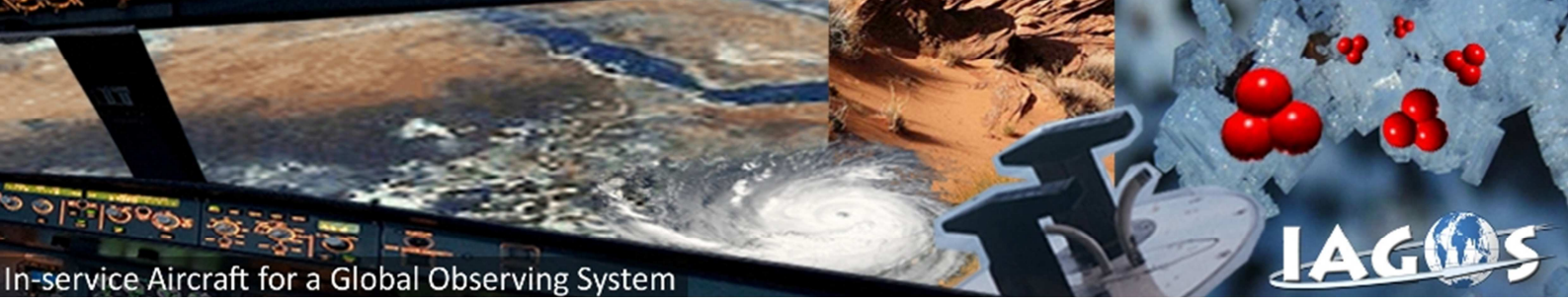

MOZAIC-IAGOS Scientific symposium on atmospheric composition observation by commercial aircraft
Toulouse France 13-15 May 2014

Book of abstracts

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1 Evaluation/validation of satellites and surface remote sensing

1.0 - Use of IAGOS data for verifying remote sensing observations and evaluating atmospheric chemistry model results (keynote)

Peter van Velthoven[1]

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Ankie Piters[1], J. de Laat[1], B. Barret[2], C.A.M. Brenninkmeijer[3], K.-P. Heue[3], A. Zahn[4], C. Gerbig[5]

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Presenter : Peter van Velthoven

The IAGOS-core (formerly MOZAIC) and the IAGOS-CARIBIC observations have in the last decade been used extensively for evaluation of remote sensing measurements and atmospheric chemistry models. We present a number of highlights to illustrate the promise that the IAGOS data hold for the future, e.g. for evaluation of COPERNICUS atmosphere services. For example, IAGOS data have been used to validate SCIAMACHY, MOPITT and IASI carbon monoxide, IASI ozone and GOME-2 SO2 and BrO observations, as well as FTS surface observations of carbon dioxide and methane. We will present a few methods for remote sensing evaluation and discuss caveats. The quasi-horizontal flight legs of IAGOS aircraft in the UTLS have proven value for evaluating vertical transport processes in atmospheric chemistry models. The variety of measurements collected on board the IAGOS CARIBIC aircraft have allowed evaluation of the simulation of many different species by atmospheric chemistry models, including those of ozone, carbon monoxide, methane and acetone.

1.1 - Validation of Aura MLS stratospheric water vapor measurements by the NOAA frost point hygrometer

Dale Hurst

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Presenter : Dale Hurst

Differences between stratospheric water vapor measurements by NOAA frost point hygrometers (FPHs) and the Aura Microwave Limb Sounder (MLS) over Boulder, Colorado, Hilo, Hawaii, and Lauder, New Zealand are evaluated for the period August 2004 through December 2012. Only those MLS profiles spatially and temporally coincident with monthly FPH soundings at each site are evaluated. FPH profiles are convolved with the MLS averaging kernels for 8 pressure levels from 100 to 26 hPa (~16 to 25 km) to reduce their vertical resolution to that of the MLS water vapor retrievals. From 100 to 26 hPa the mean FPH–MLS differences are well within the combined measurement uncertainties of the two instruments. However, the mean differences at 100 and 83 hPa over these sites are statistically significant, ranging from –0.46±0.22 ppmv (–10±5%) to –0.10±0.05 ppmv (–2±1%). From 68 to 26 hPa the FPH–MLS differences average 0.8% (0.04 ppmv). For each site the time series of differences at each pressure level are examined for trends using weighted linear regression analyses. The vast majority of these trends are not statistically significant and most are smaller than the minimum detectable trends. Except at 100 and 83 hPa the average agreement between MLS retrievals and FPH measurements of stratospheric water vapor is better than 1%.

1.2 – Summertime tropospheric ozone assessment over the Mediterranean region using the thermal infrared IASI/MetOp sounder and the WRF-Chem model

Sarah Safieddine

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A. Boynard, P.-F. Coheur, D. Hurtmans, G. Pfister, B. Quennehen, J. Thomas, J.-C. Raut, K. S. Law, Z. Klimont, J. Hadji-Lazaro, M. George1 and C. Clerbaux

Presenter : Sarah Safieddine

Over the Mediterranean region, elevated tropospheric ozone (O3) values are recorded, especially in summer. We use the Infrared Atmospheric Sounding Interferometer (IASI) and the Weather Research and Forecasting Model with Chemistry (WRF-Chem), to understand and interpret the factors and emission sources responsible for the high O3 concentrations observed in the Mediterranean troposphere. Six years of IASI data have been analyzed and show consistent maxima during summer, with an increase of up to 22% in the [0-8] km O3 column in the eastern part of the basin compared to the middle of the basin. MOZAIC-IAGOS data over the same IASI period will be investigated. We use 2010 as an example year to investigate the processes that contribute to these summer maxima. Using two modeled O3 tracers (inflow to the model domain and local anthropogenic emissions), we show that between the surface and 2 km, O3 is mostly formed from anthropogenic emissions and above 4 km, is mostly transported from outside the domain. Evidence of stratosphere to troposphere exchanges (STE) in the eastern part of the basin is shown, and correspond with low relative humidity and high potential vorticity.

1.3 - Consistency of measurement of tropospheric ozone by different platforms and techniques in the global databases

Hiroshi Tanimoto

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Regina Zbinden (Meteo France)

Presenter : Hiroshi Tanimoto

A number of tropospheric ozone observations are conducted all over the world by different platforms and techniques for different purposes and goals. These observations are used to derive and discuss seasonal cycles, interannual variations, and long-term trends of tropospheric ozone. The datasets are also compared to chemistry-transport models to evaluate the performance of the models and hence to test our current understanding of the tropospheric ozone. Here we evaluated consistency of tropospheric ozone observations in multiple platforms available in the global databases: WDCGG for surface, WOUDC for sonde, and MOZAIC/ IAGOS for aircraft observations. We also examined potential discrepancy between sensor types for sondes: ECC, Brewer-Mast (at Hohenpeissenberg), and Carbon Iodine (in Japan, until 2009). Concomitant observations are examined on hourly basis (except for daily basis for sonde-vs.-aircraft comparison) for a pair of sites whose distance is less than 100 km. We found good correlative behaviors, with regression lines being close to 1:1 lines for the comparison of sonde to surface observations at 6 sites, while its correlation is tighter by ECC sensor than by CI. The correlations found for the 6 aircraft-vs.-surface comparison are poorer than for the sonde-vs.-surface comparisons, indicating that insufficient

representativeness of the sites and/or the impact of local production at the airports. The 8 comparisons of sonde to aircraft datasets show mean biases of 5 ppbv at all altitude levels up to 9 km. While the overall difference is within 10 ppbv among surface, sonde, and aircraft at Hohenpeissenberg, the sonde-vs.-surface difference can be more than 20 ppbv in summer (June-September) at Tsukuba, with its magnitude depending on year, suggesting that special care is needed in using sonde data below 3 km at Tsukuba. These results suggest that local time and sampling frequency do matter in the analysis of seasonal cycles, long-term trends, and interannual variations of tropospheric ozone.

1.4 - Role of dynamics and biomass burning on CO and O3 over Asia: MOZAIC vs. Models

Varun Sheel

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Lokesh Sahu, Physical Research Laboratory, Ahmedabad, India
M. Kajino, Meteorological Research Institute, Tsukuba, Japan
Valerie Thouret, Laboratorie d'A'erologie, CNRS, Toulose, France

Presenter : Varun Sheel

Anthropogenic emissions in Asia are increasing due to rapid urbanisation and industrial growth. Trace gases like CO and O3 emitted in the Asian region are susceptible for long rang transport and are known to affect the remote regions of the atmosphere.

We study the seasonality in the vertical distribution of CO and O3 over Asia using the MOZAIC aircraft data and chemistry transport models. The objective of this study is to investigate the effect of biomass burning and long range transport, on the variability of these trace gases.

While the PBL CO is predominantly influenced by strong winds, bringing regional background air from marine and biomass burning regions, under calm conditions CO levels are elevated by local emissions. Back trajectories and fire count map indicate the role of long-range transport and regional biomass burning on the lower tropospheric O3. On the other hand, in the free troposphere, seasonal variation reflects the impact of long-range transport associated with the ITCZ and biomass burning. The inter-annual variations were mainly due to transition from El Nino to La Nina conditions.

1.5 - Analysis of tropospheric ozone and carbon monoxide profiles over South America based on MOZAIC/IAGOS database and model simulations

Marcia Yamasoe

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Bastien Sauvage, Valerie Thouret, Philippe Nedelec

Presenter : Marcia Yamasoe

In this study, we analyzed ozone and carbon monoxide profiles measured, during landing and taking off over Caracas, in Venezuela, and Rio de Janeiro and São Paulo, in Brazil, by commercial aircrafts from the MOZAIC/IAGOS fleet. The analyzed database include years from 1994 to 2009 over Caracas, from 1994 to 2005 over São Paulo, from 1994 to 2004 and 2012 over Rio de Janeiro. In terms of ozone concentrations, results showed clean atmosphere over Caracas with seasonal mean partial ozone column, integrated from the surface up to 200 hPa, of less than 25 DU, presenting the highest seasonal mean in March, April and May. During this season, carbon monoxide concentration reached values as high as 400 ppbv below 750 hPa, particularly in the years 2003 and 2007. Backward trajectories analyses with FLEXPART, of case studies for which the measured concentrations were high, showed that local sources were the main contributors to the observed loadings. Fire pixels from MODIS indicated local biomass burning activities as the possible source, since the period corresponds to the peak of the dry season above the equator, once the ITCZ is further south. Sensitivity studies performed with the chemical transport model GEOS-Chem version 9-01-01 could not capture the influence of biomass burning, what could be due to an underestimation of GFED v2, the emission inventory used in this study. Over São Paulo and Rio de Janeiro, the highest ozone concentration was observed during September to November, with mean partial ozone column, integrated from the surface up to 200 hPa, reaching about 30 DU in 2012. Lightning and biomass burning from South America and local urban pollutions were the main sources of ozone precursors with no contribution of long range transport from Africa. Although no trend analysis could be performed due to the rather irregular frequency of flights over this area, results indicated a possible tendency of increasing concentrations in the recent years at 95% significance level

1.6 - Monitoring and Estimating of NOx emission from space using WRF_Chem Model for Air quality study over south Asia

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Sachin D. Ghude, G.G. Pfister

Presenter : Chinmay Kumar Jena

Accurate NOx emissions are essential for air pollution quantification and mitigation. Combustion processes in the transportation, industrial, and residential sector and emissions from power plants are the dominant anthropogenic sources of NOx. In addition to contributing to the formation of ground-level ozone and fine particle pollution, high levels of NOx are linked with a number of adverse effects on the respiratory system. In this work, we map and develop for the first time an independent satellite constrained NOx emission inventory for India for 2005 on seasonal scale using an inverse technique and iterative procedure. The study combined OMI tropospheric NO2 column retrievals over the Indian region with tropospheric NO2 columns simulated by the WRF-Chem model using the INTEX-B emission. The study uses the WRF-Chem model covering a domain of south Asia at 0.5°×0.5° horizontal resolution to simulate tropospheric NO2 columns. The model uses MOZART-4 gas phase chemistry linked to the GOCART aerosol scheme. For consistency with satellite retrievals, model output at each day is interpolated in time and space to locations of valid satellite retrievals and convolved with the averaging kernels used in the DOMINO retrievals. By determining the local relationship between modeled emissions and tropospheric columns and iteratively applying this relationship to OMI observations, an optimized NOx emission inventory was derived for each season of 2005. Locally and regionally differences, however, can be high. The top-down inventory captures many of the missing hotspots in the original inventory. We further compared the OMI tropospheric columns NO2 with model simulated tropospheric column NO2 using EDGARv4.2, RAS, MACCity, SAFAR and showed significant differences in spatial distribution of NO2. Evaluating the effect of the INTEX-B, EDGARv4.2, RAS, MACCity, SAFAR and top-down inventory on surface ozone clearly indicates significant changes in the spatial distribution. Model results are compared with satellite retrievals of tropospheric column amounts of carbon monoxide (CO) from MOPITT and nitrogen dioxide (NO2) from OMI. While there are still remaining uncertainties in the top-down estimate, this method provides a potential basis for mapping and evaluating robust independent NOx emissions and trends, particularly for regions with large uncertainties such as India, and provides important information to air quality modelers and policy makers.

2 Long-range transport of air pollutants

2.0 – Hemispheric Ozone : Current understanding and future directions (Keynote)

Kathy Law

LATMOS-IPSL

2.1 - Source regions of elevated carbon monoxide in the northwestern Pacific upper troposphere

Kuo-Ying Wang

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Philippe Nedelec, Jean-Pierre Cammas, Valerie Thouret, Andreas Volz-Thomas, Andreas Wahner, Andreas Petzold

Presenter : Kuo-Ying Wang

Asia Pacific regions are main regions for the release of anthropogenic pollutants from sources at the surface to the global atmosphere. However, routine in-situ measurements of these pollutants to show transport pathways of these anthropogenic pollutants in the troposphere are rare. Most of current understanding of the spatial and temporal distribution of these surface pollutants relies on satellite remote sensing and ground based monitoring stations. In this work, we show profiles of routine CO measurements from the Pacific Greenhouse Gases Measurement (PGGM) air-based measurement project over the Pacific regions since June 2012. These CO measurements were made by using the In-Service Aircraft for a Global Observing System (IAGOS) Package 1 installed on a routine in-service commercial Airbus A340-300 passenger aircraft operated by China Airlines. The PGGM IAGOS data set collected during the July 2012-February 2013 period contains more than 500 profiles of CO measurements from surface to 10 km altitude during the descending and ascending phases of the flights. In addition, CO measurements were conducted over the Pacific in the upper troposphere at aircraft cruise altitude. The sources of elevated and low CO in the Pacific upper troposphere and in the lower troposphere at 2-km altitude measured by the PGGM IAGOS flights were determined by using the HYSPLIT model. Our model results consistently show that the Asian continent is a main source for elevated CO measured by the IAGOS instrument in the upper troposphere over the Pacific regions, while low CO in the upper troposphere contains airs that are not associated with the Asian surface sources. Together, the IAGOS data and the HYSPLIT model show distinctive transport pathways between the polluted air and the clean air over the northwestern Pacific region.

2.2 - Impact of Eurasian biomass burning emissions on the springtime lower-tropospheric ozone in North China and the rest of Northeast Asia

Hongyu Liu

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Jiayue Huang, James Crawford, Valerie Thouret, Jose Rodriguez, Susan Strahan, Megan Damon, Stephen Steenrod, and Sarah Strode

Presenter : Hongyu Liu

Intensive biomass burning activities take place in Eurasia from April through September, severely degrading regional air quality. We examine the impact of Eurasian biomass burning on the springtime (April-May) lower-tropospheric (LT) ozone over Northeast Asia in the Global Modeling Initiative (GMI) chemistry and transport model driven by the Modern Era Retrospective-Analysis for Research and Applications (MERRA) meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO). We evaluate model simulated ozone against the multi-year (1995-2012) aircraft ozone profiles over Beijing from the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program and ozonesonde measurements at Japanese stations obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC). The model-simulated large-scale temporal and vertical variability in ozone is similar to that in the MOZAIC observations, but the model often underestimates the magnitude of the observed ozone enhancements in the LT during April-May. By conducting model sensitivity simulations, we quantify and contrast the impacts of Eurasian biomass burning on the springtime LT ozone over Northeast Asia in the years with and without active biomass burning activities, respectively. Extremely high Eurasian biomass burning emissions in combination with stronger northward transport of Chinese anthropogenic emissions resulted in very large ozone enhancements in the LT over Northeast Asia in May 2003. We find that the impact of Eurasian biomass burning emissions on the springtime LT ozone in Beijing can be comparable to, or even higher than that of Chinese anthropogenic emissions. These results have important implications for predicting the air quality of the North China Plain as well as understanding the interannual variability of springtime tropospheric ozone in Northeast Asia.

2.3 - YAK-AEROSIB: continental scale aircraft measurements of trace gases gases above Siberia

Jean Daniel Paris

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Presenter : Jean Daniel

Despite the unique scientific value of better knowing atmospheric composition over Siberia, regional observa
tropospheric composition over this region are still lacking. Large local anthropogenic emissions, strong ec

exchange across the vast forest expanse, and processes feeding back to global climate such as wildfire events, w
emissions, seabed hydrates destabilization and degrading permafrost make this region particularly crucial to
Anthropogenic emissions at the regional scale are also an increasing pressure that is poorly known.
We aim at addressing this need in the YAK-AEROSIB program [1,2] by collecting high-precision in-situ measure
vertical distribution of CO2, CH4, CO, O3, black carbon and ultrafine particles distribution in the Siberian troposphe
other parameters including aerosol lidar profiles, on a pan-Siberian aircraft transect. Campaigns are performed ann
regular route, while special campaigns are occasionnally arranged to sample the troposphere elsewere (e.g. R
coast).
We show the background tropospheric composition obtained from these surveys, the variability and the impact o
transport of anthropogenic emissions from Europe and Asia, as well as the impact of biomass burning plumes bo
wildfires (2012) and from remote sources elsewhere in Asia. Long range transport of anthropogenic emissions is sh
a discernible impact on O3 distribution, although its lower-tropopsheric variability is largely driven by surface d
Regional sources and sinks drive the lower troposphere CO2 and CH4 concentrations. Recent efforts a
understanding the respective role of CH4 emission processes in driving its large scale atmospheric variability over t
Generally, the YAK AEROSIB provide unique observations over Siberia, documenting both direct impact of regi
and aged air masses experiencing long range transport toward the hig

[1] J.-D.Paris, et al. 2010, B. Am. Meteorol. Soc., 91 (5), 625-641.
[2] Antokhin, P. N. et al. 2012, J. Atmos. Oceanic Technol., 29(1), 64-75.
[3] A. Berchet, et al. 2013, Tellus B 2013, 65, 19688.

2.4 - Lightning NOx influence on large scale NOy and O3 plumes observed over the northern mid-latitudes

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Presenter : Alicia Gressent

This work is a climatology of NOy plumes originating from lightning emissions using 4 years (2001-2005)
measurements in the upper troposphere of the northern mid-latitudes, together with ground- and space-based ob
lightning flashes and clouds. The study investigates the influence of lightning NOx (LNOx) emissions on large s
2000 km) plumes (LSPs) of NOy and O3. 127 LSPs (6% of the total MOZAIC NOy dataset) have been attribu
emissions. Most of the lightning-related LSPs were recorded over North America and the Atlantic mainly in spring
during the maximum lightning activity occurrence. The majority of the LSPs (74%) is related to warm conveyor bel
tropical cyclones originating from North America and entering the intercontinental transport pathway between N
and Europe, leading to a negative (positive) west to east NOy (O3) zonal gradient with -0.4 (+18) ppb difference
and -0.6 (+14) ppb difference in summer. Such transatlantic LSPs may have a potential impact on the European p
rest of the LSPs are related to mesoscale convection over Western Europe and the Mediterranean Sea (18%) a
convection (8%).

2.5 - Variability of tropospheric ozone over an urban site in India: A study based on MOZAIC and CCM vertical profiles over Hyderabad

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Presenter : Lokesh Sahu

The Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) data of O3 over Hyderabad in India during years 2006-2008 have been analyzed. Vertical profiles of O3 show seasonal variation with higher and lower values during pre-monsoon and monsoon seasons, respectively. In lower troposphere, monthly O3 was highest of 54 ppbv in January and lowest of 18 ppbv in August. Back trajectories and fire count map indicate the roles of long-range transport and regional biomass burning. The lower levels of O3 in monsoon season were due to flow of marine air and negligible local biomass burning, while higher levels in other seasons were due to transport of continental air. During monsoon and post-monsoon seasons, lower levels of O3 in upper troposphere were associated deep convection. In upper troposphere, levels of O3 in the pre-monsoon of 2006 were higher by about 30 ppbv compared to same season of year 2008. Higher and lower levels were associated with strong wind shear in the year 2006 and weak shear in the year 2008. The year-to-year variations were mainly due to transition from El Niño to La Niña conditions. The vertical variations of O3 and lapse rate were anti-correlated in the troposphere. The lower O3 levels were observed in the stable layers, but higher O3 in the mid-troposphere were caused by the long-range transport. In the PBL region, the mixing ratio of O3 shows strong dependencies on meteorological parameters. The CCM2 reasonably reproduced the observed profiles of O3 except for pre-monsoon season.

2.6 - The Indian Summer Monsoon: Investigating Pollution Import, Chemical Processing, Mixing and Subsequent Export, Based on Measurements by the IAGOS-CARIBIC Observatory in the Upper Tropospheric Anticyclone

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Presenter : Carl Brenninkmeijer

The CARIBIC Observatory was deployed to survey the Indian Summer Monsoon using Lufthansa flights to Chennai in India. Based on a fully fledged (and fully automated) flying analytical laboratory including a remote sensing instrument and air sample and aerosol particle sample collection systems, all integrated in an air freight container (gross mass 1.5 ton), aerosol and over 100 trace gases could be assayed. The monthly CARIBIC flight tracks (2 passages from Frankfurt, Germany to Chennai, India, and back) crossing the western lower edge of the oblong upper tropospheric large anticyclone (over up to 3000 km) give a consistent picture of the chemical evolution and demise of this important tropical/subtropical meteorological phenomenon. The conclusions for greenhouse gas emissions from South Asia and a study of the sources

of methane have been published. Likewise an estimate on the emissions of methyl chloride from South Asia was published. We now present further progress using chemical tracers observed by the CARIBIC system and meteorological analyses to obtain a more complete picture of the mixture of trace gases inside the Indian Summer Anticyclone, the chemical age of the air masses, photochemical tendencies, as well as the source regions and export pathways of the air masses characterized by the accurate and extensive CARIBIC measurements.

2.7 - Ozone and Carbon Monoxide Climatologies from the Trajectory Mapping of Global MOZAIC-IAGOS Data

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Three-dimensional (i.e. latitude, longitude, altitude) gridded climatologies of ozone and carbon monoxide have been developed by trajectory mapping of global MOZAIC-IAGOS data. Ozone and carbon monoxide measurements made during ascent and descent, comprising nearly 58,000 profiles at more than 150 airports worldwide from 1994 to 2012 are used. Forward and backward trajectories are calculated from meteorological reanalysis data in order to map ozone and carbon monoxide measurements to other locations, and so to fill in the spatial domain. This domain-filling technique employs over 700,000 calculated trajectories to map otherwise sparse MOZAIC-IAGOS data into a quasi-global field. This is possible because the respective lifetimes of both ozone and carbon monoxide in the troposphere are generally of the order of weeks.

These quasi-global climatology datasets facilitate visualization and comparison of different years and seasons, and offer insight into the global variation and trends of ozone and carbon monoxide. The results are archived monthly for decades from 1994-2012 on a grid of $5^{o}\times5^{o}\times1$ km (latitude, longitude, and altitude), from the surface to 13 km altitude. They are also archived seasonally, monthly and yearly from 1994 to 2012. Major regional features of the global ozone and carbon monoxide distributions are clearly evident in the maps. The ozone climatology shows excellent agreement with a similar product derived from ozonesonde data. Comparison of the CO climatology with MOPITT data will also be presented.

2.8 - MOZAIC/IAGOS airborne measurements to evaluate CO emission inventory

Fabio Boschetti

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Huilin Chen, Julia Marshall, Philippe Nedelec, Valerie Thouret, Christoph Gerbig

Presenter : Fabio Boschetti

In recent year a new stream of data has become available for atmospheric investigations: airborne measurements from commercial airliners.

The main advantage of using airborne platforms for sensors is their ability to collect meteorological and mole fraction measurements at different height covering most of the troposphere. However, mainly due to the cost of rental aircraft, the number of flights is usually quite limited, with direct consequences on measurement availability. The MOZAIC/IAGOS program address this issue making use of commercial airliners to provides CO mole fraction measurements on a regular basis.

In this study MOZAIC/IAGOS measurements have been used together with a Lagrangian particle dispersion model (STILT) to evaluate the representativeness of the EDGAR emission inventory (version-4.2 , horizontal resolution: 10 km) at 3 different locations (Frankfurt, Paris and Vienna) for the time frame 2004-2011.

Key concepts in this study are:

a) Mixed layer: part of the atmosphere where signals resulting from CO emissions are reasonable well mixed, and lead to an enhancement of CO within the mixed layer.

b) Free troposphere: surface placed two km above the mixing height (top of the mixed layer)

We investigate the CO enhancement relative to values in the free troposphere for both the upper and lower half of the mixed layer. Our main hypothesis is that the CO enhancement in the upper half of the mixed layer is more representative for larger spatial scales, as the vertical distance to the surface and its emission sources is increased compared to that of the lower half. CO enhancements calculated from observed vertical profiles are compared with the corresponding values from CO mole fractions modeled by the emission inventory and coupled with the transport model. Modeled global CO fields from the MACC project will be used in this study as lateral boundary conditions; the domain of the transport model is approximately coincident with the territory of the EU.

We found that simulated results tend to significantly underestimate the measured CO mole fractions, indicating a low bias of about a factor of two in the inventory-reported CO emissions. In addition, the lower half of the mixed layer usually shows a smaller bias between the observed and modeled CO than does the upper half of the mixed layer. Potential reasons for the discrepancy between simulations and observations will be assessed.

3 Recent and new technical developments

3.0 - MOZAIC-IAGOS Technical history over 20 Years (Keynote)

Philippe Nédelec

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From an Airbus initiative to better understand the aircraft impact on the atmosphere, the Mozaic project has been started in 1992, with the objective of installing atmospheric monitoring systems on commercial Airbus operated by major European airlines as Lufthansa and Air France. In addition to the scientific interest of the measurements, this technical instrumental development had to face to major constraints related to the hard conditions for routine airborne operations : aircraft environment, aeronautic certification and instruments reliability for unattended operation up to 12 months.

The initial objective was to measure Ozone (by CNRS) and water vapor (by FZ Julich). Developed with a strong collaboration with Airbus, Mozaic systems have been certified by Airbus and installed on 5 new Airbus A340, in Airbus final assembly line. Initially planned for 2 years and as the project proved its operational capacity and its strong scientific interest, it was continued over years. From 2001, 2 new instruments have been developed : CO analyzer by CNRS, installed on the 5 Mozaic aircraft, and a complex NOy analyzer by FZ Julich installed on 1 aircraft only. In 2014, the 2 last Mozaic equipped aircraft are to be retired from service after 20 years of operation...

As Mozaic was a technical and scientific success, the continuation project IAGOS was started in 2005, with the objectives of equipping other aircraft (A340 and A330) with a new designed EASA certified system, with optimized compacted instrumentation and enhanced measurements capabilities. A basic system has been realized for O3, CO (by CNRS), H2O (by FZ Julich) and Clouds (by University of Manchester). In April 2014, 5 IAGOS equipped aircraft are operational with the basic systems, which includes provision for installation of optional instruments (NOx, NOy, Aerosols, CO2/CH4, SO2), presently under final certification process.

3.0 - Improving and enhancing the IAGOS data stream: recent developments within the IGAS project

Christoph Gerbig,

MPI-BGC
Julia Marshall, and the IGAS Team

IGAS Team:
Karl Beswick (University of Manchester), Damien Boulanger (CNRS - Laboratoired'Aérologie),
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Marc Pontaud (CNRM), Armin Rauthe-Schöch (MPI for Chemistry), Hans Schlager (DLR),
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The project IGAS (IAGOS for the GMES Atmospheric Service) was launched in 2013 with the main aim to better link and enhance data streams of atmospheric measurements from commercial airliners provided by IAGOS (In-service Aircraft for a Global Observing System; www.iagos.org) to scientific users and to the Copernicus Atmosphere Service (formerly known as GMES Atmosphere Service).

A major objective is to increase the accessibility of the data through the development and implementation of database tools, and by ensuring that the central IAGOS database is fully interoperable with the DLR flight campaign archive and the Copernicus data archive in Jülich, from which all data will be accessible via the Jülich OGC Web Services Interface (JOIN). This also involves making IAGOS-CARIBIC data available at the central IAGOS database. The application of IAGOS aerosol and GHG measurements in the Copernicus Atmosphere Service and for satellite validation will be developed and assessed. Also near-real-time (NRT) transmission of IAGOS-core data is under development. Furthermore, a major effort is undertaken to evaluate and harmonize the quality of the measurements, ensuring full documentation and traceability to WMO reference standards and conformance with WMO measurement guidelines. For this, a regular documentation of QA/QC efforts is involving internal and external consistency assessments as well as reviews by external experts. Finally, IGAS seeks to enhance IAGOS measurement capabilities through targeted instrument development for the measurement of aerosol light extinction, VOCs, speciated cloud water/ice/volcanic ash particles, and water vapour. The presentation will introduce the project and will present results obtained within the project so far.

3.1 - Latest developments for the IAGOS database

Damien Boulanger

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Presenter :

The IAGOS central database in Toulouse, that also includes the former MOZAIC database, is an essential part of the global atmospheric monitoring network. Data access is handled by open access policy based on the submission of research requests which are reviewed by the PIs. Users can access the data through the web sites: http://www.iagos.fr and http://www.pole-ether.fr as the database is part of the French atmospheric chemistry data centre ETHER (CNES and CNRS).

The IAGOS information system is in continuous development and improvement. New tools are being developed in order to facilitate and accelerate data validation. The web interface is being enhanced by implementing added-value web services for data visualisation such as maps, scatter plots, quicklooks, etc. New added-value products will be made available like information on the observed air masses origins and associated pollution sources, tropopause height, potential vorticity, co-location with IASI data, etc. The database is being optimized to support the growing number of users and the integration of data from the IAGOS-CARIBIC project and species from new IAGOS instruments.

In the framework of the IGAS (IAGOS for GMES/ Copernicus Atmospheric Service) project, major achievements will be reached for standardisation and interoperability. IAGOS data will be provided in a self-describing format (NetCDF or NASA Ames) including redefined metadata compliant with ISO 19115, INSPIRE and NetCDF-CF standards.

We also are implementing interoperability between all the involved IGAS data services, including the IAGOS central database, the DLR Research Aircraft database and the Jülich WCS web application JOIN which combines model outputs with in situ data for intercomparison. These data services will also be made interoperable with other international portals and databases.

The JOIN application will be enhanced in order to allow graphical comparison between in situ and spatial observations and model results from interoperable web services, such as the daily products from the Copernicus atmospheric service.

3.2 - Quantifying the source/receptor link for the IAGOS observation database

Antoine Auby

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Presenter :

Since 1994, the IAGOS program has produced in-situ measurements of the atmospheric composition during more than 40000 commercial aircraft flights. In order to help analyzing these observations and understanding the processes driving their evolution, we are developping add-value products quantifying their source/receptor link.

We improved the methodology used by Stohl et al. (2003), based on the FLEXPART plume dispersion model, to simulate the contributions of anthropogenic and biomass burning emissions from the ECCAD database (http://eccad.pole-ether.fr) to the measured carbon monoxide mixing ratio along each IAGOS flight. Contributions are simulated for the last 20 days before the observation, separating individual contributions from the different source regions.

The main goal is to supply add-value products to the IAGOS database showing pollutants geographical origin and emission type. Using these informations, it will be possible to link trends in the atmospheric composition to changes in the transport pathways and to the evolution of emissions. We will also show that this tool can be used for statistical validation and intercomparison of emission inventories using large amounts of data, as lagrangian models are able to bring the global scale emissions down to a smaller scale, where they can be directly compared to the in-situ observations from the IAGOS database.

3.3 - Ice or Ash? Real time detection using the next generation IAGOS BCP

Darrel Baumgardner

Droplet Measurement Technologies, United States of America, darrel.baumgardner@gmail.com
Martin Gallagher (University of Manchester), Andreas Petzold (Forschunsgzentrum Juelich),
Angela Dean (FAAM)

Presenter : Darrel -Baumgardner

The backscatter cloud probe (BCP) has been providing measurements of cloud size distributions since its implementation as part of IAGOS since July, 2011 and is now operating on six A-340 passenger aircraft. Like the majority of single particle optical spectrometers, the BCP measures the equivalent optical diameter (EOD) of particles but is unable to identify the type of particles, e.g. if it is a water droplet, ice crystal, dust or volcanic ash. In addition, because of the close proximity of the sensitive sample volume to the aircraft skin, many of the ice crystals that are counted and sized could be fragments of ice crystals that have impacted the fuselage in front of the BCP but cannot be distinguished from natural crystals.

To address the shattering issue and to expand the capability of the BCP to differentiate different types of atmospheric particles, Droplet Measurement Technologies (DMT) was given financial support from the University of Manchester and Forschungszentrum Juelich to develop a next generation BCP that incorporated the technology that had already successfully been implemented in the DMT CAS-POL that can separate ice crystals from ash and water droplets. A prototype Backscatter Cloud probe with Polarization Detection (BCPD) has been developed with a form factor only 10% larger than the BCP but with the capability of differentiating between droplets, ice crystals and potentially volcanic ash. In addition,

because it measures the interarrival times of particles, some rejection of fragments of shattered ice crystals is possible. An added feature is the processing of the particle transit times and shapes of the transit signal to provide additional information on particle morphology.

The design and operating principles of the BCPD will be presented and examples from flights given in comparison with other cloud probe measurements.

3.4 - Simultaneous Measurement of Particle Extinction and Scattering Using the CAPSssa Monitor

Timothy Onasch

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Presenter : Andrew Freedman, Aerodyne Research, Inc., USA - af@aerodyne.com

We present data demonstrating the performance of an instrument (the CAPS PMssa) that simultaneously incorporates measurements of both airborne particle extinction and scattering coefficients and thus single scattering albedo (SSA) on the same sample volume. Based on cavity attenuated phase shift (CAPS) techniques, the sensor, which detects optical extinction and scattering within a 10 nm bandpass band centered around a particular wavelength, comprises a light emitting diode (LED), an enclosed measurement cell (~26 cm in length) incorporating a resonant optical cavity of near-confocal design and a vacuum photodiode detector. To measure light scattering, a 10 cm diameter integrating sphere with a photomultiplier tube detector is incorporated into the middle of the measurement cell to collect scattered light at all angles with minimal bias. The scattered-light detection circuitry is gated so as to only measure the scattered light during the beam-off phase of the cycle. The use of a low loss optical cavity ensures that there is sufficient optical circulating power that remains within the cell after the LED has been turned off to perform this task. Noise levels (1 sigma) in both the scattering and extinction measurement channels are below 1 Mm-1 with 1 second sampling. Error estimates indicate that the instrument can provide an absolute accuracy in the SSA of better than ±0.03. The absolute accuracy of the individual scattering and extinction measurements are limited by the accuracy of the particle counting equipment used to provide a correction for the sampling cell geometry and in the case of scattering , the calculation of the truncation correction.

4 UTLS chemical composition and trends

4.0 - UTLS chemical composition and trends (Keynote)

William Randel

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Presenter : William Randel

The upper troposphere – lower stratosphere (UTLS) is a region of complex interplay between dynamical, radiative and chemical behaviors, influenced by large- and small-scale circulations and transport from above and below. In situ aircraft observations from MOZAIC-IAGOS are providing novel long-term characterization of chemical variability and links to circulation, and promise to be a key data source for the future. This talk will provide an overview of UTLS chemical behavior and highlight current 'hot topics', including chemical aspects of monsoonal circulations, influences of persistent deep convection, and long-term variability and changes in UTLS composition in models and observations.

4.1 - Comparison of ozone concentrations in the UTLS as measured by ozone sondes and commercial airliners (MOZAIC)

Johannes Staehelin

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Johannes Staufer, Fiona Tummon, Rene Stuebi, Herman Smit and Valerie Thouret

Presenter : Johannes -Staehelin

Ozone sondes have been launched at selected sites since the late 1960s to measure vertically highly-resolved ozone profiles used to construct ozone climatologies (e.g. for comparison with numerical simulations) and to deduce long-term ozone changes. However, the reliability of long-term tropospheric trends deduced from Brewer Mast sondes (mainly flown at European sites in previous decades) has recently been questioned. Furthermore, systematic differences have been found in the data series obtained from the most commonly used EEC sonde types (i.e. regarding impacts of the sensor manufacturer and solute concentrations, based on chamber studies and dual flights).

Since 1994, ozone in the upper troposphere-lower stratosphere (UTLS) has been continuously monitored by measurements from regular aircraft as part of the project MOZAIC (Measurement of Ozone and Water Vapor by Airbus in Service Aircraft Program). In this study, a method will be presented to compare ozone measurements from MOZAIC derived at flight altitude (largely in the UTLS) with ozone sonde data from various launch sites by identifying the same air masses using trajectory analysis. The method was tested extensively and an uncertainty of ±2% was found. Most ozone sonde data in the northern extra-tropics show encouraging agreement for the period after 1998 (in the order of 5-10%, consistent with laboratory studies). Prior to this, however, discrepancies are much larger. Potential reasons for these discrepancies will be discussed in the presentation.

4.2- Free Tropospheric Ozone Trends over Southern Africa (1990-2008) Re-visited: A Study with MOZAIC and SHADOZ Data

Anne M. Thompson

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Presenter : Anne M. Thompson

Five years ago Clain et al. (2009) reviewed 18 years of ozone soundings from Irene (South Africa, 25S, 28E) and Réunion island (21.5S, 55E), that is frequently downwind of South Africa. They reported that, on an annually averaged basis, trends in boundary layer (BL) ozone over Irene, were +14%/decade from 1990 through early 2008. During the SHADOZ period, 1998-2008, the BL ozone increase reached 30%/decade. For the mid-troposphere, a statistically significant ozone increase was only found in winter; no trend was reported in the upper troposphere. At Réunion, with a sounding record from 1992 to 2007, the corresponding trends were only significant above 10 km. Recently we re-analyzed the sounding data for the BL and free troposphere (FT, 4-11 km for Irene; 4-15 km over Réunion). Trends were determined with a multivariate regression model that accounts for ENSO, QBO and seasonal cycles in ozone. At Irene, MOZAIC data from nearby Johannesburg International Airport were used to fill in a sonde gap for 1995-1999. The results are summarized as: (a) a drift in launch time between the 1990-1993 Irene record and sondes launched after 1998 is responsible for at least some of the large BL ozone increases of Clain et al. (2009). (B) From 6-11 km, in late fall-early winter (April-July) there is a +20-25%/decade ozone increase over Irene from 1990-2007. Over Réunion, for 1992-2011, from 5-13 km, there is a stronger trend: 40%/decade. Both locations displayed a lesser FT ozone increase in December, with +50%/decade in ozone near the tropopause. For the latter change, we examined dynamical markers but did not find a clear explanation. For the wintertime FT ozone increases, changes in emissions of ozone precursors and back-trajectories from the soundings were studied as possible causes. The hypothesis that long-range transport (LRT) of pollution from Africa, Madagascar and South America is a contributor appears to be confirmed by the trajectories. Over Réunion the large ozone increase maybe due to LRT from rapidly developing regions in south and/or southeast Asia.

4.3 - High resolution simulations of indian pollution transport in the UTLS during the Asian summer monsoon.

Flore TOCQUER

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BARRET Brice, MARI Céline

Presenter : Flore TOCQUER

The Asian summer monsoon is a prominent feature of the global circulation that is associated with an upper-level anticyclone (ULAC) which, stands out vividly in satellite observations of trace gases as CO. The contribution of surface emissions up to this altitude is only possible through intense dynamic processes. Recent studies have shown that during the monsoon period, air masses could be uplifted from the surface to the upper troposphere by deep convection occuring over the Tibetan plateau and the southern slopes of the Himalayas. Furthermore, the continuous and fast increase of pollutant emissions from South Asia makes this transport pathway particularly relevant for the O3 budget in the UTLS.

Simulations with the Méso-nh model were carried out to characterize the convective processes responsible for the vertical transport of surface pollutants to high altitudes and to better understand their impact on the composition of the ULAC. The first step has been to document convective systems from satellite data in order to identify different types of events. The next step has been to set up a case study corresponding to an extreme event and simulate the emission and transport of a passive CO tracer. A simulation including complete NOx-COV chemistry will allow us to further assess the impact of such an event to the composition of the UTLS. IASI O3 and CO data will be employed to validate the O3 and CO distributions simulated by Méso-NH. Finally, case studies corresponding to convective systems with different charactersitics (region, depth, horizontal extent...) will enable to determine the main factors controling the convective transport of pollution to the UTLS.

4.4 - Ozone budget over South Asia during the summer monsoon: chemistry transport modelling versus satellite and IAGOS observations

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Bastien Sauvage, Yasmine Bennouna and Eric Le Flochmoën

Presenter : Brice Barret

Satellite observations have documented the accumulation of primary pollutants (CO, HCN) into the Asian Monsoon Anticyclone (AMA) up to the lower stratosphere. Global chemistry transport models have shown that this accumulation is due to south and east Asian pollution transported by large scale and convective uplift during the Asian Summer Monsoon. This uplift of lower tropospheric air masses has a potentially strong impact upon the Upper Troposphere-Lower Stratosphere (UTLS) O3 and water vapour budget and therefore also on the surface radiative forcing. Satellite observations and models have shown that O3 concentrations are lower within the AMA than in the surrounding tropical UT, as a result of the uplift of BL O3-poor air masses to the UT. Nevertheless, boundary layer O3 precursors (VOCs, NOx) uplifted from the densely populated Asian regions are responsible for the photochemical production of O3. Other sources have a potentially large impact on UT O3 enhancement : the production of NOx by lightnings (LiNOx) within the monsoon convective systems, stratospheric intrusions from the subtropical jet and long range transport. Furthermore, the large scale AMA circulation and its variations strongly interacts with these potential sources of O3.

The aim of our study is to quantify the relative impact of these different processes on the Asian UT O3 concentrations during the pre-monsoon to post-monsoon period. We have therefore performed simulations with the GEOS-Chem global chemistry transport model for the year 2009. Distributions of CO and O3 from a reference simulation are first evaluated versus IAGOS observations from Hyderabad in central India and versus IASI retrievals over Asia. For budget assessment we have performed sensitivity simulations with the NOx emissions from different regions switched off. The tagging of stratospheric O3 in the reference simulation also enabled us to determine the impact of stratospheric intrusions on O3 in the Asian UT. We will present the results of this set of simulations focusing on the role of the AMA.

4.5 - Ten Years of Carbon Monoxide Measurements from the MOZAIC Program

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V. Thouret, P. Nedelec and the MOZAIC team

Presenter : Hannah Clark

The program MOZAIC (Measurement of Ozone and Water Vapour on Airbus in-service Aircraft) began in 1995 with commercial aircraft equipped with scientific instruments to measure ozone and water vapour in the atmosphere. After 7 years of successful operation, the range of chemical species measured was expanded to include carbon monoxide. We present a climatology of CO measured in the upper troposphere and lower stratosphere at cruise altitudes between 9 and 12 km over the period 2002-2012. The distribution of CO in the upper troposphere is strongly influenced by natural and anthropgenic emissions in the boundary layer. In particular, we note seasonal differences in biomass buring over equatorial Africa and the signatures of boreal forest fires over Siberia and North America. Concentrations are seen to increase by 20% on passing from Western to Eastern hemispheres. In the North American, Atlantic and European sectors, CO concentrations show an annual reduction of 2%. In the Siberian/North Asian sector a trend is more difficult to determine due to the influence of some particularly pronounced boreal fire events.

4.6 - Nitrogen oxides in the UTLS: Long term observations with CARIBIC

Helmut Ziereis

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A. Zahn, Institute for Meteorology and Climate, Karlsruhe Institute of Technology, Germany
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Presenter : Helmut Ziereis

The upper troposphere/lower stratosphere (UTLS) is a region of special interest for atmospheric chemistry and dynamics. The climate forcing of radiatively active gases like ozone and methane is particularly sensitive to concentration changes in this region.

Nitrogen oxides and total reactive nitrogen have a decisive influence on the chemistry in the UTLS. They act as catalyst in several reaction chains influencing the cycling of OH, the production of ozone and the lifetime of methane. The impact of additional emitted nitrogen oxides on these gases, for example by aircraft, also depends on its background concentration. However, due to their comparatively short lifetime and their variety of sources, there is a high uncertainty of the abundance of nitrogen oxides in the UTLS.

Nitric oxide (NO) and total reactive nitrogen (NOy) have been measured within the CARIBIC (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container) project since 2002. More than 300 flights have been performed on a monthly base using a Lufthansa Airbus A340-600 based in Frankfurt/Germany. Long-distance flights have been performed to destinations in North- and South America, Asia and Africa.

Short lifetimes and a complex source distribution complicate the establishment of a climatology for reactive nitrogen species in the UTLS. However, the present data set is suitable to form the basis for it, at least for some regions. The large scale seasonal and regional distribution of nitric oxide and total reactive nitrogen in the UTLS is presented and compared to other measurements and to the results of model simulations. Significant regional and seasonal differences in the NO and NOy abundance have been observed. Signatures of aircraft emissions or biomass burning respectively have been regularly observed. The extensive spectrum of concomitant measurements by CARIBIC gives additional insights.

4.7 - Seasonal changes of CO2, CH4, N2O and SF6 in the upper troposphere/lower stratosphere over the Eurasian continent observed by commercial airliner

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Presenter : Sawa Yousuke

Atmospheric mixing ratios of greenhouse gases at about 11 km altitude were analyzed from monthly air sampling aboard commercial airliner during the flights between Europe and Japan from April 2012 to August 2013. Compared to the subtropic, higher CH4 and SF6 mixing ratios, similar values of N2O, and larger seasonal changes of CO2 were found in the upper troposphere. CH4, N2O and SF6 in the lower stratosphere, above the tropopause up to 30 K in potential temperature, showed simultaneous increases from June to October, and faster decreases at higher altitudes from January to March. Mean age of the air in the lower stratosphere was estimated based on SF6 mixing ratios to be about 2 years in late spring and 1 year in autumn, suggesting stronger influences on the mixing ratios in the stratosphere from troposphere in summer.

4.8 - Aerosol Particle Distributions, Sources and Trends in the UT/LMS Measured by the CARIBIC Observatory since 2000

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Presenter : Denise Assmann, Leibniz Institute for Tropospheric Research, Germany, assmann@tropos.de

Submicrometer aerosol particles in the upper troposphere and lowermost stratosphere (UT/LMS) influence the Earth`s radiation budget directly and, more important, indirectly, by acting as cloud condensation nuclei and by changing trace gas concentrations through heterogeneous chemical processes. Since 1997, the Leibniz Institute for Tropospheric Research, Leipzig, Germany and the University of Lund, Sweden, are conducting regular in situ aerosol particle measurements in the UT/LMS onboard passenger aircraft with the CARIBIC (now IAGOS-CARIBIC) observatory (www.caribic-atmospheric.com). Aerosol particle number concentrations and the particle size distribution are measured in situ using four particle counters (3 CPCs, 1 OPC). Moreover, particle elemental composition is determined using an aerosol impactor sampler and postflight ion beam analysis (PIXE, PESA) of the samples in the laboratory.

Based on this unique data set and meteorological analysis, we present near-global distributions of particle concentrations in 8-12 km altitude and how clouds in different regions of the world influence these concentrations. Besides clouds, volcanic eruptions influence the UT/LMS aerosol most strongly. Hence, we discuss the volcanic influence on the particle elemental composition and on particle concentrations. Moreover, we present trends in number concentrations, but also in the elemental composition, determined from our regular measurements over more than a decade.

4.9 - The extratropical tropopause inversion layer

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Alison Ming, DAMTP, University of Cambridge

Presenter : Peter Haynes

The extratropical tropopause inversion layer (TIL) is a layer with thickness of a few km of high static stability lying in the extratropical stratosphere immediately above the tropopause. The layer can been identified in vertical profiles of static stability extracted from high resolution radiosonde measurements or radio occultation measurements but can also be identified in profiles extracted from large-scale meteorological analysis datasets. It is particularly clear when the profiles are composited with respect to the local tropopause height (defined by a lapse-rate criterion) rather than with respect to height.

It has been suggested that the presence of the TIL is important for the distribution of chemistry species in the tropopause region.

Two key questions are (i) what are the physical mechanisms leading to the formation of the TIL and (ii) what are the implications of compositing with respect to tropopause height. (i) is addressed by simulations in an idealised general circulation model with a simple radiative relaxation state. We demonstrate, following previous authors, that a TIL-like structure can be generated by the large-scale dynamics without any need for similar vertical structure in the restoration state. However, this does not always occur. We explain the formation of the TIL in terms of the E-P flux convergence and identify the essential feature as there being two separate regions of convergence, one in the upper troposphere and the other in the lower stratosphere. (ii) is addressed by considering 'synthetic' temperature profiles in which fluctuations of different types are imposed on a background and the corresponding composite profiles are calculated. The conclusion is that the compositing cannot by itself generate a TIL-like structure, but that it can act to accentuate a TIL-like structure that is pre-existing in the background. The implications for chemical species are considered, bearing in mind that dynamical fluctuations that are relatively long-lived, e.g. associated with potential vorticity anomalies, are likely to have different effects than those that are short-lived, e.g. associated with gravity waves.

4.10 - Seasonality of the mean age in the UTLS region: Hemispheric differences and impact of the Asian monsoon

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Presenter : Paul Konopka

The seasonality of the composition of air in the UTLS region is determined by the seasonality of different transport processes like convection, Brewer-Dobson circulation (BDC) and two-way irreversible isentropic transport across the tropopause. Whereas during winter (seasons are related to the northern hemisphere), the subtropical jets form a strong transport barrier between the tropics and extratropics, this barrier weakens significantly in the northern hemisphere during summer. This is a result of the hemispheric asymmetry of the land-sea distribution and of the orography, which leads to hemispheric differences in the distribution and intensity of the wave drag driving the BDC.

Based on a multi-annual CLaMS simulation covering the period from 2001 to 2012 with the model transport driven by the ECMWF ERA-Interim reanalysis, we discuss the seasonality of the mean age (measuring the mean transport time of an air parcel traveling from the boundary layer) in the tropical tropopause layer (TTL) and in the extratropical lowermost stratosphere (LMS). During the considered period, the simulated trace gases (like CH4, N2O, F11, CO2, CO, H2O and O3) are in fairly good agreement with in-situ and satellite observations, especially in the lower stratosphere and around the tropopause.

In the TTL, the mean age shows a pronounced annual cycle that is driven by the seasonality in tropical upwelling and horizontal transport from the extratropics (inmixing) with youngest air during late boreal winter and oldest air during late boreal summer, respectively. On the other side, strong hemispheric differences can be diagnosed in the polar high latitude LMS. Here, air in the northern hemisphere is much younger during summer than during the same season on the southern hemisphere. A regionally resolved climatology of the mean age further shows youngest air in the TTL in winter above the West Pacific warm pool, whereas in summer the Asian summer monsoon forms the key pathway for transport into the LMS

By quantifying the wave forcing in terms of the transformed Eulerian mean formalism (TEM), we derive respective climatologies of dynamical sources (EP-flux divergence) and explain transport and its seasonality within this framework. In this way, we trace back the seasonality and hemispheric differences of the mean age to the respective differences in the strength of the Arctic and Antarctic polar vortices, different climatological patterns of the upper tropospheric anticyclones and of the orographic Rossby waves in the troposphere. By analysing the TEM version of the transport equation we also quantify the impact of the residual circulation and of eddy mixing on causing the mean age seasonality. Regionally resolved analysis emphasizes the importance of the Asian continent and in particular of the Asian monsoon on the composition of air in the UTLS region over the northern hemisphere.

5 Monitoring atmospheric composition, climate and air quality

5.0 - The importance of IAGOS in-situ observations for the Copernicus Atmosphere Service

Vincent-Henri Peuch

European Centre for Medium-Range Weather Forecasts
Head of Atmospheric Composition Division, MACC-II co-ordinator

Copernicus, previously known as GMES (Global Monitoring for Environment and Security), is the programme for the establishment of a European capacity for Earth Observation. Over the next years, Copernicus will strengthen the European infrastructure for monitoring the Earth's environment (the "Sentinels" satellites) and will continuously deliver operational services addressing six thematic areas: land, marine, atmosphere, climate change, emergency management and security. These services will support a wide range of applications as well as policymakers and public authorities who need the information to develop environmental legislation and policies or to take critical decisions. Based on the Copernicus services and on the data collected through the Sentinels and the contributing missions, many value-added services can be tailored to specific public or commercial needs, resulting in new business opportunities.
The Copernicus Atmosphere Service (CAS) addresses the aspects relative to atmospheric composition, by providing monitoring, forecast or retrospective information on greenhouse gases, reactive gases and aerosol at the global scale and (at higher resolution) over Europe. This Service is based upon a heritage of successive European FP projects (GEMS, MACC, MACC-II), which have supported ambitious scientific and technical developments over the last decade. These projects have made routine use of IAGOS data, which provide rare and invaluable altitude in-situ information. IAGOS observations have been particularly useful for independent validation of the products provided by the projects, while their potential has been explored for data assimilation (as "anchor" for bias-correction schemes or "active" assimilation). The future deployment of a larger fleet of equipped IAGOS aircraft and covering various atmospheric constituents constitutes a very important perspective for the CAS. The NRT processing and data transmission capabilities of IAGOS will also make it possible to include the corresponding data stream as part of the operational processing, just like Aircraft Meteorological Data Relay (AMDAR) for Numerical Weather Prediction.
This talk will present some of the insight and experience gained with the use of IAGOS data in the currently on-going project MACC-II, highlighting in particular the usefulness of the altitude in-situ information in cases of significant large-scale pollution episodes.

5.1 - Validation of the MACC-II atmospheric composition global forecasting service

Henk Eskes

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MACC-II validation partners

Presenter : Henk Eskes

The MACC-II (Modelling Atmospheric Composition and Climate, http://www.gmes-atmosphere.eu/) project is establishing the core global and regional atmospheric environmental services delivered as a component of Europe's GMES (Global Monitoring for Environment and Security) initiative. The global near-real time service of MACC-II consist of daily analysis and forecasts of atmospheric composition produced with the MACC system. These analysis are obtained by assimilating trace gas concentrations and aerosol properties as measured by operational and research satellites. Apart from the near-real time forecasts, MACC provides reanalyses of atmospheric composition for the time period 2003-2012. A dedicated

validation activity exists within MACC-II to provide a routine validation of these services based on independent observations (in-situ, remote sensing and satellite).

In our contribution we will discuss the validation approach, system evolution, and validation results of the MACC-II NRT global atmospheric composition forecasting service. The performance of the system is assessed in two ways: both the longer-term mean performance (seasonality) as well as its ability to capture recent events are documented. During MACC II a series of validation reports is produced, updated every 3 months, to document the validation statistics of the near-real time service. These reports cover results for a period of at least one year to document the seasonality of the biases. Changes in the performance related to model upgrades are documented by comparing with previous versions and stand-alone model results. In my talk I will discuss in particular the contribution of MOZAIC/IAGOS to the validation of O3 and CO of the MACC-II daily forecasts and reanalysis.

5.2 - Technical challenges to integrating aircraft measurements of atmospheric composition with numerical models

Mark Parrington

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Presenter : Mark Parrington

Routine in situ measurements of atmospheric constituents from civilian aircraft under IAGOS-MOZAIC are expected to play a significant role in future monitoring of atmospheric composition and climate through the MACC-II and follow-on MACC-III pre-operational services, and the operational Copernicus atmospheric service. Ozone (O3) and carbon monoxide (CO) data from IAGOS are currently used in the MACC-II project for verification of global satellite data assimilation using the ECMWF Integrated Forecast System (IFS) and a framework for integrating these data with the IFS will be implemented for MACC-III. An additional project (IAGOS for the GMES Atmospheric Service, IGAS) will further increase the operational capability of the IFS to include greenhouse gas and aerosol measurements made by the IAGOS fleet. While the precise role of these data has yet to be fully established, it is anticipated that they will provide critical information required for monitoring, bias correction, and assimilation of atmospheric composition in the IFS. To evaluate some of the technical challenges to integrating in situ aircraft data with numerical models we present an overview of assimilation experiments using measurements made during a research campaign (BORTAS) over the North Atlantic in the summer of 2011. Profiles and flight-level measurements of tropospheric O3 and CO made during BORTAS by the UK FAAM BAe-146 research aircraft, and coincident IAGOS measurements, have been assimilated into a chemistry transport model using both sequential (Kalman filter) and variational (4D-Var) techniques. Lessons learned from these assimilation experiments and their potential application to IAGOS data in an operational setting, to define, for example, the observation operator and error correlation length-scales, and first results of assimilating IAGOS O3 and CO profiles in the IFS will also be presented.

5.3 - Use of simultaneous O3 and CO data from MOZAIC-IAGOS to evaluate the MACC-II Reanalysis in the UTLS – Tellus-B : no

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H. Clark, V. Thouret, L. Jones, A. Inness, H. Eskes, V. Huijnen, P. Nédélec, MACC team, IAGOS team

Presenter : Audrey Gaudel

Tropospheric ozone plays a major role in the chemistry of the troposphere by exerting a strong influence on the concentrations of oxidants such as hydroxyl radical (OH) and is the third greenhouse gas after carbon dioxide and methane. Its radiative impact is of particular importance in the Upper Troposphere / Lower Stratosphere (UTLS), the most critical region regarding the climate change. Carbon Monoxide (CO) is one of the major ozone precursors in the troposphere. In the UTLS, it also has implications for stratospheric chemistry and indirect radiative forcing effects.
The aircraft of the MOZAIC program have collected simultaneously O3 and CO data regularly all over the world since the end of 2001. Most of the data are recorded in northern mid-latitudes, in the UTLS region (as commercial aircraft cruise altitude is between 9 and 12 km).
We will present and discuss the performance of the MACC-reanalysis, including the ECMWF-Integrated Forecasting System (IFS) coupled to the CTM MOZART with 4DVAR data assimilation, to reproduce ozone and CO in the UTLS, as evaluated by the observations of MOZAIC between 2003 and 2008.
In the UT, the model tends to overestimate O3 by about 30-40 % in the mid-latitudes and polar regions. This applies broadly to all seasons and this is even stronger in DJF and MAM. In tropical regions, the model underestimates UT ozone by about 20 % in all seasons and even stronger in JJA.
Upper-tropospheric CO is globally underestimated by the model in all seasons, by 10-20 %. In the southern hemisphere, it is particularly the case in SON in the regions of wildfires in South Africa. In the northern hemisphere, the zonal gradient of CO between the US, Europe and Asia is not well-captured by the model, especially in MAM.

5.4 - Evaluation of hindcast simulations using aircraft and other types of observations

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Presenter : Thierno Doumbia, UPMC Univ. Paris 06, LATMOS-IPSL, France, doumbia@latmos.ipsl.fr

Global chemistry-climate models have been used to simulate the evolution of the atmospheric composition over the past decades. These simulations have been performed in free-running and specified dynamic modes, using the NCAR Community Earth System Model. The simulations have been done at different horizontal and vertical resolutions. We have analyzed the long-term changes as well as the interannual variability of several atmospheric compounds, with a focus on ozone, carbon monoxide and nitrogen dioxide. We have investigated the behavior of these species by focusing on two regions, Europe and Asia.

In Europe, surface emissions have decreased significantly since the 1980s, which have led to a decrease in the concentrations of several tropospheric compounds. On the contrary, emissions in Asia have dramatically increased, particularly during the past two decades, which has resulted in large increases in the atmospheric content of several species.We have compared the model simulations to different types of observations. We have used the MOZAIC and IAGOS observations of CO and ozone to analyze the model results, and focused on the vertical profiles measured in different airports in the regions under consideration. We have also compared the model results with satellite observations (IASI, MOPITT, OMI, GOME and SCIAMACHY) and ozone sondes.

5.5 - Assessing the regional representation of MOZAIC trace gas soundings in the lower troposphere and suitability for use in regional chemical transport model evaluation

Morgan Silverman

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Presenter : Morgan Silverman

Aircraft measurements provide a unique data set to study tropospheric concentrations and transport, and provide useful information for chemical transport model evaluation. With continued stringency to the United States Environmental Protection Agency (EPA) national ambient air quality standards (NAAQS) for ground-level O3, there is increased emphasis on the important to accurately simulate the contributions of O3 and its precursors aloft to concentrations at the surface. In the past, surface measurements have been the primary data source used for model evaluation. While these analyses are essential, profile data are needed to assess processes such as vertical mixing between the boundary layer and free troposphere, hemispheric transport, lightening generated NOx, and stratospheric intrusions.
Since the 1990's several programs have installed automatic measuring instruments on commercial airliners (NOXAR (Brunner et al., 1998), CARIBIC (Brenninkmeijer et al., 1999), and MOZAIC (Marenco et al., 1998). From these, MOZAIC flights are the only in-service flights to U.S. airports. Recent interest in using MOZAIC data to evaluate regional chemical transport models, such as the community multiscale air quality model (CMAQ) (Solazzo et al., 2013, Hogrefe et al., 2013) raises questions regarding the suitability of MOZAIC data in the mid to lower troposphere (<6 km) during take-off and landing.
Fischer et al. (2006) suggests that because the trace gas profiles provided via commercial flights are often restricted to the vicinity of heavy-duty airports, the observation do not provide a are not good representation of the background atmosphere, and hence may be a biased data set and limit use for regional chemical transport model evaluation in the lower parts of the troposphere. This raises the question, to what extend are aircraft data influenced by aviation emissions along flight paths and how well does the data represent the regional environment at lower altitudes? This work characterizes MOZAIC measurements of O3, CO, and NOy from June, July, August, and September 2002-2004 at major U.S. airports. The MOZAIC data is compared to aircraft profiles conducted under the NASA Earth Venture-1 DISCOVER-AQ (Deriving Information on Surface conditions from COlumn and VERtically resolved observations relevant to Air Quality) mission, which occurred during July 2011. We find that MOZAIC is representative of an urban environment. There is no clear aircraft or flight path influences that are seen in the dataset.

5.6 - Three-dimensional behaviors of atmospheric CO2 revealed by CONTRAIL project and their use for carbon cycle studies

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Presenter : Toshinobu Machida

Frequent measurements of atmospheric CO2 using Continuous CO2 Measuring Equipment (CME) as well as other greenhouse gases by Automatic Air Sampling Equipment (ASE) onboard the commercial airliners under the CONTRAIL project brought us huge numbers of CO2 data in upper air and revealed latitudinal, longitudinal and vertical difference in CO2 variation worldwide. The CONTRAIL project has been conducted since 2005 using 8 aircraft operated by Japan Airlines. Until the end of 2013, more than 10,000 of CME flights were made between Japan and Europe, South Asia, Southeast Asia, East Asia, Australia, Hawaii and North America, and 17,000 vertical profiles have been obtained over there.

Seasonal variations of CO2 observed in northern mid-high latitudes are 7-12 ppm. Seasonal amplitudes are generally larger in lower altitudes over the continent, because of the strong influence by land biosphere, but the vertical differences are small over the Pacific islands such as Hawaii and Guam. Amplitudes are gradually weakened along the latitude and are only 1-2 ppm in the southern hemisphere. Unlike the northern mid-high latitudes, larger amplitudes are found in upper troposphere over Sydney, Australia compared to the surface. This is due partly to the efficient inter-hemispheric transport in upper troposphere for high CO2 from the Northern Hemisphere. The rapid increase in the upper southern lower latitudes is equivalent to about 0.2 Pg increase in carbon.

An inversion analysis was performed using the CONTRAIL-CME with the NICAM based transport model. The inversion result shows that the CONTRAIL data have significantly large impacts on estimates of tropical terrestrial fluxes. Comparing inversion with surface data alone, posterior errors were reduced by up to 64 % in the Asian tropics. The inversion with the CONTRAIL data yields the global carbon sequestration rates of 2.22±0.28 Pg C yr-1 for the terrestrial biosphere and 2.24±0.27 Pg C yr-1 for the oceans.

5.7 - Large-scale distributions of methyl chloride in the upper troposphere observed by CARIBIC - Tellus- B : no

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Presenter : Taku Umezawa

CARIBIC is a flying observatory using a Lufthansa A340-600 aircraft for measuring various atmospheric compounds at monthly intervals. We present spatial and temporal variations of methyl chloride (CH3Cl) in the upper troposphere (UT) observed mainly by CARIBIC for the years 2005–2011. The CH3Cl mixing ratio in the UT over Europe was higher than that observed at a European surface baseline station throughout the year, indicative of a persistent positive vertical gradient at NH mid latitudes. A series of flights over Africa and South Asia show that CH3Cl mixing ratios increase toward tropical latitudes, and the observed UT

CH3Cl level over these two regions and the Atlantic was higher than that measured at remote surface sites. Strong emissions of CH3Cl in the tropics combined with meridional air transport through the UT may explain such vertical and latitudinal gradients. Comparisons with carbon monoxide (CO) data indicate that non-combustion sources in the tropics dominantly contribute to forming the latitudinal gradient of CH3Cl in the UT. We also observed elevated mixing ratios of CH3Cl and CO in air influenced by biomass burning in South America and Africa, and the emission ratios derived for CH3Cl to CO in those regions agree with previous observations. In contrast, correlations indicate a high CH3Cl to CO ratio of 2.9+/-0.5 ppt ppb-1 in the Asian summer monsoon anticyclone and domestic biofuel emissions in South Asia are inferred to be responsible. We estimated the CH3Cl emission in South Asia to be 134+/-23 Gg Cl yr-1, which is higher than a previous estimate due to the higher CH3Cl to CO ratio observed in this study

5.8 - Global distribution and trends of tropospheric ozone : A review

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Presenter : Owen Cooper

A comprehensive understanding of global surface ozone trends has eluded the scientific community due to limited long-term in situ observations and relatively few ozone monitors in regionally representative rural or oceanic regions. Furthermore, satellite records of lower tropospheric ozone mixing ratios are presently too short to yield robust results. However, in recent years several studies have provided updates to ozone trends at long-established sites, or reported trends at many newer sites that now have lengthy records sufficient for trend analysis. To pull all these new findings together into a single reference, many colleagues and I have produced a review article on the global distribution and trends of tropospheric ozone [Cooper et al., 2014]. Relying upon this synthesis I will review all of the current ozone trend analyses in the peer-reviewed literature, focusing on rural rather than urban monitoring sites to facilitate understanding of ozone changes across broad regions. Trends at rural sites are also more easily compared to global chemistry-climate models. The earliest reliable ozone records began in the 1950s and 1970s with more and more sites becoming available in the 1980s and 1990s. The ozone rate of change has varied in magnitude and even sign over the decades with the strongest changes occurring in East Asia, eastern North America and Western Europe where changes in domestic ozone precursor emissions have been greatest. Ozone trends since the 1990s will be compared to global images of satellite-detected tropospheric column NO2 to identify regions where ozone trends are consistent, or inconsistent with observed ozone precursor changes and our general understanding of tropospheric chemistry. The important contribution of MOZAIC data to this global ozone trend review will also be highlighted. Finally, I will present a new global analysis of 9-years of tropospheric column ozone as detected by the polar orbiting OMI and MLS instruments on the NASA AURA satellite, contrasting interannual ozone variability in the northern and southern hemispheres.

Cooper, O. R., D. D. Parrish, J. Ziemke, Stefan Gilge, Niels Jensen, Manuel Cupeiro, J.-F. Lamarque, L. Horowitz, V. Naik, D. T. Shindell et al. (2014), Global distribution and trends of tropospheric ozone: A review, Elementa, submitted.

5.9 - Inter-annual variability of ozone and carbon monoxide in tropospheric columns derived from MOZAIC/IAGOS and compared to satellite data

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Presenter : Régina M. ZBINDEN

The tropospheric study, derived from the MOZAIC/IAGOS program, discuss the ozone (O3) and carbon monoxide (CO) inter-annual variability (IAV) within 08/1994-02/2011 with a focus on the 2000's.

Due to the irregular MOZAIC/IAGOS sampling frequency, the analyse is based on the most visited sites which are located in the mid-northern latitudes: - Germany (i.e. a cluster of Frankfurt and Munich, 18 043 O3 profiles and 8 527 CO profiles), - Eastern USA (i.e. a cluster of Boston, New York, Philadelphie, Washington, 5 842 O3 profiles and 1 370 CO profiles), - Vienna (4 752 O3 profiles and 1 518 CO profiles), Paris (4364 O3 profiles and 695 CO profiles) and - Japan (i.e. a cluster of Tokyo, Nagoya and Osaka, 3 199 O3 profiles and 1 0001 CO profiles). The first previous first study on tropospheric columns inter-annual variability (Zbinden et al., 2006) is improved because we are now able to quantify the amounts of O3 or CO in the MOZAIC unvisited tropospheric layer in high tropopause conditions (Zbinden et al. 2003). All tropospheric columns with a 50 m vertical resolution are now fully defined from the surface to the dynamical tropopause fixed at 2 pvu extracted from the ECMWF analysis (T213) and interpolated for the aircraft position along the track. Note, the potential vorticity pressures are available on the data base, with a 150m vertical resolution.

We examine the whole time-series, two sub-periods before and after 2000 because the IAV is significant whereas the late years (Hess and Zbinden, 2013; Parrish et al., 2013; Cooper et al., in preparation) and the seasonal IAV.

Additionally a comparison with satellite results is performed using TES and OMI/MLS for O3 and AIRS for CO. This analyse is not a validation because it is not a truly one to one comparison like in Worden et al. (2007) for TES O3 and in Mac Millan et al. (2011) for Airs CO. Nevertheless, this simple comparison is interesting by itself because it results from two independent data sets, with their own limit and performance.

5.10 - Impacts of Indian emissions in the tropospheric ozone levels in South Asian region

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Presenter : E.Surendran Divya

We have investigated the impact of chemical emissions in the Indian subcontinent on the tropospheric abundance of ozone and its precursors in the surrounding south Asian region. The study makes use of the global Chemistry-Transport Model (MOZART) version-4 forced with dynamical fields provided by meteorological analyses from Modern Era Retrospective-analysis for Research and Applications (MERRA) of the Goddard Earth Observing System Data Assimilation System (GEOS DAS) and with HTAP emission inventories established for 2008. Ozone production in this region is dominated by the anthropogenic emissions of carbon monoxide, oxides of nitrogen and non-methane hydrocarbons. The influence of Indian emissions over most of the surrounding South Asian region leads to an increase in the ozone and CO concentrations in most part of the free troposphere. During the monsoon period of July, the marine winds transport boundary level ozone and CO towards the northeastern region of South Asia as opposed to what is occurring during the pre-monsoon period. However, due to the short residence time of NOx, convective transport of nitrogen oxides is not strong and hence changes in the concentration of this compound are small in the free troposphere. The background level of ozone over the Indian region, (fraction of ozone

present in a given area that is not attributed to anthropogenic sources in this area) is approximately 10-30 ppbv and is even larger in the northern part of India during the monsoon period. The contribution of long-range transport of ozone from distance pollution sources in south Asian region to the Indian region is substantial for ozone and CO, but insignificant for NOx.

6 Water vapour and clouds

6.0 - Ice supersaturation and cirrus clouds in the tropopause region

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Cirrus clouds, i.e. clouds consisting exclusively of ice crystals are very frequent in the tropopause region. Since in situ formation of ice crystals takes place far away from thermodynamic equilibrium, large regions in the upper troposphere are in the status of ice supersaturation. During the last twenty years, cirrus clouds and their potential formation regions (so-called ice-supersaturated regions, ISSRs) were investigated using measurements (e.g. MOZAIC data), model simulations and theoretical approaches. However, there is still lack of understanding of key processes for the formation and evolution of cirrus clouds and ISSRs.

In this contribution I will give a short overview about cirrus clouds and ISSRs in the tropopause region. Some new results as obtained recently will also be presented. Finally, I will discuss open questions and possible strategies for solving these issues.

6.1 - Reanalysis of upper tropospheric humidity data of the MOZAIC programme for the period 1994 to 2009

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Presenter : Herman G.J Smit

Water vapour plays a dominant role in the atmospheric energy budget and greenhouse effect. It is one of the key drivers for atmospheric transport and has a strong influence on the oxidative capacity of the atmosphere. Therefore, it takes a key position in the description of climate, chemistry and their interactions. Due to a range of small-scale dynamical and microphysical processes, the abundance of upper tropospheric humidity is highly variable on spatial and temporal scales that cannot be resolved, neither by the global radiosondes network nor by satellites.
Since 1994 high spatial and temporal relative humidity data are provided by the MOZAIC/IAGOS-project (www.iagos.org). The data set emerging from this long-term observation program builds the backbone of the first climatology of upper tropospheric humidity (UTH). First analyses of MOZAIC measurements for the period 1994 to 1999 showed that the UT at cruise altitude (10-12 km) is much wetter than reflected in the model analyses of the ECMWF (European Centre for Medium range Weather Forecast). The MOZAIC data has also demonstrated that ice supersaturation is often large enough to let contrails develop into cirrus but not large enough to let cirrus clouds form naturally.
In this study we have reanalysed the MOZAIC data set on relative humidity (RH) for the period 1994 to 2009. Previous analyses of probability distribution functions (PDF) of UTH- data from MOZAIC observations from the year 2000 and later indicated a bias towards higher RH values. As a result, the PDF of UTH exhibits a maximum at RH over ice (RHice) of approx. 120% instead of the maximum at 100% RHice observed in the period 1994 – 1999.
Since this bias towards higher RHice values is in contradiction to physical understanding and to observations made inside equilibrated cirrus clouds, an in-depth reanalysis of the MOZAIC data set was conducted. During this effort an error in the laboratory calibrations applied from year 2000 on was found, whereas the data of the period from 1994 to 1999 were found to be correct. The complete data set for the period 1994–2009 was reanalysed with the corrected calibration factors. As a result, the PDF of the

corrected data set exhibit their maximum at RHice ≅ 100%, in agreement with PDF of RHice presented in the literature. Applied correction schemes and a revised error analysis are presented along with the PDF of RHice for various regions of the global UTLS.

6.2 - The evaluation of MOZAIC/IAGOS Humidity Devices during airborne field studies CIRRUS-III and AIRTOSS-ICE

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Presenter : Patrick Neis,

Water vapour is one of the most important parameters in weather prediction and climate research. Particularly the interaction between the water vapour in the upper troposphere and lowermost stratosphere (UTLS) and tropopause dynamics are not well understood. The difficulty is that the abundance of upper tropospheric humidity (UTH) is highly variable on spatial and temporal scales that cannot be resolved, neither by the global radiosondes network nor by satellites.

Since 1994, data with high spatial and temporal resolution for relative humidity are provided by the in-situ measurements aboard civil passenger aircraft from the MOZAIC/IAGOS-programme (www.iagos.org). The data set emerging from this long-term observation effort builds the backbone of the ongoing in-situ UTH climatology and trend analyses.

In order to assess the validity of the long-term water vapour data and its limitations, an analysis of the humidity data sets of two field campaigns is presented. The validation of applied measurement methods, i.e. the MOZAIC/IAGOS Humidity Devices, is valued on the basis of the aircraft campaigns CIRRUS-III (2006) and AIRTOSS-ICE (2013), where research-grade water vapour instruments were operated simultaneous to the MOZAIC/IAGOS Humidity Devices.

The performance of the MOZAIC Humidity Device (MHD; operated from 1994 to 2013 on MOZAIC aircraft) and the advanced IAGOS Humidity Device (IHD; operated since 2011 on IAGOS aircraft) are explored in clear sky, in the vicinity of and inside cirrus clouds as a blind intercomparison to the reference measurement instrument FISH (Fast In-Situ Hygrometer).

From these intercomparison distinct values for precision and limit of detection are determined for the two sensors MHD and IHD, respectively. The qualification of the capacitive humidity sensor for the use in long-term observation programmes is successfully demonstrated and the continuation of high data quality is confirmed for the transition from MHD to IHD.

6.3 - Processes controlling H2O in the upper troposphere / lowermost stratosphere: An analysis of eight years of monthly measurements by the IAGOS-CARIBIC observatory

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Presenter : Andreas Zahn

The water vapor (H2O) data obtained by the IAGOS-CARIBIC passenger aircraft since 2005 at 10-12 km altitude is analyzed. A precise description of the (seasonally varying) vertical distribution of H2O from the upper troposphere (UT) via the extra-tropical tropopause mixing layer (exTL) into the background lowermost stratosphere (LMS) is given. Furthermore the transport pathways and air flows determining H2O in the UT/LMS are analyzed. Compared to mid/longer-lived trace gases, H2O is strongly variable in the UT and exTL and additionally undergoes considerable seasonal variation with peaking concentrations in summer. The seasonal variation of H2O is in phase at all altitudes from the UT up to ~4 km above the mid-latitude tropopause. The transport and dehydration pathway of air starting in the lower troposphere and ending at 10-12 km altitude is reconstructed based upon (i) the potential temperature (theta), (ii) the relative humidity w.r.t. ice (RHi), and (iii) the starting point of back trajectories as function of the H2O concentration and altitude relative to the tropopause. The instantaneously measured RHi was found to be primarily determined by vertical movement, i.e. cooling during ascent/expansion and warming during descent/compression before sampling. Using back trajectory analyses, the data show with surprising clarity that H2O and RHi at 10-12 km altitude are controlled by three dominant transport/dehydration pathways: (i) the Hadley cell circulation, i.e. convective uplift in the tropics and pole-ward directed subsidence from the tropical tropopause layer (TTL) with observed RHi down to 2%, (ii) warm conveyor belts and mid-latitude convection transporting wet air into the UT with RHi usually above 60%, and (iii) the Brewer-Dobson lower and higher branches supplying the 10-12 km altitude region with very dry air with RHi values down to 1%.

6.4 - First regular in-situ observations of the isotopic composition of water vapor (HDO/H216O) in the upper troposphere within IAGOS-CARIBIC

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Amongst water vapor (H2O) concentration mesurements, a new dimension for a better understanding of the hydrological cycle of the atmosphere opens the analysis of the isotope ratios of H2O. These are modified, i.e. fractionated, basically by each process in which water vapor is involved, in particular during H2O phase changes such as the formation of clouds. In the troposphere, the H2O isotope ratios trace transport pathways and the type of (lofting) process which bring water vapor from the Earth surface to sampling altitude.

Since April 2010, a tunable diode laser absorption spectrometer (ISOWAT) measures the isotope ratio HDO/H216O onboard the IAGOS-CARIBIC aircraft (given in the δ-notation as difference to an isotope standard). The data reflect the large impact of deep convection in the tropics which leads to reduced H2O isotope fractionation in the tropical upper troposphere. The relevant effects of ice lofting, mixing processes and condensate retention are assessed. Comparison to a Rayleigh model that only considers the

isotopologue-dependent vapor pressure and kinetic fractionation processes during cloud formation, we find a mean enhancement of δD(H2O) by more than 100 ‰ above the three continents Africa, South America, and East Asia. We further develop a meridional profile of δD(H2O) in the upper troposphere and assign deviations from the mean profile to certain processes, e.g. isotopically strongly depleted δD(H2O) to air masses processed in an Atlantic hurricane which may be due to a super Rayleigh fractionation process.

6.5 - Role of deep convection in moistening the stratosphere: LES of Hector the Convector

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Presenter : Thibaut Dauhut,

The trend of stratospheric water vapour during the past decades is not correctly reproduced by current GCMs. This may be due to lack of representation of rapid water transfers from troposphere to stratosphere. Our modeling study focused on a particular case of tropical deep convection which takes an active part in this transport. We aimed at understanding its dynamics and the stratosphere moistening processes. We selected a Hector thunderstorm observed on 30 November 2005 over Tiwi Islands, Australia, during the SCOUT-O3 field campaign. Plumes of ice particles reaching 19 km altitude were measured by lidar aboard the Geophysica stratospheric aircraftt. We performed a Large-Eddy Simulation of Hector (100 m horizontal resolution) using cutting-edge computing resources, as well as a series of simulations with coarser and coarser horizontal resolutions, from 200 m to 1600 m. Strong morning sea breeze deviated boundary layer westerlies and led to intense convergence of humid air over Tiwi Islands. Deep convection triggered around 1 pm. The most intense upward transport started straight after and lasted around 2 hours. Updraft cores statistics showed that stronger upward winds in the boundary layer and at the cloud base lead to weaker cloud fraction aloft and higher hydrometeor content in updraft cores. Turbulence analysis also showed that the faster the updrafts are, the lower the dilution and the more efficient the vertical transfer of water. As a result, some updrafts overshooted the tropopause and carried ice crystals in the stratosphere. Overshoots in the LES compared well with the observations. Part of the ice particles precipitated then whereas the remainder sublimated in lower stratosphere. The consequent vapour pockets were transported and diluted within the stratosphere by easterlies. In total, 2776 tonnes of water were transported from troposphere to stratosphere. Associated net hydration of the lower stratosphere was found with a 16 % increase in water vapour. Upscaling this result using 5-year TRMM radar observations we found that deep convection penetrating 380 K potential temperature level could represent 18 % of troposphere-stratosphere total water flux. While moistening appeared to be robust with respect to the grid spacing used, grid spacing on the order of 100 m may be necessary for a reliable estimate of hydration. This study was supported by the StratoClim project.

6.6 - Cloud Property Measurements from Commercial Aircraft

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Presenter : Karl Beswick

A compact (500 cm3), lightweight (500 g), near-field, single particle backscattering optical spectrometer is described that mounts flush with the skin of an aircraft and measures the concentration and optical equivalent diameter of particles from 5 to 75 µm. The backscatter cloud probe (BCP) was designed as a real-time qualitative cloud detector initially only for data quality control of trace gas instruments developed for the climate monitoring instrument packages that are being installed on commercial passenger aircraft as part of the European Union In-Service Aircraft for a Global Observing System (IAGOS) program (http://www.iagos.org/). Subsequent evaluations of the BCP measurements on a number of research aircraft, however, revealed that it is capable of delivering semi-quantitative dust and cloud particle data products including size distributions, liquid water content and other information on dust and cloud properties. We demonstrate the instrument's capability for delivering useful long-term climatological, as well as aviation performance information, across a wide range of environmental conditions.

The BCP has been evaluated by comparing its measurements with those from other cloud particle spectrometers on research aircraft and six BCPs are currently flying on European and Asian commercial A340/A330 Airbus passenger airliners. The design of the BCP is described in this presentation, along with an evaluation of measurements made on the research and commercial aircraft. Results from more than 8,000 hours of airborne measurements by the BCP on five Airbus A-340s (Lufthansa, China Airlines, Air France and Iberia) operating on routine global traffic routes show that more than 700 hours of cloud data, in > 20000 clouds wider than 25 km, have been recorded at normal cruise altitudes (> 10 km) since September, 2011. These data are a valuable contribution to data bases of cloud properties, including sub-visible cirrus, in the upper troposphere and useful for validating satellite retrievals of cloud water and effective radius as well as providing a broader, geographically and climatologically relevant view of cloud microphysical variability useful for improving parameterizations of clouds in climate models. They are also useful for monitoring the vertical climatology of clouds over airports, especially those over mega-cities where pollution emissions may be impacting local and regional climate.

6.7- Development of an airborne dual-channel hygrometer based on photoacoustic spectroscopy

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Presenter : David Tatrai

Application oriented photoacoustic (PA) spectroscopy related research and measuring system development was started at the University of Szeged in the mid-1990's. One of the first instruments was a moisture measuring system for natural gas industry. Based on this system and by using newly developed telecommunication DFB diode lasers, a new dual channel hygrometer was developed to be applied within the CARIBIC project as of 2005. The dual-channel layout makes it possible to measure water vapor and total water (the sum of vapor and cloud water/ice) simultaneously when connected to an appropriate sampling system. With the development of a new control electronics enabling several self-checking methods, an upgraded version of the CARIBIC system was made by 2009 and has been applied since

2010. The system used within CARIBIC project can be viewed as prototype as providing only raw signals that are continuously calibrated using a chilled mirror hygrometer working in parallel.
Goal of a EUFAR project was the development of a reliable standalone system. The first important development was the improvement of the wavelength setting method of the DFB laser. As a result the uncertainty of the wavelength is less than 40fm, which corresponds to less than 0.05% of PA signal uncertainty. This PA signal uncertainty is lower than the noise level of the system itself. The other main development was the improvement of the concentration determination algorithm. For this purpose several calibration and data evaluation methods were developed. The combination of the latest ones have made the system traceable to the humidity generator applied during the calibration within 1.5% relative deviation or within noise level whichever is greater.
The improved system was several times tested at the Environmental Simulation Facility (Forschungszentrum Jülich, Germany) in pressure and humidity ranges possible in in-service aircraft operation (150-950 mbar and 1-15000 ppmV). Furthermore the system was tested and compared to other instruments in three flight campaigns based on a research aircraft (Learjet 36A). The test results both in the laboratory and both in the field shows that the developed system is a promising tool for further airborne environment research.
Based on the introduced results now a new more compact, faster and more flexible system is being developed based on a new process control and data acquisition electronics.
The past and actual development process with the main results will be presented.

POSTERS

P 1.1 - First simultaneous space measurements of atmospheric pollutants in the boundary layer from IASI: a case study in the North China Plain

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Presenter : Anne Boynard

An extremely severe and persistent smog episode occurred in January 2013 over China. The levels of air pollution have been dangerously high, reaching 40 times recommended safety levels and have affected health of millions of people. China faced one of the worst periods of air quality in recent history and drew worldwide attention. This pollution episode was caused by the combination of anthropogenic emissions and stable meteorological conditions (absence of wind and temperature inversion) that trapped pollutants in the boundary layer. To characterize this episode, we used the IASI (Infrared Atmospheric Sounding Interferometer) instrument onboard the MetOp-A platform. IASI observations show high concentrations of key trace gases such as carbon monoxide (CO), sulfur dioxide (SO2) and ammonia (NH3) along with ammonium sulfate aerosol. We show that IASI is able to detect boundary layer pollution in case of large negative thermal contrast combined with high levels of pollution. Our findings demonstrate the ability of thermal infrared instrument such as IASI to monitor boundary layer pollutants, which can support air quality evaluation and management.

P 1.2 - Preliminary analysis of long-term variability of upper tropospheric humidity in the northern

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We use 30 years of intercalibrated brightness temperature data from the High-Resolution Infrared Radiation Sounder (HIRS) onboard the NOAA series of satellites to produce a 30 year data set of upper tropospheric humidity with respect to ice (UTHi). The upper tropospheric humidity (a mean relative humidity in the upper troposphere roughly between 300 and 500 hPa) is derived from the measured brightness temperatures in HIRS channels 12 and 6 using the formula proposed by Jackson and Bates (2001). We have produced daily files of UTHi for each NOAA satellite that carries the HIRS instrument, from which we have calculated monthly means in 2.5 × 2.5 degrees resolution for the northern mid-latitudes (30°–60°N). We present a preliminary analysis of climatology and long-term vari-ability of UTHi in the past three decades over the northern mid-latitudes.

P 2.1 - Atmospheric conditions associated with high and low summertime ozone concentrations in the lower troposphere and the boundary layer over some eastern Mediterranean airports

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Presenter : Pavlos Kalabokas

The data obtained by the MOZAIC (Measurement of Ozone and Water Vapor by Airbus in Service Aircraft) program revealed enhanced ozone mixing ratios in the lower troposphere over the Eastern Mediterranean during summertime in comparison to Central Europe. In order to further evaluate the findings, vertical profiles of ozone measured in the period 1994–2008 in the framework of the MOZAIC project over the Eastern Mediterranean basin (Cairo, Tel-Aviv, Heraklion, Rhodes, Antalya) were analyzed, focusing on the free lower troposphere (1.5–5 km) and on the boundary layer (0-1.5km). In general, the maximum vertical ozone concentrations during days of highest ozone mixing ratios over all the examined Eastern Mediterranean airports are observed above the boundary layer, at 2-3km altitude. Within the boundary layer, ozone is decreased on average in all airports, especially in Tel-Aviv and Cairo, which might be attributed to boundary layer processes such as nitrogen oxides originating from local urban pollution, atmospheric particles (e.g. desert dust) or dry deposition on the ground.

Based on this analysis, it turns out that the lower tropospheric ozone variability over the Eastern Mediterranean area is controlled mainly by the synoptic meteorological conditions, combined with local topographical and meteorological features. In particular, the highest ozone concentrations in the lower troposphere are associated with large scale subsidence of ozone-rich air masses from the upper troposphere under anticyclonic conditions, whereas the lowest ozone concentrations are associated with low pressure conditions inducing uplifting of boundary layer air with low ozone and high relative humidity into the lower troposphere. Other meteorological processes within the boundary layer (e.g. sea-breeze) might have an important influence on the ozone variability, depending on the particular location characteristics. The results of our analysis show that the influence of tropospheric ozone on both the boundary layer and surface ozone values, mainly through the process of atmospheric subsidence, is quite variable among the examined Eastern Mediterranean airports with the highest impact detected over the Aegean Sea airports of Heraklion and Rhodes, which seem to be more exposed to the free tropospheric influence due probably to the combined effects of synoptic meteorology and topography.

P 2.2 - Ozone and carbon monoxide distributions in the African upper troposphere: 5 years of pollution observations

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Presenter : Bastien Sauvage

Between 2006 and 2011 more than 2,000 MOZAIC-IAGOS (http://www.iagos.org/) daily flights were performed from Namibia (Windhoek) to Europe. In the meantime IASI ozone observations and MLS CO are used for a larger sampling of the African upper troposphere. The study updates our knowledge on the upper tropospheric seasonal distributions of ozone, water vapour and carbon monoxide along hemispheric and meridional transects over Africa. First salient aspects of these seasonal distributions were analysed by Sauvage et al. (JGR, 2007) and modelled with a two-dimensional approach by Saunois et al. (JGR, 2008) and a global 3D CTM by Barret et al. (ACP, 2008). Contributions of surface emissions injected by convection in the Hadley cells over continental Africa and zonally transported with upstream source regions as far as Asia or South America are investigated through a modelling approach using the Lagrangian FLEXPART model coupled with emission inventories.

P 3.1 - Measuring Aircraft Engine Soot Emissions with the CAPS PMex Extinction Monitor

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The optical properties of plumes emitted from a high bypass turbofan aircraft engine (V2527) and a sector rig of an aircraft engine combustor (Pratt and Whitney) were measured using a combination of the CAPS PMex particle extinciton monitor (operated at a wavelength of 630 nm) and a multi-angle absorption spectrometer(MAAP). These meaurements indicated that the single scattering albedo (SSA) of aircraft engine soot is on the order of 0.05, a result that is compatible with in situ measurements of the particle size distributions which indicated that the soot particle volume-weighted mobility diameter distribution peaked well below 100 nm. These results are in agreement with the only other measurements of the optical properties of aircraft engine soot emissions (Petzold, et al. J. Geophys. Res. 22:171-181 (1999)).

P 3.2 - Development and Evaluation of Novel and Compact Hygrometer for Airborne Research (DENCHAR): In-Flight Performance During AIRTOSS-I/II Research Aircaft Campaigns (RNTD

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Presenter : Herman G.J. Smit

Water vapour is one of the most important parameters in weather prediction and climate research. Accurate and reliable airborne measurements of water vapour are a pre-requisite to study the underlying processes in the chemistry and physics of the atmosphere. Presently, no airborne water vapour sensor exists that covers the entire range of water vapour content of more than four order of magnitudes between the surface and the UT/LS region with sufficient accuracy and time resolution, not to speak of the technical requirements for quasi-routine operation.

In a joint research activity of the European Facility for Airborne Research (EUFAR) programme, funded by the EC in FP7, we have addressed this deficit by the Development and Evaluation of Novel and Compact Hygrometer for Airborne Research (DENCHAR), including the sampling characteristics of different gas/ice inlets. The new instruments using innovative detecting technics based on tuneable diode laser technology combined with absorption spectroscopy (TDLAS) or photoacoustic spectroscopy: (i) SEALDH based on novel self-calibrating absorption spectroscopy; (ii) WASUL, based on photoacoustic spectroscopy; (iii) two commercial WVSS-II, also a TDLAS hygrometer, but using 2f-detection technics.

We will present an evaluation of the in-flight performance of three new hygrometer instruments, which is based on the results obtained during two research aircraft campaigns in 2013 as part of the AIRTOSS (AIRcraft Towed Sensor Shuttle) experiments. Aboard a Learjet 35A research aircraft the new instruments were operated side by side with the well established Fast In-Situ Hygrometer (FISH), which is based on Lyman (alpha) resonance fluorescence detection technics and calibrated to the reference frost point hygrometer MBW DP30 at the ground based FISH calibration bench. We also will report on the performance of the different inlet systems used during both campaigns under different cloud and non-cloud conditions.

The instruments showed very good and consistent in-flight performance over a wide range of humidity levels covering more than three orders of magnitudes between 10 ppmv and 20,000 ppmv water vapour mixing ratios. Flight by flight the DENCHAR-instruments showed a very consistent behaviour among each other as well as against the FISH-instrument. Within their uncertainty range (5-10%) all instruments agreed very well and were traceable within about 10% uncertainty to the accurate DP30 (MBW) frost point hygrometer.

P 3.3 - The IAGOS GHG package: a measurement system for continuous airborne observations of CO2, CH4, H2O and CO

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Presenter : Annette Filges

Abstract for
- Session 4: Recent and New Technical Developments
- or Session 5: Monitoring Atmospheric Composition, Climate and Air Quality

Within the framework of IAGOS-ERI a cavity ring-down spectroscopy (CRDS) based measurement system for greenhouse gases was designed, tested, and qualified for deployment on commercial airliners. The design meets requirements regarding physical dimensions (size, weight), performance (long term stability, low maintenance, robustness, full automation) and safety issues (fire prevention regulations). The system uses components of a commercially available CRDS instrument (G2401-m, Picarro Inc.) mounted into a frame suitable for integration in the avionics bay of the Airbus A-340.

To enable robust and automated operation of the IAGOS GHG package over six-month deployment periods, numerous technical issues had to be addressed. An inlet system was designed to eliminate sampling of larger aerosols, ice particles, and water droplets, and to provide additional positive ram-pressure to ensure operation throughout an aircraft altitude operating range up to 12.5 km without an upstream sampling pump. Furthermore, no sample drying is required as the simultaneously measured water vapor mole fraction is used to correct for dilution and spectroscopic effects. This also enables the collection of science-quality water vapor measurements throughout the atmosphere. To allow for trace gas measurements to be fully traceable to WMO scales, a two-standard calibration system has been designed and tested that periodically provides calibration gas to the instrument during flight and on ground for each six-month deployment period.

The first of the IAGOS GHG packages is scheduled for integration in spring 2014. The aim is to have five systems operational within four years, providing for regular, long-term GHG observations with near-global coverage.

We present results from recent test flights and laboratory tests that document the performance for GHG and water vapor measurements. Furthermore, future applications of the IAGOS-GHG data stream, provided in near-real-time via SatCom to the weather prediction centres, will be discussed, including the validation of remote sensing and of forward transport modeling of GHGs, assessment of the impact of transport uncertainties, and the use as input for inverse modeling.

P 3.4 - The New NOx Instrument Deployed in IAGOS – Design and First Results

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Presenter : Andreas Volz-Thomas

Nitrogen oxides (NOx, i.e., NO and NO2) play an important role in atmospheric chemistry related to ozone and oxidation capacity (OH and NO3 radicals). They are observed at mixing ratios ranging from a few ppt in the remote marine boundary layer to a few ppb in continental background and up to several hundreds of ppb in urban environments (c.f. Emmonds et al., 1997, 2000). Sources of NOx are combustion processes (power generation, transportation, biomass burning, wild fires), soil emissions, lightning and photochemical production from N2O in the stratosphere. The most important sources to the upper troposphere are, lightning, transport from the boundary layer and aircraft emissions. Measurements of NOx in the free troposphere and lower stratosphere are important for understanding the local photochemistry and for assessment of the impact of aircraft on the budgets of greenhouse gases such as ozone and methane. Because of the strong influence of UV radiation on the ratio between NO and NO2, it is important to measure both species or at least the sum of both. The NOx instrument (P2b) deployed in IAGOS-CORE is designed for autonomous operation during several months. The measurement principle is chemiluminescence (CLD) with photolytic conversion of NO2 at wavelengths of 395 ± 5 nm. The poster describes the design and capability of the instrument and highlights examples of measurements made aboard an Airbus A340-300 operated by Lufthansa (D-AIGT). Because of the limitations provided by the physical boundary conditions for installation in the IAGOS rack and the requirement of long deployment periods, the instrument employs much lower flow rates than instruments flown on research aircraft and, hence, has a much lower sensitivity (0.7-1 cps/ppt). Consequently, the main focus of the NOx measurements made in IAGOS-CORE is on tropospheric profiles and on the detection of plumes arising from long range transport.

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P 3.5 - Development and first application of the IAGOS aerosol sensor package

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The natural variability of aerosol particles both in space and time is still one of the largest sources of uncertainty in global climate models. While ground-based networks for in-situ and remote-sensing measurements are well developed (see www.actris.net for more information), no comparable infrastructure is available for in-situ measurements of aerosol properties in the upper troposphere and lowermost stratosphere (UTLS).
In the framework of IAGOS, a robust instrument for the routine measurement of the aerosol particle size distribution and the integral numbers of particles and for non-volatile particle cores aboard long-haul in-service aircraft was developed. The aerosol size information for the so-called accumulation mode covers the range of particles available for the formation of liquid water and ice clouds. The total number concentration provides information on gas-to-particle conversion and particle nucleation at flight altitude level. The number concentration of the non-volatile particle cores yields complementary information on the anthropogenic contribution to the atmospheric aerosol burden. Also, non-volatile soot particles emitted by aircraft are thought to play a role in the indirect aerosol effect on climate by acting as ice nuclei for cirrus particles.
The aerosol package IAGOS P2c is designed for automated, low-maintenance operation. It contains one instrument for measuring particles of the aerosol accumulation mode by light scattering techniques (optical particle counter; Grimm OPC model 1.129: > 250 nm in diameter), and a two-channel instrument for the measurement of particle number concentrations down to the nm scale by means of condensation particle counters (CPC; Grimm CPC model 5414: > 5 nm in diameter). The dual-channel set-up permits the separation of total aerosol particles and non-volatile aerosol particles by applying a thermal denuder while the reported upper size limits of approx. 3 µm in diameter are determined by the maximum diameter of particles passing the inlet system.
Here we present the performance analysis of IAGOS P2c conducted in the Jülich Aerosol Lab facilities. First airborne measurements of this device will also be presented from the RACEPAC Campaign where IAGOS P2c is operated onboard the POLAR 6 research aircraft of Alfred Wegener Institute (AWI). This field study has its focus on arctic clouds and haze over the Beaufort Sea near Inuvik, located at the Northern Territories, Canada, and takes place in April/May 2014. The instrument passes the certification and the corresponding test-flight. Here data of these flights over the North-Sea close to Bremerhaven in November 2013 and hopefully first preliminary data of the Campaign will be shown.
Acknowledgements
This work was supported by the EC project IGAS (Grant Agreement No. 312311) and by the Federal Ministry of Education and Research, Germany in IAGOS–D (Grant Agreement No. 01LK1223A). We kindly express our gratitude to the Leipzig Institute for Meteorology and Alfred Wegener Institute Bremerhaven coordinating the RACEPAC campaign for giving us the opportunity to join the campaign.

P 4.1 Water vapor transport in the lower stratosphere during summer linked to Asian monsoon and horizontal transport

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Presenter : Felix Ploeger

We compare global water vapor observations from Microwave Limb Sounder (MLS) and simulations with the Lagrangian chemistry transport model CLaMS (Chemical Lagrangian Model of the Stratosphere) to investigate the pathways of water vapor into the lower stratosphere during northern hemisphere (NH) summer. We find good agreement between the simulation and observations, with an effect of the satellite averaging kernel especially at high latitudes. The Asian and American monsoons emerge as regions of particularly high water vapor mixing ratios in the lower stratosphere during boreal summer. In NH mid latitudes and high latitudes, a clear anticorrelation between water vapor and ozone daily tendencies reveals a large region influenced by frequent horizontal transport from low latitudes, extending up to about 450K during summer and fall. Analysis of the zonal mean tracer continuity equation shows that close to the subtropics, this horizontal transport is mainly cause d by the residual circulation. In contrast, at higher latitudes, poleward of about 50N, eddy mixing dominates the horizontal water vapor transport. Model simulations with transport barriers confirm that almost the entire annual cycle of water vapor in NH mid latitudes above about 360K, with maximum mixing ratios during summer and fall, is caused by horizontal transport from low latitudes. In the model, highest water vapor mixing ratios in this region are clearly linked to upward transport within the Asian monsoon in the subtropics and subsequent poleward horizontal transport.

P 4.2 - Acetone in the UT/LMS

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Presenter : Marco Neumaier

Onboard the IAGOS-CARIBIC passenger aircraft, acetone as one of the most abundant oxygenated volatile organic compounds (VOCs), along with acetonitrile and methanol, is measured with a proton-transfer-reaction mass spectrometer, which constitutes the most complex instrument available in IAGOS. Thereby, since 2005 the largest in-situ dataset of acetone from the upper troposphere / lowermost stratosphere (UT/LMS) is gathered. Acetone shows a pronounced seasonal variation that maximizes in summer with mixing ratios up to ~1100 pptv at the tropopause (TP), mainly caused by the then peaking emission of biogenic acetone and enhanced secondary production from e.g. propane oxidation. Folding the acetone CARIBIC data with 5-day back trajectories from the ECMWF model (i) increases the global coverage considerably and (ii) reveals further features affecting the UT like the Asian summer monsoon or biomass burning events over Africa in winter.
A strong linear correlation between acetone and CO was found during sporadic research aircraft campaigns (e.g. Reus et al. 2003). Based on the IAGOS-CARIBIC data, this study was considerably expanded to better understand the sources of acetone, its chemical processing and its surprisingly good CO correlation. The highest and most variable [acetone] / [CO] correlation slopes are found over North America in summer (~35±25 pptv/ppb), while over Asia significantly lower values show up (~15±10 pptv/ppb). In winter in both regions comparable values of ~10±5 pptv/ppbv are observed. Since in the UT

the lifetime of acetone is a factor of ~3 shorter than that of CO, the correlation slope is expected to decrease in aged air masses.
Acetone was in the past assumed to be a dominant HOx source in the dry extra-tropical upper troposphere (ex-UT). However the measurement of significantly smaller photodissociation quantum yields of acetone by Blitz et al. 2004 challenged that assumption. Based on diverse CARIBIC trace gas data and non-observed parameters taken from the model EMAC, we quantified the HOx source in the UT/LMS from (photo-) oxidation of acetone (Neumaier et al., revised to GRL). The findings are contrasted to HOx production from ozone photolysis, overall the dominant tropospheric HOx source. We show that HOx production from acetone (photo-) oxidation reaches up to ~92% of the HOx source from ozone photolysis in October in the UT and on average ~54% in summer, i.e. acetone is a significant source of HOx in the UT/LMS.

P 4.3 - An insight on Asian pollution transport to the UTLS over the region of the Asian Monsoon Anticyclone with MOZAIC-IAGOS data

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Presenter : Yasmine Bennouna

With high anthropogenic emissions and natural biomass burning from wildfires, South and East Asia represent the largest source region of atmospheric pollution. During the Asian Summer Monsoon (ASM), the Asian Monsoon Anticyclone (AMA) is a significant dynamical feature of synoptic circulation in the upper troposphere-lower stratosphere (UTLS). A number of recent studies based on satellite observations have shown that carbon monoxide (CO) and ozone (O3) exhibit respectively low and high concentrations whithin the AMA. These features are related to the fast convective uplift of polluted boundary layer air masses to the upper troposphere (UT). Once in the UT, the pollution is trapped within the strong anticyclonic circulation and slowly ascends to the lower stratosphere.
Our study exploits the potential of the MOZAIC-IAGOS in-situ observations from regular Asia-Europe and Asia-Middle East flights, to document the UTLS composition within and around the AMA. The studied area is chosen to encompass mainly the Indian subcontinent, the Arabian Peninsula and most surrounding regions of South-East Asia (0N-50N,10E-150E). Both profiles and cruise data for O3 and CO available in the region are examined in relation with meteorological fields from the MERRA reanalysis. To complement and support the information provided by IAGOS, data from other sources are also presented, including UTLS satellite observations from IASI (Infrared Atmospheric Sounding Interferometer) and MLS (Microwave Limb Sounder). Furthermore, simulations with the GEOS-Chem model are performed for the year 2009 to quantify the O3 budget in the Asian UTLS during the ASM. The 2009 IAGOS observations are used to validate the CO and O3 distributions simulated by the model over South-Asia.

P 4.4 Case studies of ozone stratospheric intrusions events over South America

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Presenter : Marcia Yamasoe

Ozone concentration enhancement due to stratosphere-troposphere exchange was analyzed based on MOZAIC-IAGOS flights over Caracas, Venezuela, São Paulo and Rio de Janeiro, Brazil. The analyzed databases consisted on data collected during landing and taking off, i.e. from the surface up to 200 hPa. In São Paulo and Rio de Janeiro, the identification of layers for which ozone enhancements were due to stratospheric intrusions was performed based on potential vorticity (PV). Layers with absolute PV above 2 PVU were considered, allowing a 15 hPa threshold. In Caracas, this PV threshold could not be used, since some ozone enhanced concentrations at upper tropospheric layers were also identified, but with lower PV values. FLEXPART back-trajectories confirmed the stratospheric origin of such airmasses. Satellite imagery corroborated the origin. In the water vapour channel images, very dry airmasses were observed and enhancements of total ozone column upstream from the flow were also identified. In São Paulo and Rio de Janeiro, the intrusions events were associated to the subtropical jet, with maximum contribution during winter time (June, July and August). In São Paulo, during this season, around 200 hPa, 21% of all the analyzed data got contribution from the stratosphere, increasing the mean ozone concentration at that pressure level by about 11.4 ppbv, compared to the climatological value estimated without this source contribution.

P 5.1 - Temporal Variation and Source Apportionment of Atmospheric Mercury Concentrations at Two Regional Background Sites in China

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Presenter : Lei Zhang

Long-term continuous monitoring of speciated atmospheric mercury concentration is an important approach to study the characteristics and sources of mercury pollution in key regions. Gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particulate-bound mercury (PBM) were measured at two regional background Sites (Miyun, Beijing and Chongming, Shanghai) respectively in North China Plain (NCP) Region and Yangtze River Delta (YRD) Region in China. The average total mercury concentrations of Miyun site and Chongming site were 3.33 and 2.65 ng/m3, respectively, about 1.5–2 times of the background mercury concentration of Northern Hemisphere. The highest average GEM concentration occurred in summer at Miyun site while in winter at Chongming site, indicating the influence of mercury emission sources in NCP Region through long-range transport of polluted air mass. The peak value of the RGM concentration occurred in autumn at Miyun site while in summer at Chongming site, revealing that Miyun was more affected by anthropogenic sources while Chongming was more affected by natural sources. The highest PBM was found in autumn at both sites, which was mainly caused by the adverse diffusion conditions in autumn. The diurnal distributions of GEM and PBM had the highest values in the late night at Miyun site while in the morning at Chongming site, which showed that the accumulation time of mercury pollution was longer in coastal area than in inland area. The ratio of GEM to CO indicated that residential boilers played an important role in the elevation of GEM in winter. The ratio of RGM to O3 could be an indicator of the contribution of local primary sources. The ratio of (PBM/PM2.5)/RGM was positively correlated with temperature at Chongming site. HYSPLIT back-trajectory analysis identified the local and regional mercury emission sources in heavy pollution episodes at these two sites. The results from the potential source contribution function (PSCF) model based on the HYSPLIT results indicated that the air mass transported predominantly from the northwest (Mongolia and Xinjiang area) contributed to the elevated atmospheric mercury at Miyun site in autumn and winter, while NCP Region and the north part of YRD Region were the major source areas for mercury pollution in spring and summer. The total mercury concentration at Chongming site was mainly contributed by YRD Region in summer while by both NCP Region and YRD Region in autumn and winter.

P 5.2 - Variations of tropospheric methane over Japan during 1988–2010

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Mixing ratios of greenhouse gases and related trace gases have been measured using chartered and commercial aircraft in the lower to upper troposphere (LT and UT) over Japan by Tohoku University. We present variations of CH4 during 1988–2010. The analysis is aided by simulation results using an atmospheric chemistry transport model (i.e. ACTM). Tropospheric CH4 over Japan shows altitude-dependent interannual and seasonal variations, reflecting differences in air mass origins at different altitudes. The long-term trend and interannual variation of CH4 in the LT are consistent with previous reports of measurements at surface baseline stations in the northern hemisphere. However, those in the UT show excursions from those in the LT. In the UT, CH4 mixing ratios show seasonal maximum in August due to efficient transport of air masses influenced by continental CH4 sources, while LT CH4 reaches its seasonal minimum during summer due to seasonally maximum chemical loss. Vertical profiles of the CH4 mixing ratios also vary with season, reflecting the altitude-dependent seasonal cycles. In summer, transport of CH4-rich air from Asian regions elevates UT CH4 levels, forming the uniform vertical profile above the mid troposphere. On the other hand, CH4 decreases nearly monotonically with altitude in winter–spring. The ACTM simulations with different emission scenarios reproduce general features of the tropospheric CH4 variations over Japan. Tagged tracer simulations using the ACTM indicate substantial contributions of CH4 sources in South Asia and East Asia to the summertime high CH4 values observed in the UT. This suggests that our observation data over Japan are highly valuable for capturing CH4 emission signals, particularly from the Asian continent.

P 5.3 - Ozone and carbon monoxide in tropospheric columns: a climatology (24°N-50°N) derived from MOZAIC/IAGOS on a 1994-2011 peri od.

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Presenter : Régina M. ZBINDEN

We propose to complete the new tropospheric climatology of ozone (O3) and carbon monoxide (CO) in the northern hemisphere derived from the MOZAIC/IAGOS program in terms of seasonal-mean profiles and the monthly-mean column contents, with in addition the boundary layer, mid-troposphere and upper-troposphere partial columns contents (Zbinden et al., 2013).

This work is based on the huge amount of O3 and CO MOZAIC/IAGOS in-situ observations performed by the instruments on board of commercial aircraft (Airbus A340). We select only the ascent and descent part of the flights which mostly document the tropospheric layers with a 50 m vertical resolution. The new and comprehensive climatology is refined compared to Zbinden et al. (2006), giving for each profile a complete description of the hole troposphere, i.e. from the surface up to the dynamical tropopause fixed at 2 pvu (i.e. excluding stratospheric contaminations but not stratospheric intrusions). The potential vorticity pressures are extracted from the operational European Centre for Medium-Range Weather Forecast (ECMWF)

analyses (T213), are interpolated for the specific aircraft position with a 150m vertical resolution and are available on the MOZAIC/IAGOS data base. The methodology, extensively explained in Zbinden et al. (2013), will be recapitulated.

The period to derive the results from the calibrated data now extend from 08/1994 to 02/2011 for O3 and from 01/2001 to 02/2011 for CO, including more than 50 000 ozone profiles. The sites located from [24-50° N] and [119° W – 140° E] are updated and improved c ompared to Zbinden et al. (2013). The presentation highlights the most interesting characteristics of the troposphere for both species.

Our climatology provides an accurate insight into the troposphere nearby cities with extended coverage compared to the one of ozonesondes network, whereas satellite data map regularly wide areas never elsewhere documented. We finally compare the climatology using remote sensing and in-situ data and point out the discrepancies and the benefits for tropospheric chemical studies.

P 5.4 - Aerosol sources over Jaipur in Northwestern India from ground based AERONET measurements

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Presenter : Sunita Verma

The aerosol properties retrieved from AERosol RObotic NETwork (AERONET) measurements during the period 2009 to 2012 over Jaipur (27.3 N, 94.6 E), Northwestern India are utilized for the first time to identify the types of aerosols. A novel approach to consider the appropriate threshold of aerosol optical thickness (&#964 at 500 nm (τ500) and Angstrom exponent (&#945 in the spectral band 440-870 nm has been applied for the identification process. Five prevailing aerosol classes are identiﬁed: Desert dust, Biomass, Maritime, Semi-arid Background and mixed aerosols. Dust and semi-arid background aerosol are the most common at Jaipur (13.6% and 34.7% of the cases, respectively), with a wide variability in both τ and α. Only in about 8.4% of the cases can aerosol be classiﬁed as maritime however mixing with other aerosol is also substantial.
Spectral optical thickness and the refractive index estimated at visible and near-infrared wavelengths is also used to account for the nature of atmosphere aerosols and being compared with other AERONET sites located in India based upon their geographical distribution and extensive data availability. Simultaneously, the single scattering albedo of dust is also inferred for all the considered sites. The comparative analysis with other sites over India suggests that Jaipur arid background is more scattering in nature than Northern and Western regions in India. Seasonally, the absorption is least in summer and most in winter over the site.
Key Words: Aerosols, Optical Thickness, Desert Dust, Arid, Biomass, Angstrom Exponent

P 5.5 - Observation projects for atmospheric greenhouse gases by Japan Airlines (JAL)

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Commercial airlines provide a powerful observational platform for obtaining free tropospheric CO2 systematically for long periods of time over a large geographical space. An earlier observation between Australia and Japan was carried out in 1984 and 1985 by Tohoku University in Japan, in cooperation with Japan Airlines (JAL) (Nakazawa et al., 1991). The JAL project was re-started in 1993 over the same western Pacific region, with the collaboration of scientific institutes in Japan, the JAL Foundation, JAL, aircraft engineering companies, and aviation regulatory agencies. The first phase of the JAL project from 1993 to 2005 was carried out using an Automatic air Sampling Equipment (ASE) for flask sampling system to obtain a long-term record of CO2 and other trace gases. The CO2 record over the western Pacific has provided valuable information on the latitudinal distribution of the atmospheric CO2 seasonal cycle and on the inter-annual variation of long-term increasing trends in the upper troposphere of both the Northern and Southern Hemispheres (Matsueda et al., 2002).

For the second phase of the JAL project (Comprehensive Observation Network for TRace gases by AIrLiner: CONTRAIL), an improved Automatic air Sampling Equipment (ASE) for flask sampling and a new Continuous CO2 Measuring Equipment (CME) for in-situ CO2 measurements were installed on Boeing 747-400 and Boeing 777-200ER aircraft (Machida et al., 2008; Matsueda et al. 2008). In all, one or both of these instruments have been installed on several Boeing aircraft operated by JAL with regular flights from Japan to Australia, Europe, East, South and Southeast Asia, Hawaii, and North America, providing significant spatial coverage, particularly in the Northern Hemisphere.

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P 5.6 - Potential of in-service aircraft based greenhouse gas observations within IAGOS for constraining regional carbon budgets.

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Presenter : Shreeya Verma

The spatial and temporal variations of atmospheric CO2 contain information about carbon sources and sinks and characteristics of the CO2 exchange processes between the atmosphere and the surface of the earth. Within the recently established European Research Infrastructure IAGOS (In-service Aircraft for a Global Observing System), highly accurate and precise in-situ observation of greenhouse gases is foreseen in the near future. The objective of this study is to quantify the reduction in uncertainty in estimates of carbon sources and sinks brought about by the use of this newly developed data stream. Anticipating the deployment of five GHG observing systems within IAGOS, the flight tracks from five in-service aircraft within MOZAIC, the predecessor of IAGOS, are used in an inversion system to assess the constraint on the carbon budget and quantify the potential for reduction in posterior CO2 flux uncertainties. These measurement locations are used to evaluate the impact of data from aircraft on the reduction of flux uncertainties compared to that based on the existing global observation network, and furthermore to identify areas where the addition of these measurements would be of greatest impact. We use the Jena Inversion System that employs the Global Atmospheric Tracer Model TM3 for atmospheric transport,

focusing on the period 1996-2004. The vertical aircraft profiles are input into the inversion as two partial-column averages instead of point measurements, the lower partial column completely containing (and exceeding) the boundary layer. Thus the error due to imperfect model representation of the boundary layer height can be diminished, and results in the reduction of the overall model-data mismatch error. The experimental design is such that in each simulation the existing measurement network is augmented by pseudo-observations from up to five simulated IAGOS aircraft. Uncertainty reduction from each of these simulations is compared to the uncertainty reduction from simulations employing only IAGOS or only the existing observation network. Additional constraint on regional carbon budgets is expected from the reduced model-data mismatch error when using vertical profiles as compared to using point measurements within the atmospheric boundary layer only

P 5.7 - Five Years of NOy Measurements in the UTLS from MOZAIC

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Nitrogen oxides play an important role in atmospheric chemistry related to ozone and oxidation capacity. Once released in the atmosphere, NOx is quickly oxidised to, e.g., nitric acid (HNO3) and organic nitrates, most importantly PAN, from where NOx can be recycled via photolysis and thermal decomposition. NOy (the sum of NOx and its atmospheric oxidation products) is invariant to homogeneous chemistry and hence is a good tracer for long range transport of NOx emissions, particularly in the upper troposphere.. The most important sources of NOy to the upper troposphere are lightning, transport from the stratosphere and the boundary layer and aircraft emissions. Measurements of NOy in the free troposphere and lower stratosphere are important for understanding of the impact of air transportation on the budgets of greenhouse gases such as ozone and methane.
An instrument for autonomous NOy measurements was flown continuously aboard the MOZAIC aircraft D-AIGI operated by Deutsche Lufthansa between April 2001 and May 2005 during more than 2000 flights, 1533 of which(8500 FLH) yielded high quality data. Coincidental measurements of NOy, O3 and CO are available on 1125 flights between January 2002 (installation of the CO instrument) and May 2005.
The poster presents the climatology of MOZAIC NOy data in the UT and typical vertical profiles. In winter, NOy concentrations in the upper troposphere (UT) are usually well below 0.5 ppb, whereas concentrations of several ppb are frequently observed in spring and summer, in particular over the western North Atlantic, but also over Europe and the Arabian Peninsula. The data are compared to global CTM simulations from the MACC reanalysis with IFS-MOZART3 for O3, CO and NOy. The comparison focuses on the climatology in the upper troposphere, as well as correlations and probability distribution functions (PDF) of the chemical species as a function of different vertical coordinates (height, potential temperature and potential vorticity) for different geographical regions (Europe, US East coast, East Asia).

P 5.8 - Observations of atmospheric composition, clouds and precipitation in Dronning Maud Land, East Antarctica

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The radiative forcing by aerosol-cloud interactions is one of the key atmospheric processes in the climate system with remaining high uncertainty. To reduce this uncertainty it is essential to investigate the relationship between changes in aerosol properties, cloud macro- and microphysical properties including precipitation, and the atmospheric dynamic conditions. Such investigations are in particular relevant, yet sparse, concerning the Antarctic ice sheet for which precipitation is the only source term in the surface mass balance. A better understanding of aerosol-cloud interactions in Antarctica is important to be able to improve climate models for this region.

At the Belgian Antarctic research station Princess Elisabeth, a monitoring observatory exists for atmospheric aerosol (optical extinction, absorption, scattering, number size distribution, total number, total mass concentration and concentration of light-absorbing aerosol), clouds, (cloud base height, vertical extent, icy or liquid, cloud base temperature) and precipitation (snowfall rate and vertical profile), complemented by a specific Antarctic automatic weather station. Additional temporary measurements during austral summer include measurements of the total ozone column, aerosol optical depth in the UV-B, balloon radio soundings and measurements with a cloud condensation nuclei counter. It is a summer research station, situated around 180 km inland, north of the Sør Rondane Mountains in Dronning Maud Land, East Antarctica (71º57'S, 23º20'E, 1390 m asl). During the rest of the year, the station and monitoring observatory is operated automatically and controlled remotely.

Analyses of the meteorological and dynamical conditons show two predominant meteorological regimes (cold catabatic ; air masses from Antarctic interior) and synoptic (extratropical cyclones and frontal systems). Accumulation by precipitation is restricted to few events with large annual variability of the quantity. We will show for some precipitation events the respective aerosol and cloud properties. In addition, seasonal optical and physical properties of the atmospheric aerosol will be presented.