid hydrogen production process have not been included in the current estimate.

Tradeoffs in Contrail and CO_2 Radiative Forcing by Altered Cruise Altitudes

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Within the 5FP EU project TRADEOFF, the impact of revised cruise altitudes was examined in a parametric study. Cruise altitudes were changed by +2,000, -2,000 and –6,000 feet and the consequential CO_2 and NO_x emissions calculated for the global fleet. A new contrail coverage calculation was performed using distance travelled rather than fuel with the GCM ECHAML39(DLR)/CHEM. In addition the radiative forcing from line-

shaped contrails was calculated. The largest signal arose from the –6,000 ft case, which increased CO_2 emissions by a few percent by decreased linear contrail coverage by 43% and radiative forcing by 45%.

Policies for Mitigating Contrail Formation from Aircraft

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One possible approach to mitigating the production of contrails from aircraft is to place restrictions on cruise altitudes based upon ambient atmospheric conditions. Temperature and humidity conditions in the atmosphere are a determinant of contrail formation, which in general is more likely the lower the temperature and the higher the humidity levels. This research examined the ability to restrict cruise altitudes as a policy for reducing contrail formation. A simulation model of European airspace was used to examine seasonal altitude restrictions and the effect on carbon emissions (fuel burn), travel times and air traffic controller workload. Seasonal altitude restrictions were based upon monthly average atmospheric conditions that resulted in winter-time restrictions of 24,000 feet and summer-time restrictions of 31,000 feet. Results showed only a small increase in carbon emissions and travel times but more severe implications for controller workload. Further analyses examined longer haul North Atlantic flights that would need more severe altitude restrictions. This sort of policy was still found to be feasible for some longer haul flights, but would be less effective than for short haul flights. Potential further research and policy implications are discussed.

Greener by Design

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In the coming century, the impact of air travel on the environment will become an increasingly powerful influence on aircraft design. Unless the impact per passenger kilometre can be reduced substantially relative to today's levels, environmental factors will increasingly limit the expansion of air travel and the social benefit that it brings. Of the three main impacts, noise, air pollution around airports and influence on climate change, the third is considered to have the greatest long-term importance. Of the three main contributors to climate change from aircraft - CO_2 emissions, NO_X emissions and the creation of persistent contrails - it is the last two which are the most promising targets. Ways of reducing the impacts of these two are discussed and it is noted that, in each case, the best environmental result is likely to entail some increase in CO_2 emissions. It follows that regulatory or economic measures to reduce impact on climate should be framed so as to do just that. Measures framed purely in terms of CO_2 emissions are likely to be counter-productive. Nevertheless, the design of aircraft to reduce fuel burn and hence CO_2 emission remains a key long-term objective and, in this context, the paper considers the potential offered by new technology and new design concepts.

Poster Presentations

Poster Session 1: Engine Emissions and Plume Processes / Transport and impact on chemical composition

CCN Activation of Jet Engine Combustion Particles During PARTEMIS

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METHODS AND RESULTS

During the EU project PartEmis, the microphysical properties of the particles in the exhaust of a jet engine combustor were investigated for fuels of different sulphur content (FSC 50, 410 and 1280 µg/g). The combustor was operated in two temperature and pressure conditions resembling the average conditions of old and more modern aircraft. In this contribution, the focus is on the ability of exhaust particles to act as cloud condensation nuclei (CCN) at water vapour supersaturations around 0.7%, a value that is slightly higher than the 0.5% usually assumed for stratus cloud formation. CCN were measured with a static thermal diffusion chamber developed at the University of Vienna. The instrument was calibrated both in terms of counting efficiency and supersaturation. As the concentrations of the exhaust particles (i. e. particles 9 nm) were a function of operation condition and FSC, the relative concentration of CCN (i. e. the activation ratio) was used to compare the different conditions. Increasing the FSC from low to mid to high FSC gave an increase of the activation ratio by factors of 0.9 10-3, 1.43 10-3 and 5.15 10-3 (old conditions) and 0.67 10-3, 3.04 10-3 and 7.94 10-3 (modern conditions). The activation behaviour of the exhaust particles was also modelled using Kelvin theory (appropriate for insoluble but wettable spherical particles), Köhler theory (both for completely soluble particles and for insoluble particles with a shell of soluble material) and a semi-empirical model using measured hygroscopicities under subsaturated conditions that also takes some restructuring of agglomerated particles into account. As a first estimate, the soluble material was assumed to be sulphuric acid. The thickness of the soluble shell was estimated from volatility measurements. A comparison of modelled and measured activation behaviour showed the best agreement with the semi-empirical model.

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Gas and Aerosol Chemistry of Commercial Aircraft Emissions Measured in the NASA EXCAVATE Experiment

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An aerosol mass spectrometer (AMS) and tunable infrared laser absorption spectrometer (TILDAS) were deployed to sample emissions from the Langley B757 as part of the NASA EXCAVATE program in January, 2002. Aerosol emissions were sampled at 1, 10, 25 and 35 meters behind the RB211 engine with the AMS, and gases at 1 and 10 meters with the TILDAS. Engine power was varied from idle to near take-off thrust and sulfur loading in the fuel was also varied. The AMS samples submicron aerosol (30 to 1000 nm diameter), aerodynamically sizes particles and detects chemical composition via thermal vaporization and mass spectrometric analysis. This detection approach (which utilizes electron impact ionization) provides real-time, quantitative analysis of volatile aerosol components (e.g. organic carbon and sulfate), though

refractory components such as elemental carbon are not detected. The TILDAS utilizes direct absorption spectroscopy in the near infrared to monitor specific gas phase species (including NO, NO2, HONO and SO2) with high sensitivity and fast time resolution.
The size and mass loading of fine aerosol (30nm diameter) grew as a function of distance behind the engine. Sulfate and organic aerosol were externally mixed in small and large modes (~60 and 200nm aerodynamic diameter, respectively), with organic carbon (OC) loading about 50 times larger than sulfate. The mass spectrum of the OC matches that of lubricant oil samples from the engine while the sulfate appears to be largely pure sulfuric acid. The increase in aerosol loading with distance presumably reflects condensation of vapor species as the plume cools and ages. During transition between low (idle) and high (cruise) engine power settings, dramatic increase in OC aerosol loading (up to a factor of 1000) were observed. These transients have significant implication for airport emissions from aircraft. The AMS results will be discussed in comparison with other (physical) aerosol measurements, including discussion of possible inlet effects, particularly directly behind the engine.
TILDAS measurements focused on quantifying HONO EIs as the RB211engine was varied over the EXCAVATE operating range. HONO is produced in the post combustor flow path, driven by available OH radical levels as the temperature falls in the turbine and tailpipe. HONO is a sensitive measure of post combustor oxidative processes and its concentrations are expected to depend on the temperature history through the engine hot sections, which will vary as the engine operating conditions vary. Clear variation with engine operation was observed, with HONO concentration increasing to over 2ppmv at the highest power setting.

Sulfur (VI) in the simulated internal flow of an aircraft gas turbine engine: first measurements during the PartEmis project

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Gaseous S(VI) (SO3+H2SO4) has been measured for the first time by chemical ionization mass spectrometry (CIMS) in the simulated internal flow of an aircraft gas turbine in a test rig at ground level during the PartEmis 2002 campaign. Building on S(VI) and the calculated total sulfur St the abundance ratio e=S(VI)/St was determined. Measurements were made in the three pressure stages of the rig (Low, Intermediate and High pressure stages) for two engine test conditions, representative of old and modern aircraft cruise. For FSC = 1270 ppm, old cruise condition and at the LP stage an e = 1.4±0.7 % was obtained. For the modern cruise condition, which corresponds to higher combustor exit pressure, temperature and higher fuel flow in comparison with the old cruise, an e = 2.3±1.2 % was obtained. Our results suggest an increase of e with combustor exit temperature, pressure and fuel flow. This e is consistent with previous direct measurements of e in the exhaust plumes of aircraft gas turbine engines which build on direct S(VI) measurements by MPI-K Heidelberg using CIMS. However our present e is much smaller than some previous reported indirectly inferred from measurements of aerosol volatility, SO2 and impactors. The present values support the view of relatively small e in the few per cent range. Our findings have important implications for volatile aerosol formation and soot particle activation in aircraft wakes and their role in contrail and cloud formation.

Emission of Volatile and Non-Volatile Ultrafine Particles from a Combustion Source During PARTEMIS

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The formation of volatile nanoparticles from gaseous precursors in the cooling exhaust gas of combustion sources is a well-known phenomenon. In particular the emission of condensation particles by aviation at cruise is discussed as a possible source for sulphuric acid particles in the upper troposphere. The size of these nucleating particles depends strongly on the relative humidity of the atmosphere, but remains in

general below 20 nm in diameter. Furthermore, the occurrence of non-volatile nanoparticles of diameter 5 nm is reported for soot forming flames. These non-volatile nanoparticles are regarded as precursor of the combustion aerosol particles (soot). Both types of nanoparticles were observed in the exhaust of a jet engine combustor. The effect of combustor operation conditions and fuels sulphur content (FSC) on both types of nanoparticles is investigated using experimental methods and modelling studies.
The measurement of the nanoparticle mode (D < 20 nm) was performed in the diluted exhaust gas. Size resolution in the scale D < 20 nm was achieved by operating a multi-channel Condensation Particle Size Analyzer CPSA. The CPSA provides number concentrations in the size bins D = 4-7 nm, 7-9 nm, 9-20 nm, and >20 nm. The size distribution in the combustion aerosol size range (D 10 nm) was measured with a Scanning Mobility Particle Sizer (SMPS). The mixing state of the total aerosol was measured at sizes D = 15nm, 30 nm, 50 nm, and 80 nm by using a Differential Mobility Analyzer combined with a thermodenuder system.
At low and medium FSC, particles of the smallest size class occur by a factor of about 20 less frequently like combustion aerosol particles, while at high FSC they are up to 7 times more frequent than combustion aerosol particles. In contrast, the occurrence of particles in size bins 7 -9 nm and 9 - 20 nm is almost independent of the FSC. From a volatility analysis of the sub-20-nm fraction it is concluded that volatile condensation particles are composed of sulphuric acid while non-volatile nanoparticles most likely consist of carbonaceous material.

Kinetics of Binary Nucleation in Aircraft Exhaust Plume

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Civil aviation releases various components that can affect natural atmospheric processes. In particularly, the sulfur and water vapours emitted by engines may be converted to liquid and ice aerosol particles that may act as cloud condensation nucleus. The key crucial point in this process is an initial phase of nucleation of numerous new particles in an exhaust plume during its cooling and expansion in the ambient atmosphere. In this article, the kinetics of non–steady nucleation and time lag for binary homogeneous nucleation of sulfuric acid-water aerosols is considered in a comparison with the usually used classical steady-state nucleation theory. Classical nucleation analysis assumes: (1) that the timescale for establishing a steady-state sub-critical clusters population is very short compared a change of the nucleation rate which in turn out depends on the temperature and gas species concentrations change (steady-state clusters population approach); (2) that the concentration of monomers is much higher than the concentration of sub-critical clusters, so cluster-cluster collisions are negligible compared to monomer-cluster collisions (monomer-cluster collision approach); (3) that the nucleating system is near the equilibrium and the concentration of monomers required to establish the steady-state clusters population is much higher than the total concentration of monomers incorporated in clusters (monomers reservoir approach). The purpose of this paper is to investigate these assumptions for a practically important case of the binary nucleation during the aircraft exhaust plume cooling. For this, a model which directly consider the dynamics of clusters population (birth-death equations including monomers) with accounting for the cluster-cluster collisions and cluster dissociation into two smaller clusters together with the cooling of gaseous molecule-cluster system is proposed. It is shown that for many typical conditions the assumptions leading to the classical nucleation rate are invalid. For example, (i) may be important collisions of two sub-critical clusters, which result in the formation of „critical“ or even of larger size cluster; (ii) in the aircraft exhaust plume there is a relatively slow sub-critical clusters build-up compared to the rate of plume expansion and cooling. Also, the comparison between different models describing the energetic of first clusters formation, have shown that considered nucleating process of neutral clusters may be limited by the initial „nucleation steps“ (i.e. the formation of dimer, trimer etc.).

A USA Commercial Flight Track Database for Upper Tropospheric Aircraft Emission Studies

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Prediction of the atmospheric effects of air traffic on the atmosphere require a realistic representation of the density and timing of flights at different altitudes. Simulations of air traffic typically involve the use of fuel use data to represent flight duration at particular levels. Such datasets have been valuable but are limited in information and have not been updated for many years. This paper describes a new database of upper tropospheric commercial flights over the contiguous United States of America (USA). It is currently available and being continuously updated with new data on a daily basis. Commercial flight information taken in real time over the USA from the FlyteTrax system developed by FlyteComm, Inc. has been archived at NASA Langley research Center since September 2000. The raw data consist of 2-, 5-, or 10-minute reports of flight number, aircraft type, time, latitude, longitude, altitude, heading, destination and origination locations, speed, and departure and arrival times. All reported portions of flights above 25,000 ft (7.6 km) within the domain bounded by 20°N - 50°N and 60°W - 135°W are quality controlled after sorting the data by flight number and time. Flights remaining after passing the quality control checks are then used to develop the database, which is divided into two parts: linear and gridded. The former computes the node points for each flight track on 1° latitude-longitude grid using interpolation along great circle arcs between each report. These standardized flight track positions comprise the linear database in the form of one file for each flight along with a header describing the general flight characteristics. The gridded database uses the standardized flight tracks to determine for each hour the number and total length of flights within a 1-km vertical range in a given 1° grid box. The linear dataset should be useful for detailed simulation studies, while the gridded data should be more valuable for use in climate simulations. Statistics on the flight lengths, vertical distribution, and temporal variability at various scales will be presented.

Interaction of NO and ice crystals produced from combustion generated warer vapor in a simulated jet engine exhaust gas plume

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This paper describes the results of a preliminary experimental study on the NO-ice crystals interaction in a simulated jet engine exhaust plume. The formation of ice crystal (snow) by condensation of water vapor in the exhaust gas simulates the formation of contrail. The experiments were conducted in the coldest season at Rikubetu in Hokkaido, Japan. Exhaust gas was prepared by a kerosene-fueled small regenerative combustor. It was pre-cooled in a convection-cooled tube and injected into co-axially flowing atmospheric air of temperatures from -20 to -30 C in a vertically positioned 30-cm diameter duct of 3 m in length. Two funs were installed at the inlet or the exit of the duct to feed the atmospheric air and the reference velocity was changed stepwise by choosing single or dual operation. The NO concentrations in the exhaust gas were varied from 250 to 500 ppm mainly by increasing fuel-air ratio and by a change in the axial fuel nozzle position in the burner while maintaining fuel flow rate. The NO2 concentration was negligible. The sulfur content in the kerosene used was 0.003 wt%. The residence times in the duct were estimated as 2 ms for single fun operation and 4 ms for dual fun operation. The exhaust gas was diluted 30 and 60 times at the exit. Ice crystals were collected at the exit of the duct and the concentrations of NO2-1, NO3-1 and SO4-2 ions in the samples were determined by ion chromatography.
The pH of the samples was around 6.45, being independent of operating conditions. The maximums of NO2-1and NO3-2 ion concentrations measured in the present experiment were 0.5 and 2.5ƒÊg /ml, respectively though the effects of NO concentration in the exhaust on these ion concentrations was not clear since the dependency changed depending on the air flow in the duct. The measured maximum of SO4-2 was about 10ƒÊg /ml. The variation of SO4-2□@with NO concentration in the exhaust is similar to that of NO3-2.

Validation of the Kinetic Soot Model: An Experimental and Theoretical Study on Soot Formation using LII and Shifted Vibrational CARS

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The reduction of pollutants from aeroengines is an important challenge for the design of new combustion systems. Increasing efforts aim at studying the processes contributing to soot formation and oxidation. There are two main approaches towards a comprehensive understanding of these reactions: experimental determination of physical properties in sooting flames and theoretical modelling of the underlying chemical processes. For the soot model development the validation by experimental data in simplified combustion systems is necessary. An extensive pool of validation data that contains different equivalence ratios, pressures and fuels is desirable. The precise temperature determination is equally important for the model validation since temperature has a strong influence on the gas phase soot precursor chemistry.
For this validation, well defined experimental boundary conditions of the flame under study are necessary. Our new burner design permits the separation of soot growth and oxidation by preventing the entrainment of secondary air into the sooting region of the flame. The investigated flame is surrounded by a non-sooting methane/air coflame which acts as a hot gas shield against secondary air.
Experimental results are presented for laminar premixed ethene/air and propene/air flames at equivalence ratios between 2 and 3 and for pressures up to 5 bar. Soot concentrations are measured by 2D-Laser-Induced Incandescence (LII) using 1064 nm excitation. Calibration of the LII signal is obtained by 532 nm extinction measurements using the same optical pathway.
Application of conventional vibrational N_2 CARS in sooting flames fails since under sooting conditions the C_2 Swan band at 473 nm interferes with the N_2 signal spectra. We modified the conventional excitation scheme by using a narrowband dye laser instead of the Nd:YAG laser‘s pump wavelength at 532 nm, thus shifting the CARS spectrum out of the interference region. Therefore, temperatures from sooting flames are now accessible with high precision using shifted vibrational CARS spectroscopy (SV-CARS).
The kinetic calculation contains two steps: a DLR-modified gas phase model applying detailed chemistry and a postprocessing soot module. The experimental information is used to support these computations in two respects. First, the real temperature information from the experiment is used for the calculation, thus including effective energy loss by radiation. Second, the predicted soot formation at given temperatures can be compared to the experimental soot volume fraction profiles. Comparison of theory and experiment permits further refinement of the kinetic soot formation model.

Jet Engine Combustion Particle Hygroscopicity under Subsaturated Conditions During PARTEMIS

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METHODS AND RESULTS
Hygroscopic properties of jet engine combustion particles were investigated within the EU-PartEmis project. Focal points were the influence of fuel sulphur content (FSC), engine operating condition and of the turbine section on the particle properties. A jet engine combustor was operated on a test-rig at QinetiQ, Farnborough, UK. The turbine section was simulated by a three-stage heat exchanger. Two different engine operating conditions (old and modern cruise), and three different FSCs (~50, ~400, and ~1300 µg S / g fuel; low, mid, and high FSC, respectively) were investigated. Hygroscopic growth factors (HGF = D(RH)/Do, D = diameter) of dry Do = 30, 50 and 100 nm particles at relative humidity RH = 95% were measured using a hygroscopicity tandem differential mobility analyser.
The combustion particles were not hygroscopic at low FSC, but HGFs increased distinctly with increasing FSC, i.e. HGFs (RH = 95%, Do = 50 nm, modern cruise, combustor exit) were 1.01, 1.10, and 1.16 at low, mid, and high FSC, respectively. This increase of hygroscopicity is attributed to a sulphuric acid coating of

increasing thickness. Generally the engine operating conditions had no significant effect on the hygroscopicity, only at mid FSC were HGFs somewhat higher under modern cruise conditions. The turbine section had little effect on HGFs at low and mid FSC, i.e. HGFs (RH = 90%, Do = 50 nm, modern cruise, mid FSC) were 1.06 and 1.07 at the combustor exit and after the turbine section, respectively. However, corresponding HGFs at high FSC were 1.09 and ~1.18, respectively, indicating an increase of particle hygroscopicity through the turbine section. Under identical conditions HGFs also depended on the initial particle size, small particles were more hygroscopic than larger particles.

ACKNOWLEDGEMENTS
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AvioMEET Inventory Tool and its Applications

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Increasing numbers of flights and still unknown effects of exhaust gases on the high atmosphere have drawn most attention on air traffic and its emissions. In Europe, many institutions are working in this area, collect traffic and emission data, create emission inventories and assess effects. That lead to some work done in parallel while using different databases and methodologies which often lead to results that cannot be compared or matched.
COST 319 action and MEET project were a starting point for a dialogue and discussions between the different communities involved and thus gave an incentive for harmonisation. MEET project came up with a methodology for estimating air pollutant emissions from present and future air traffic. Methodology and emission indices are now used for strategic environmental assessment and transport policy making.
The COST 319 working group D2 – air traffic – has proposed minimum requirements for an harmonised approach to generate emission indices. This seems to be the only way make results from different inventories comparable and exchangeable. Harmonisation work is going on under the umbrella of the Thematic Network AERONET. Its subgroup on Harmonisation of Emission Inventories and Modelling is aiming at comparing existing data sets and defining needs and an interface to atmospheric and climate modelling.
Methodology used in MEET project and presented here is based on a flexible design that allows to adjust it to the user requirements as well as on air traffic data and emission factors (easily) available.
Based on the MEET methodology an MS-Access computer tool was created, called AvioMEET, which uses most of the Emission Indices published in the Emission Index Sheets of MEET/Deliverable 18.
TRENDS finally uses all this more or less theoretical knowledge to apply it on existing traffic activity data to come up with a database of environmental indication for air transport.

Air Parcel Trajectories in the South-European UTLS: Implications for the Impact of Air Traffic Emissions

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This poster reports on meridional and vertical transport in the region of the tropopause during the APE-INFRA 2002 and Geophysica-ENVISAT satellite test and validation campaigns from Forli, Italy during July and October 2002. The *Geophysica* high-altitude research plane (July, October) and the DLR Falcon (October) were used during these campaigns and the flight paths are designed to converge with the footprint of the ENVISAT satellite. This poster discusses back trajectory modelling analysis of air parcels from a specified grid (34-48^{o}N and 0-22^{o}E) and over a vertical domain of isentropic surfaces from 300K to 500K. Data from in situ instruments measuring aerosol number, ozone, water vapour, NO_y and NO_x etc are analysed.
5-day reverse domain-filling (RDF) trajectory studies are presented, illustrating - on regional scales - the origin of air parcels each day for one month using data from the EMCWF (ERA-40 re-analysis). Initial

analysis has focused on meridional, zonal and vertical transport over 5 days. Along with July and October 2002, ten-years climatology (1992-2001) have also been studied, for comparison and to build a climatology of the region. Back trajectory modelling has shown that a number of air parcels have both descended from the lower stratosphere into the emissions zone (UTLS); while a smaller number have risen through the mid-tropospheric layer to the upper troposphere. Rapid cross-isentropic transport/dispersion is reported in the UTLS. This rapid vertical transport is unsurprising in the troposphere, but the cause of rapid vertical transport in the lower stratosphere is still under investigation.
Since we would expect air from high latitudes to be chemically different to air originating in the sub-tropics in the lower stratosphere, we discuss how the relative abundance of high and low latitude air in the region of air traffic emissions will influence the likely impact of these emissions.
Initial results from in-situ instruments aboard both the Geophysica and Falcon aircraft for the "NERC" flight of the 17/10/2002 show distinct signals of aged aircraft exhaust plumes. This signal is particularly prominent in many chemical species such as: Ozone, water vapour, NO, NO_y; along 0.3-20μm aerosol size distributions and condensation nuclei (CN) number concentrations. Further investigation of these aircraft contrails is currently being carried out.

The impact of aircraft on the chemical composition of the atmosphere and options for reducing the impact. A 3D CTM model study.

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In the framework of the EU project TRADEOFF, a 3-D global chemical transport model driven by ECMWF meteorological data is used to calculate the impact of NOx emissions from aircraft on the chemical composition of the atmosphere in the year 2000. The model applies two comprehensive numerical schemes for tropospheric and stratospheric chemistry, respectively, and calculates advective transport based on the accurate Second Order Moments scheme. The vertical resolution is better than 1 km in the tropopause region.
In our model calculations we use a set of different emission scenarios, which were developed by QinetiQ in the TRADEOFF project. The focus of this study is on the aircraft impact and its sensitivity to flight altitude and flight routing (polar routes). For comparison, we calculate the aircraft impact for an emission scenario provided by NASA, which was used in the IPCC report on aviation and the global Atmosphere.
We investigate geographical and temporal variations in the impact on ozone, reactive nitrogen (NOy), NOx (NO+NO2), OH, and the lifetime of CH4. Finally a calculation of aircraft impact in the year 2050 using emissions estimated by NASA is presented, illustrating the effect of a changing background atmosphere and the increase in NOx emissions from aircraft. In this context the non-linearity of the ozone response due to NOx emissions is clearly revealed.

Modelling the Impact of Subsonic Aircraft Emissions on Ozone

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The impact of aircraft NOx emissions on ozone in the UTLS region has been studied within the framework of the TRADEOFF project. Model integrations have been performed using TOMCAT, a 3-dimensional tropospheric chemistry transport model. Perturbations to aircraft emissions have been incorporated by changing the cruise altitude and flight routing of the present-day subsonic fleet. The effects of tropospheric gas-phase chemistry on ozone mixing ratios and tropospheric ozone column have been investigated. A selection of results from the TRADEOFF project will be presented here.

Uptake of Nitric Acid in Cirrus Clouds

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Cirrus clouds have attracted increasing attention in recent years, in particular because of their role in the radiative forcing of climate, indirect aerosol forcing as well as their relevance for the chemistry of upper tropospheric ozone. One possible mechanism important for atmospheric chemistry and trace gas distribution is the denitrification of the tropopause region by sedimenting cirrus ice particles. However, up to now the question on the partitioning of nitric acid in a cirrus cloud situation including the efficiency of nitric acid scavenging by ice particles is not satisfactorily answered.
From a synopsis of field, laboratory and model studies at T205K as well as from the field experiments Polstar at T<205K we derive a general picture of the partitioning of nitric acid (HNO3) in cirrus clouds and a new hypothesis on the uptake of HNO3 on ice particles:
A substantial part of nitric acid remains in the gas phase under cirrus cloud conditions. The HNO3 removed from the gas phase is distributed between interstitial aerosol and ice particles in dependence on ice surface area and temperature, respectively. In cold cirrus clouds with small ice surface areas (T<205K) the partitioning is strongly in favour of interstitial ternary solution particles while in warmer cirrus clouds with large ice surface areas the uptake on ice dominates. Consequently, denitrification via sedimenting ice particles may occur only in the -more frequently occurring- warm cirrus clouds.
The HNO3 coverage on ice is found to be different for ice particles and ice films. On ice films the coverage can increase with decreasing temperature from about 0.1 to 0.8 monolayer, while that on ice particles is found to decrease with temperature and the partial pressure of HNO3 from 0.1 to 0.001 monolayer. An HNO3 uptake behaviour following dissociative Langmuir isotherms where the coverage decreases for descending temperatures may explain the observations for ice particles.

Radiative Forcing on Climate from Aircraft Emissions in the Stratosphere

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The 1999 Intergovernmental Panel on Climate Change report on Aviation and the Global atmosphere estimated that emissions from a fleet of one thousand High Speed Civil Transport aircraft (flying at Mach 2.4) could produce a non-negligible impact on the radiative forcing driving changes in climate. The radiative forcing for this fleet was +0.1 Wm-2, with +0.10 Wm-2 coming from the increase in stratospheric water vapor, along with smaller contribution from increased CO2 (+0.01 Wm-2) and from effects on stratospheric ozone (-0.01 Wm-2). In this study, we reexamine the radiative forcing from fleets of aircraft flying at stratospheric altitudes. We use our narrowband radiative transfer model in these studies, along with model calculations of calculated changes in ozone and water vapor from our zonally-averaged model of atmospheric chemical and physical processes. The radiative transfer model used here has higher resolution in the tropopause and lower stratosphere region than the models used in the IPCC assessment. Preliminary results suggest that the radiative forcing for the water vapor emissions from aircraft was overestimated in the IPCC assessment. Along with reconsideration of the radiative forcing for the HSCT scenarios used in IPCC, we also consider the radiative forcing from more realistic fleets of possible stratospheric-flying aircraft.

Sources of NO_x at cruise altitudes; Implications for predictions of ozone and methane perturbations due to NO_x emissions from aircraft.

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NO_x emissions from aviation in the upper troposphere and lower stratosphere cause radiative forcing of climate through perturbations in concentrations of ozone and the lifetime of methane. Assessments using global chemical tracer models (CTMs) have shown significant differences between the estimated impacts (e.g. IPCC, 1999). Due to the non-linear nature of the photochemistry of the atmosphere, the impact of additional NO_x from aircraft is very dependent on the background concentrations of NOx. In this region of the atmosphere NO_x can originate from many sources, mainly from lightning, convective transport of surface emissions, downward transport from the stratosphere (from N2O oxidation), and from aircraft emissions. To improve our understanding of the possible environmental impacts of NO_x emissions from aircraft, it is of key importance to be able to simulate the background NO_x chemistry (total concentrations and contributions from the different sources).

Within the EU-project TRADEOFF we have performed an analysis of the contributions to the background NO_x levels at cruise altitude from different sources in 5 global CTMs. The results show that although the total concentrations of NO_x can be well represented, there are significant differences in the contribution from the different sources. Implications for predictions of current as well as future ozone and methane lifetime perturbations from aircraft will be discussed.

Postersession 2: Particles and Clouds / Mitigation

Aerosol properties measured in situ in the free troposphere and tropopause region at midlatitudes

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In the past few years the DLR Falcon 20, a German twin-jet research aircraft with a maximum ceiling of 13 km, has participated in a number of experiments devoted to the characterization of aerosol properties in the troposphere and the tropopause region. Total aerosol number concentrations for Aitken mode and ultrafine particles have been measured with condensation particle counters with different lower cut-off diameters in the range from 3 to 15 nm. For a subset of data, the fractionation between volatile, semi-volatile and refractory particles was determined. Total concentration of accumulation mode particles as well as aerosol size distributions were determined from measurements of a combination of optical aerosol spectrometer probes (PMS PCASP-100X and FSSP-300). In this contribution we report on mean tropospheric vertical profiles of aerosol properties and the statistics of aerosol concentration and size distributions in the upper troposphere for different campaigns mainly conducted in Europe but differing in location (marine/continental) and expected contribution of anthropogenic pollution.

Hygroscopicity and wetting of aircraft engine soot and its surrogates: CCN formation in UT

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The potential importance of aircraft–generated soot particles to contrail and cirrus formation in the upper troposphere (UT) are explored in field and modeling studies. But ice nucleating ability of exhaust soot is still poorly known because an unambiguous evidence that soot particles are directly involved in ice formation is difficult to obtain from in situ measurements. To improve this situation, a typical aircraft engine combustor burning aviation kerosene under cruise conditions was used to generate engine soot and characterize its properties, especially those responsible for cloud condensation nucleus (CCN) formation. Aircraft combustor soot produced by the sulfur-free fuel burning and laboratory-made kerosene flame soots were studied as surrogates for atmospheric black carbon (BC) aerosols.

This work examines the soot wetting and hydration properties to determine the possible pathways of CCN formation in the UT. The surface microstructure and chemical nature have a tremendous influence on the soot wetting properties. Engine soot is found to consist of the mixture of the graphite flackes and amorphous particles of the low surface area. Measurements of the water/ice contact angle, θ, on the soot surfaces show a range from 50 to 80 degrees. The best wettability is obtained for engine soot, $\theta \approx 50^0$, which is related to a significant amount of water soluble fraction (WSF), $\geq$4.4 wt%, and volatile compounds $\approx$ 17%. The main part of WSF is connected with sulfate whose distinctive features are observed in FTIR spectra. Engine soot demonstrates a surpriingly high hygroscopicity , with about of 20 adsorbed water monolayer at 240K. It will act as contrail condensation nuclei at the small warter supersaturations in the plume.

Combustor and kerosene soots give $\theta \approx 63^0$ and 80^0, they are belived to represent the insoluble BC particles in the UT. The amount of adsorbed water on their surfaces is around one monolayer that corresponds to carbonaceous surface of intermediate polarity. Following the inverse Kelvin effect the soot aglomerated structure is suggested to amplify the heterogeneous nucleation process due to water condensation into the interparticle cavities between the soot primary particles. This pathway is of major importance for ice nucleation on the insoluble carbonaceous particles in the UT. It provides an estimation of the critical ice supersaturations, S_c, needed for the particle growth. Combustor and kerosene soots S_c are found near 6.2% and 17.6% at T=220K, respectevely. Data of S_c are examined to determine which wetting characteristics of BC particles are required for the cirrus cloud formation in the ice - saturated regions of the UT.

Ice Water Content of Cirrus Clouds and its Dependency on different Types of Aerosols

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The aerosol impact on cloud microphysics is important because of its link to the forcing of climate. There are a few investigations concerning clouds consisting of liquid water, but the knowledge on ice clouds remains still poor. Model descriptions of the ice particle formation in cirrus clouds need a reliable parameterisation of the partitioning of the available water into the gas, liquid (aerosol) and solid phase. Until now, experimental data providing direct access to the gas phase and the ice water content with high accuracy are not available. We conducted laboratory measurements of the homogeneous and heterogeneous ice nucleation and the partitioning of the water at UT cirrus cloud conditions dependent on different temperatures and types of aerosol particles in the large coolable and evacuable aerosol chamber AIDA of IMK-AAF.
The ice supersaturation necessary for the formation of ice particles was achieved by quasi adiabatic volume expansion by controlled pumping. The onset of freezing was detected by measuring the depolarisation of scattered laser light with high sensitivity and time resolution. The Lyman-alpha- fluorescence hygrometer (FISH) of FZJ, ICG-I was used to measure the total water (gas phase + condensed phase). Simultaneously the gas phase water concentration was directly measured in situ by absorption at 1370nm with the tuneable diode laser (TDL) of University of Heidelberg and IMK-AAF. Further information about the ice phase like number concentration and size distribution of the formed ice particles was obtained both from in-situ multi-path FTIR extinction spectroscopy and an optical particle counter. With these instruments we are able to measure the partitioning of the water during ice nucleation experiments.
We will show results for the development of the ice water content after the onset of freezing until the cloud formation process has finished. The ice nucleation experiments presented here were conducted for freezing temperatures below 235 K. Different types of aerosols were used as ice nuclei: pure mineral dust; soot, coated with sulphuric acid and with ammonium sulphate, respectively; and solution droplets of sulphuric acid and of ammonium sulphate, respectively.

3D simulation of cirrus formation from airplane contrails

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An Eulerian microphysical 3D-cirrus cloud model (MPC) is developed. The model is based on a detailed microphysical description of both liquid and solid phase cloud particle size distributions. It includes nucleation, melting, condensation, evaporation and sedimentation processes. In the present contribution, MPC is used to simulate different scenarios with occurrences of contrails.

Heterogeneous nucleation effects on cirrus cloud coverage

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Current aircraft engines release a lot of aerosol into the atmosphere even when no contrails are formed. These aerosol particles may eventually become involved in heterogeneous cirrus formation processes, at ice-supersaturations lower than those needed for homogeneous nucleation. Therefore
it is generally believed that this so-called indirect effect leads to higher cirrus cloud coverage on the average,

compared to a hypothetical case with no aerosol emissions from aircraft (for example a fleet of LH2 driven airplanes only). However, this view is too simple. There are competing effects that must be considered in a complete assessment of the indirect effect. First, cirrus formed heterogeneously is probably optically thinner than homogeneously formed cirrus, because crystal numbers are less, and maximum supersaturation during the nucleation event is less. Second, after a heterogeneous cloud has formed the supersaturation is used up for a while, and a homogeneous cloud will not form whereas it would have been formed without the indirect effect. Third, lifetimes of heterogeneously formed clouds may differ systematically from those formed homogeneously. Results obtained with the ECHAM model (involving new parametrisations for cirrus coverage from heterogeneous and homogeneous processes) will be shown as examples for these possible effects. Unfortunately, there is considerable uncertainty in many of the parameters involved. Thus an assessment of the climatic role of the indirect effect does not seem to be in reach currently.

Contrail Coverage over the USA Derived From MODIS and AVHRR Data

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Contrails often lead to the development of additional cirrus clouds that can affect climate via the radiation budget. Evaluation of contrail coverage and optical properties is crucial for assessing the impact of current and future climatic effects of air traffic. Current estimates of contrail coverage over the United States of America (USA) have been based on a single NOAA-16 afternoon overpass time for recent studies and at four times of day for 1993-94 data from two satellites with different sensitivities and detection errors. Approximately 14,000 flights cross portions of the USA each day at different times of day. The commercial flight activity begins in earnest around 0600 LT and continues with high intensity before fading shortly before local midnight. Because spreading contrail lifetimes are generally less than 4-6 hours, then the atmosphere should be cleansed of most contrail coverage by the beginning of the next day. Assuming that the state of the upper troposphere is, on average, the same during the day, this daily cycle should be reflected in the contrail properties and coverage. However, preliminary studies using NOAA-15 morning overpasses suggest that the afternoon analyses may underestimate the contrail coverage because the spreading and saturation of contrails formed during the morning in areas of heavy air traffic might mask or diminish the contrails formed during the afternoon. To obtain a better assessment of the diurnal variation in contrail coverage, this study analyzes data taken over the USA from a series of satellites beginning with the NOAA-12 in the early morning period followed by NOAA-15, NOAA-17, Terra, Aqua, and NOAA-16 during the afternoon. Contrail coverage and optical properties are derived from multispectral data from the Advanced Very High Resolution Radiometer (AVHRR) on the NOAA satellites and from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra and Aqua satellites. Different sensitivities in the instruments are first compared to ensure that any derived diurnal variations are not due to sensor artifacts. Preliminary results from all of the satellites are presented showing the daily variation in contrails. Results of a more extensive analysis using only the NOAA-15 and NOAA-16 data are also shown.

Contrail Coverage over the North Pacific From MODIS and AVHRR Data

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Cirrus cloud cover has been increasing over the North Pacific since the 1970's. Although part of the increase may be due to a rise in relative humidity, some of the change is likely caused by contrails forming and spreading as a result of transoceanic air traffic. Analysis of high-resolution satellite data is required to determine the contribution by linear contrails to that increase. The Advanced Very High Resolution Radiometer (AVHRR) has been taking 1-km multispectral data from the NOAA satellites since the 1980's, but most of the archived and real-time data over the broad ocean areas are for the 4-km Global Area Coverage dataset. The NASA Earth Observing System satellites, Terra and Aqua, have been operating since March 2000 and August 2002, respectively. Each carries the Moderate Resolution Imaging Spectroradiometer (MODIS) that takes and archives multispectral data globally at resolutions from 0.25 to 1 km. To quantify the contrail properties over the North Pacific, an automated algorithm is applied to 1-km

MODIS data from Terra and Aqua and to selected sets of AVHRR data to derive contrail areal coverage, optical depth, ice particle size, and radiative forcing. The derived properties are compared to similar quantities derived from data over the continental United States of America to examine the differences between contrails formed over marine and continental areas. Preliminary statistics and comparisons are presented.

Survey of Cirrus properties from Satellite retrievals using TOVS and AVHRR observations

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Since 1979, the TOVS instruments aboard the NOAA Polar Orbiting Environmental Satellites have measured radiation emitted and scattered from different levels of the atmosphere, and therefore are an important tool for a continuous survey of the state of the atmosphere over the whole globe. The TOVS Path-B dataset provides atmospheric temperature profiles and water vapor profiles as well as cloud and surface properties at a spatial resolution of 1° latitude x 1° longitude. At present, 8 years of TOVS Path-B data (1987 - 1995) are available at LMD. Due to their relatively high spectral resolution, IR vertical sounders are especially useful for the identification of cirrus clouds (day and night). Cloud-top pressure and effective IR cloud emissivity are computed from the CO2 absorption band radiances by a weighted c2 method. Mean effective ice crystal sizes (De) and ice water path (IWP) of large-scale semi-transparent cirrus clouds are retrieved by taking advantage of the fact that spectral cirrus emissivity differences between 11 and 8 mm depend on this parameter. This method is sensitive to sizes up to 80 mm. This cirrus dataset, covering the NOAA-10 observation period (1987-1991), has been produced within the framework of the European project CIRAMOSA .
In addition the data of the AVHRR instrument (with a spatial resolutuion of 1 km) aboard the NOAA satellites have been used to analyse the temporal evolution of ice and water cloud parameters in a twelve years period from 1990 to 2001 over Europe. The underlying method is the AVHRR Processing scheme Over Land, cLouds and Ocean (APOLLO).
Cloud parameters in selected regions and time periods derived from data of both these instruments, i.e. the TOVS and the AVHRR instrument, will be presented and discussed. Special interest is directed towards studying the impact of increasing air traffic on cirrus properties and climate.

Comparison of cirrus cloud properties in the northern and southern hemisphere on the basis of lidar measurements.

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Cirrus cloud measurements have been performed during the INCA field campaigns in Punta Arenas/Chile (53.12°S, 70.88°W) and in Prestwick /Scotland (55.51°N, 4.60°W) in each hemisphere's fall in the year 2000. Additional measurements are currently performed at the Meteorological Observatory Lindenberg (MOL). From lidar backscatter profiles at 532 nm and 355 nm the optical depth (OD) of the clouds is retrieved as well as base and top altitude of the clouds and the phase of the particles.
One difference observed between the northern and southern hemisphere is the occurrence of very faint layers of particles in an altitude range of 5 to 8 km which were seen only in the North. However, for the cirrus itself no difference has been detected as far as the frequency of occurence (dt) of thin or subvisible clouds is concerned. In both campaigns about 35 % of all cirrus were subvisible (OD<0.03) and about the same fraction of thin cirrus was detected (0.03<OD<0.3).
Differences in the results from the southern and the northern hemisphere are found in the wavelength dependence of the backscatter coefficient and the depolarization behaviour. These results suggest, that there are clouds consisting of rather large particles in the South (Punta Arenas), which have not been detected in the North (Prestwick). A detailed analysis of these data requires a non-spherical scattering theory which is difficult to conduct and currently under investigation.

In summary we can state that our data suggest that the higher concentration of aerosol (including anthropogenic aerosol) in the northern hemisphere does not have an impact on the abundance of cirrus, including those in the subvisible range under the prevailing meteorological conditions of the campaigns. However, aerosols seem to have an important influence on the microphysical properties of high tropospheric clouds.

A Fast Stratospheric Aerosol Microphysical Model (SAMM)

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A fast stratospheric aerosol microphysical model (SAMM) has been developed to study the impact of aircraft imissions on the atmosphere. SAMM simulates homogeneous heteromolecular nucleation, condensational growth, coagulation and sedimentation of binary sulphuric acid-water particles to predict the composition and size-distribution of stratospheric aerosols. SAMM has been successfully applied to estimate the changes in background stratospheric aerosol surface area due to aircraft sulphur emission. The principal advantage of SAMM is that it is non-iterative, i.e. computing time is minimised by finding semi-implicit solutions to aerosol processes. In SAMM homogeneous nucleation and condensation is coupled so that there is a realistic competition between the two processes for the limited amount of vapour. With geometrically related size bin (44 bins for sulphuric acid-water particles in the range from 0.3 nm to 5 μm) and a 600 s time-step the model takes about half an hour to complete a 7 year simulation of stratospheric background aerosols on a 1.4 GHz workstation. SAMM's simulations of background stratospheric aerosols and volcanically disturbed aerosol compare favourably with results from earlier model studies and observed data.

Climate Responses of Aviation NO_x and CO_2 Emissions Scenarios

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A simple linear climate response model for CO_2 and O_3 (Sausen and Schumann, 2000) was used to explore the impacts of potential improvements in NO_x technology in a new simplified emission scenario. The new emission scenario, based upon industry projections, shows that that the impact of aircraft on climate may grow at a slightly lesser rate than was indicated by the IPCC aviation report. The climate impacts of CO_2 and NO_x emissions were compared and it was found that the comparative impacts of CO_2 and O_3 strongly depended upon the equilibrium temperature response of climate to ozone forcing and it was concluded that this was the greatest source of uncertainty in the model resutls. Climate response models are usually 'tuned' to the response of a parent GCM. In this study, we also examined the response of the simple model by tuning it to more generalized responses of GCMs, taken from the IPCC Third Assessment Report. It was shown that whilst the model response differed quantitatively, qualitatively, it did not. This indicates that the ozone response to NO_x aircraft emissions has a larger effect on climate than CO_2, than would be indicated by simple radiative forcings.