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In this Issue:

A Note from the IGAC Co-chairs: Phil Rasch and Kathy Law

This issue of the newsletter will acquaint our readers with many of the "mobile platforms" being used to produce systematic, high quality, and routine in-situ measurements of atmospheric trace constituents. It is a logical extension to a theme begun in our previous issue reviewing instruments useful in characterizing constituent distributions in the atmosphere, and the processes that control those distributions.

The previous issue focused on space based measurements for assessing tropospheric distributions. The mobile platforms treated in this issue provide a more local picture of constituents than satellite data, but a much broader view than seen with the measurement platforms fixed at a particular geographic location. As such they are particularly useful as integrators of information, connecting the global picture gained from satellite measurements with the local information associated with ground based measurement strategies. One of the other distinguishing features of these platforms is that, unlike those used in relatively brief field experiments, these measurements provide routine data over longer time periods, sometimes years, allowing a more complete picture of many regions, such as the upper troposphere/ lower stratosphere that are difficult to sample routinely and which are important to the climate system.

The articles provide a fascinating glimpse into the utility of ground (rail and ship), and commercial aircraft based platforms for these purposes. They also serve to highlight the benefits of active cooperation between the scientific community and industry. We believe the measurements and strategies used to gain information about atmospheric chemistry from these platforms are extremely valuable to IGAC activities and goals.

We hope you enjoy and learn from the articles.

GLOBAL CHANGE IGBP
A Core Project of the Internations Geosphere-Biosphere Programme

CACGP
In Cooperation with IAMAS Commission on Atmospheric Chemistry and Global Pollution

The CARIBIC aircraft system for detailed, long-term, global-scale measurement of trace gases and aerosol in a changing atmosphere

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Introduction

The CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) system involves the monthly deployment of an automated atmospheric chemistry

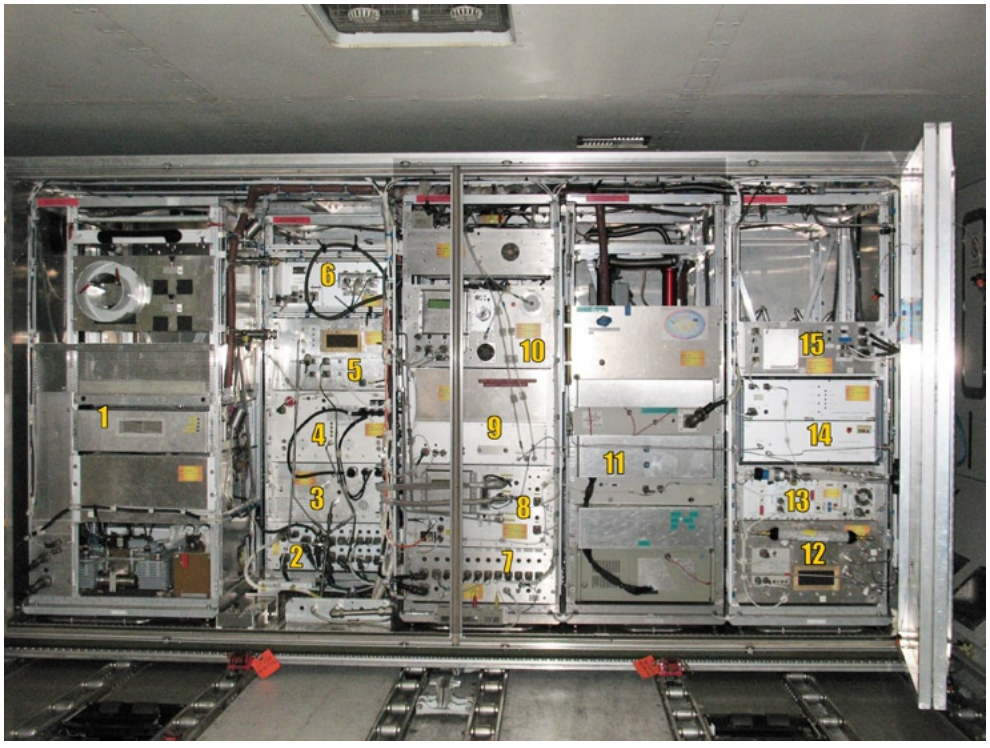

Figure 1. Frontal view of the container inside the forward cargo bay of the A340-600, facing the flight direction. Its width is 3 meters and its mass is 1.5 ton. In-flight its doors are obviously closed. 1. PTRMS. 2. Main power unit. 3 Aerosol, CPC and sampler. 4. Aerosol, 2 CPCs. 5. Aerosol OPC. 6. DOAS. 7. Base power unit. 8. Water total and vapor. 9. Mecury. 10. Ozone fast and standard. 11. NO and NOy. 12. CO_2. 13. CO. 14. O_2, 15. Master computer. At the rear, the 2 air sampling units, air pumps, a VOC sampling unit, calibration and working gas supplies are accommodated.

Figure 2. The inlet system as permanently mounted just in front of the belly fairing. The lowermost probe is the aerosol diffuser tube with a leading shroud. Left above it is the trace gas tube. PFA coated tubing is used to transport the air to the container. To the right is the smaller probe for water having a leading orifice (total water) and one at the side (gaseous water). The video camera points slightly downwards. The viewing ports of the DOAS telescopes (one nadir and 2 limb) are just visible on the right side of the main spar close to the skin of the aircraft.

observatory inside an airfreight-container (Figure 1) on board of a Lufthansa Airlines Airbus A340-600 (Figure 2). Presently, four successive long-distance measurement flights – nominally 48 hours including stopovers – can be carried out each month. CARIBIC, with its extensive scientific payload regularly probing the atmosphere at 10-12 km altitude over long distances, is providing detailed data and will be able to do so over many years, continuing to advance atmospheric research. This type of observatory has an unparalleled potential. CARIBIC is a joint endeavor by 11 institutions from 7 European countries, and as such is a large, ongoing experiment. In this IGAC communication we briefly explain the main aspects of this flying observatory and its operations. We also provide, albeit brief, information on the project and some results. For reference we point the interested reader to the CARIBIC website (www.caribic-atmospheric.com).

Background

Recent advances in atmospheric chemistry using modeling and measurements are impressive. In view of the unprecedented and still partly unpredictable course and consequences of human-induced, rapid climate change, our science discipline (and the others in IGBP) will have to provide, in a short realm of time, a detailed understanding of atmospheric chemistry and related compartments of the Earth system. It is clear that advances in modeling call for advances in measurements, and the two will go more and more hand in hand as the science evolves. (Popularly speaking: "data resistance is futile, you will be assimilated"). Moreover, a system as complex and extensive as the coupled atmosphere-biosphere-ocean-cryosphere-anthroposphere demands extensive and detailed monitoring. As a result of this all, observational data increasingly need to be detailed, continuous, consistent, and global. Joint efforts and new methods like CARIBIC are required.

Rationale

Facing global environmental change within a lifetime, it is essential to develop powerful, efficient observational tools that fill gaps in knowledge. The unique role of the NOAA-ESRL global network of stations, AGAGE, GAW (WMO), and others are well acknowledged. We also know the many possibilities of remote sensing from satellites, and the fine work done on retrieving information from spectra. Doing justice to all observational systems is beyond the scope of this article, but we do emphasize that besides the spectrum of atmospheric trace gases, aerosol particles are of special importance. Their direct and indirect climate forcing, the microphysical processes and cloud formation, combined with their large variability linked to their short lifetime, require increased research efforts.

One can easily grasp the logic of using civil aircraft for making observations, as it is inherently cost effective. With aircraft cruising at 9 to 12 km altitude (~30-40% stratospheric air) the UTLS is extensively probed in the mid- to higher latitudes (e.g., see Figure 3). The

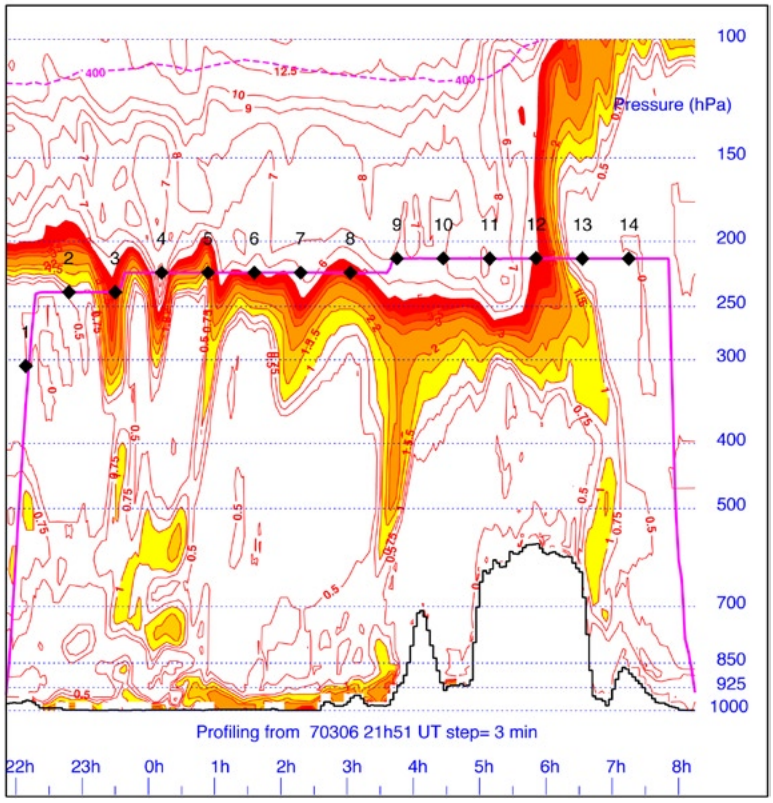

Figure 3. Vertical cross-section of the potential vorticity (PV) field along the aircraft's track for a flight from Frankfurt to Guangzhou (Cross section of 209 profiles, 7/3/2007, 0 UTC, from model level data; KNMI/ECMWF). The tropopause is highly structured, and in this particular case lowermost stratospheric air was intercepted for about 60 % of the time. PV is, as is common, given in Potential Vorticity Units. (PV is a physical property that is largely conserved, with higher values in the stratosphere).

importance of the complex UTLS interface is well acknowledged and thus requires little emphasis here. At lower latitudes, the free tropical troposphere – where data are also scarce – is also regularly probed. For flights traversing these regions, characteristics of the ITCZ and deep convection in general are documented in detail by measurements from commercial flights. Critically, these measurements can be and are executed on a regular basis. The question is of course, if one can obtain sufficiently detailed data. CARIBIC targets the maximum number of trace species and as such is a versatile, cost- effective tool for detailed global observation. After a proof-of-concept period (1997-2002, CARIBIC phase 1) it has evolved into a steady state, with a full-fledged flying observatory in operation since 2005 (CARIBIC-Lufthansa; projected to continue until 2014).

The consortium

It is useful to briefly explain the way CARIBIC functions scientifically/organizationally. The consortium of 11 partners actively operating CARIBIC and analyzing samples has gradually evolved with an underlying philosophy that institutions operate their equipment in the measurement container and use the information obtained within the framework of their own scientific mission. In some cases, the coupling between research by a given partner institution and the potential CARIBIC offers is very strong. For instance the operation of the proton transfer reaction mass spectrometer by Detlev Sprung and Andreas Zahn gives information (e.g. acetone, methanol) that is rare to such a degree that a major involvement is readily justified. In other words, in such cases CARIBIC offers a specific application great advantages. In another case, e.g. that of CO_2 measurements, the CARIBIC data are useful, but as such constitute a more modest addition to all CO_2 concentration data available worldwide through other means. Thus, in the case of continuous CO_2 measurements the embracement Institution-CARIBIC is less intensive, but of course still cordial. Having highlighted these two extremes, we emphasize that the true (long term) power of CARIBIC lies in the entire, synchronous datasets. Using CO_2 as an example, one does not merely obtain CO_2 concentration data, but a fairly complete characterization of the air masses. This type of additional information on the air matrix (e.g. COS and SF_6) is likely to become increasingly important in quantifying the atmospheric CO_2 cycle. (Note that ^{13}C and ^{18}O isotope measurements are also made of the CO_2 air samples). A very clear example is given by the NO and NO_y measurements. Such measurements are much more rare than CO_2 measurements for instance, but it is the availability of other real-time CARIBIC measurements (including aerosol) that enhance the value of these NO and NO_y measurements. Finally we reiterate that it is clear that the final, detailed and extensive output of CARIBIC should serve those who continuously improve models by offering them consistent, detailed datasets. The support in verifying satellite-based observations of an increasing number of trace gases is also of great importance.

Operation

The CARIBIC system with its measurement container (Figure 1) also includes a sophisticated air inlet system (Figure 2). Furthermore there are the provisions inside the Airbus A340-600 for electric power, flight data acquision/storage, various tubing running from the inlet to the container, and the connection interface. At Frankfurt Airport the container is installed in the Lufthansa Airbus A340-600 and uninstalled after four successive flights. Each operation takes less than half an hour. Back at the Max-Planck-Institut für Chemie (MPI-Chemistry) in Mainz, post-flight simulation tests are conducted, data are retrieved, and air and aerosol samples are unloaded. The time between flights is used for instrument calibration, improvements, repair, and air sample analyses. Prior to the next measurement flight, extensive testing is carried out. The entire operation, i.e. the activities at the airport by airline personnel and scientists and the container preparation, has become routine. Improvement/maintenance of equipment has proven to be a limiting factor. With present funding, 12 sets of 4 flights can be accomplished each year. The most cost-effective extension is to increase the number to 6 flights and beyond.

Funding

Evidently CARIBIC requires financing for operation and flight cost. The consortium pays Lufthansa Cargo the cost of airfreight at commercial rates (10 to 16 k€ per mission of four flights). This "user pays" principle gives the scientists a degree of independence towards the airline and renders the project acceptable for the airline. The financing is based on the institutions contributing person-power, equipment, consumables, and part of the mentioned flight costs, supplied within the framework of various funding schemes. German national science funding and EC funding have helped and still help CARIBIC, although the ad hoc character and the competition with "integrated projects" and "networks of excellence" in case of EC funding has proven fairly challenging for an innovative, large project like CARIBIC. One reason for this, among others, is that the EC needs science-based directives for its environmental policies. This translates into fitting scientific research into a certain framework, often based on short-term directives. In the end the Max Planck Society has supplied a special grant for CARIBIC to bridge the gap to the new EC science funding structure (European Research Council) and the development of European Infrastructures (i.e. the integration of routine aircraft measurements in a global observation system, IGACO).

What we measure with CARIBIC

For details of the instrumentation specifications we refer readers to the technical publication in Atmospheric Chemistry and Physics (ACP; Brenninkmeijer et al., 2007). Ozone is measured both by a very fast (but somewhat less accurate) analyzer and by a highly accurate analyzer with less time-resolution. Water vapor is measured using a fast photo-acoustic spectrometer as well as by an absolute system based on a chilled mirror dew/frost

point hygrometer. Total water (ice particles plus vapor) is measured using a separate photo-acoustic spectrometer. Carbon monoxide is measured using vacuum ultraviolet fluorescence. Carbon dioxide is measured using non-dispersive infrared absorption. Mercury is measured using pre-enrichment on gold followed by UV fluorescence. A unit in development measures the variations in the oxygen content of air using fuel cells. A proton transfer reaction mass spectrometer (PTRMS) measures acetone, acetaldehyde, methanol, and acetonitrile. A large system using two sensitive NO chemiluminescence detectors based on the reaction with excess ozone measures NO and NO_y (after conversion to NO on gold using hydrogen gas). Three condensation particle counters (CPCs) with different thresholds measure fine aerosol particles. An optical particle counter can measure aerosol particles up to the overall upper cut-off of about 5 micrometer. An impactor system collects aerosol particles on 14 sets of two ultra thin foils with 1.5 hour time resolution. These samples undergo extensive post-flight analyses using nuclear techniques for elemental analyses (PIXE and PESA) and microscopy. An experimental unit with 16 absorption tubes has collected Organic VOC (OVOC) samples. A system collects 28 air samples in glass containers. These air samples are used to determine concentrations of greenhouse gases, hydrocarbons and halocarbons and for isotope measurements (carbon dioxide and hydrogen). A camera provides video images of clouds, cirrus and contrails ahead. Finally a three-axis DOAS remote sensing system integrated in the inlet system measures path integrated NO_2, O_3, BrO and HCHO, under suitable conditions.

Advantages and data usage

The multitude of compounds detected by CARIBIC provides a detailed picture of the composition of the wide range of air mass types intercepted by the aircraft at cruising altitude. Figure 4 illustrates (by means of 5-day back trajectories) typically–sampled airmass pathways. The many compounds analyzed in air masses of such vastly different origins and history, coupled with the monthly recurring flights, provide detailed information about mixing, sources, and seasonal variations and their causes. In the course of years, trends and inter-annual variations will be captured. Having a single observing system also ensures a high degree of consistency in the data. We note that the container system is modular and not part of the aircraft system. This has advantages with regard to extensions and modifications of the scientific payload in view of certification issues. CARIBIC is open to participation by any other science group that can provide suitable equipment. The structure of CARIBIC, as we stated, in principle assures an active participation and interest of the various partners in using the data acquired within the realms of their research objectives. Several papers highlight this type of application (Ebinghaus et al., 2007; Zahn et al., 2002 & 2004; www.caribic-atmospheric.com). Furthermore, case studies of certain events have proven to be valuable. In the longer term, trends can be analyzed, and increasing cooperation with the modeling community (Gloor et al., 2007; Peylin et al, 2007) will unfold the full potential of the CARIBIC datasets.

Limitations

A fundamental limitation to this data set is that radical species (HOx, etc.) cannot be measured. With the existing inlet system we have already reached the limit of what scientific appendix a passenger aircraft can carry. A "wish-list" of further trace gases to be measured real-time include formaldehyde, PAN, peroxides and sulfur dioxide. A 100-sample-capacity air collector is envisaged. Practical limitations are related to the frequency of flight events (at this moment monthly) and the routes available for the specific type of aircraft. The CARIBIC system was implemented on a new type of aircraft to provide a long time horizon of operation. In the fast-moving world of commercial aviation, where new aircraft types are introduced and competition is stiff to say the least, aircraft destinations change beyond control of the scientists. As we will show in the course of the remainder of this brief article, flying from Europe produces a Eurocentric view of the chemistry of the global atmosphere. This limitation notwithstanding, the positive experience over a good number of years of operations of CARIBIC, even under troublesome funding conditions, justifies hope that such systems can be operated from other continents as well.

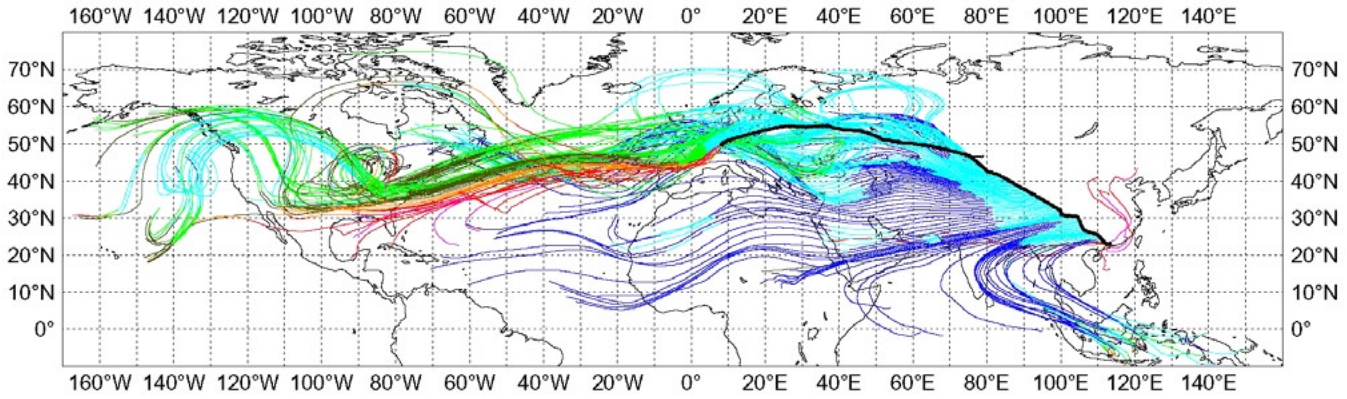

Figure 4. Five day back-trajectories for points along a single flight path (for more detail, like individual trajectories, we refer to the KNMI website for CARIBIC). Note the change in "origin" of the air upon approaching Guangzhou.

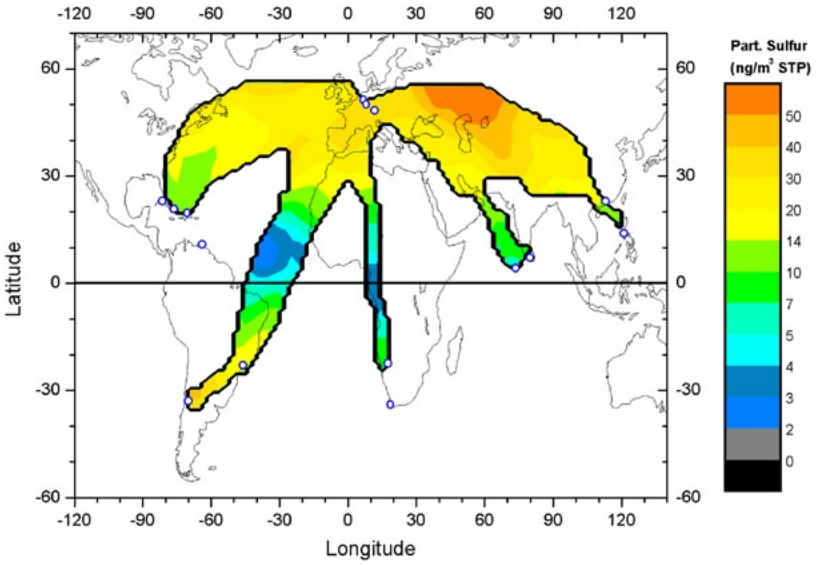

Figure 5. The concentration of sulfur (as present in aerosol particles) based on PIXE analyses carried out in Lund. Used are all data from the previous and present CARIBIC flights. At mid to higher latitudes this climatology is biased by the fraction of stratospheric air (more sulfate). For this a correction can be applied based on PV or ozone. At other latitudes the picture directly gives the tropospheric distribution. .

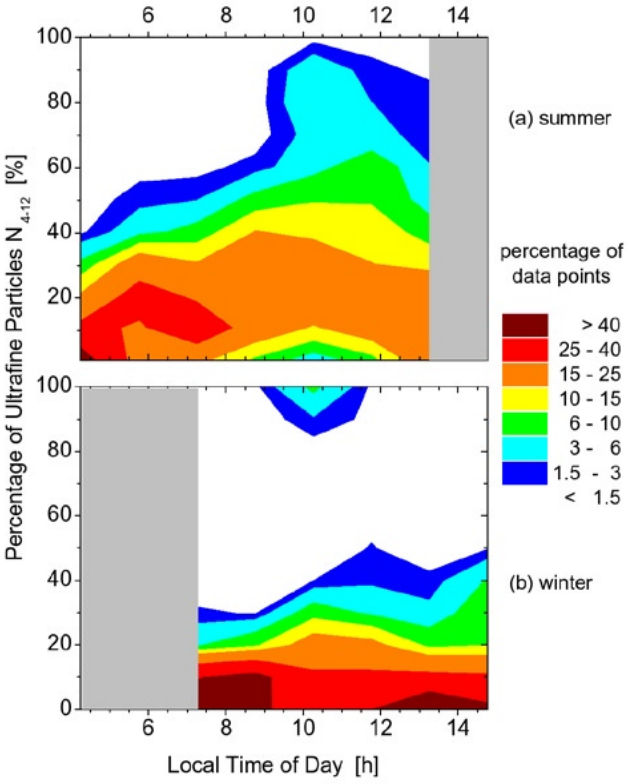

Figure 6. Probability distribution for the percentage of ultrafine particles (4 nm < diameter < 12 nm, N_{4-12}) at 8-12 km altitude, versus local time of day for tropical latitudes between 5° and 20°N over the Arabian Sea in (a) summer and (b) winter. Gray shading marks lack of data. For more info the reader is referred to the website's list of PDF files of publications (M. Hermann et al. 2003).

Examples of results

With the present CARIBIC-Lufthansa system we have been making measurements on flights from Frankfurt to Santiago via São Paulo, and to Manila via Guangzhou (Figure 4 gives an example for such a flight). Also Toronto and Houston with a "stopover" in Frankfurt are destinations. Furthermore, next to Frankfurt, Munich can also be a departure airport. Here we present some examples of CARIBIC results, starting with the extensive aerosol work.

CARIBIC aerosol particles are subjected to an ever increasing number of sophisticated analyses, including proton induced x-ray emission (PIXE), proton elastic scattering (PESA, for the light elements including hydrogen), various forms of transmission electron microscopy (TEM) and atomic force microscopy (AFM). It takes 1-2 hours' collection time in-flight to obtain an aerosol sample; thus, integration over different air masses often occurs. Figure 5 shows a climatology of sulfur at cruising altitude (typically 9.4 to 10.7 km) using the previous (Boeing 737) and present CARIBIC system. What is first clear is the diversity of CARIBIC routes originating in Europe. Flights are not restricted to narrow corridors. Second, the amount of sulfur detected varies systematically. Given that two different air inlet systems and two different samplers were used, the consistency of the results gives great confidence in the suite of results. The high values at higher latitude are related to a larger fraction of stratospheric air sampled, in particular over central Asia, and a correlation with potential vorticity is clear. Martinson et al. (2005) have derived and published a sulfur inventory for the upper troposphere and lowermost stratosphere based on their unique data.

Aerosol abundances are registered by the three condensation particle counters and an optical particle counter. Figure 6 shows the diurnal variation of the fraction of ultrafine particles (diameters between 4 and 12 nm, i.e. freshly formed particles) for summer and winter over the Arabian Sea. The importance of photochemistry during the daytime is obvious, whereas the difference between summer and winter is explained by the rarity of deep convection in winter as compared to summer. Figure 7 shows the probability of Aitken mode particles (diameter >12 nm) for flights between Frankfurt and São Paulo during the NH summer. Again the important role of deep convection for the vertical transport of precursors and particles is clear. The ITCZ at ~ 8°N can be readily discerned. Finally, Figure 8 shows – for tropical latitudes – the decline in the fraction of ultrafine particles as a function of time elapsed since the air was in contact with clouds (based on satellite imagery and back-trajectories) and its interception by the aircraft. In regions of deep convection ultrafine particles are formed and their fractional abundance subsequently decays along the pathway away from the clouds. At the same time, tropical

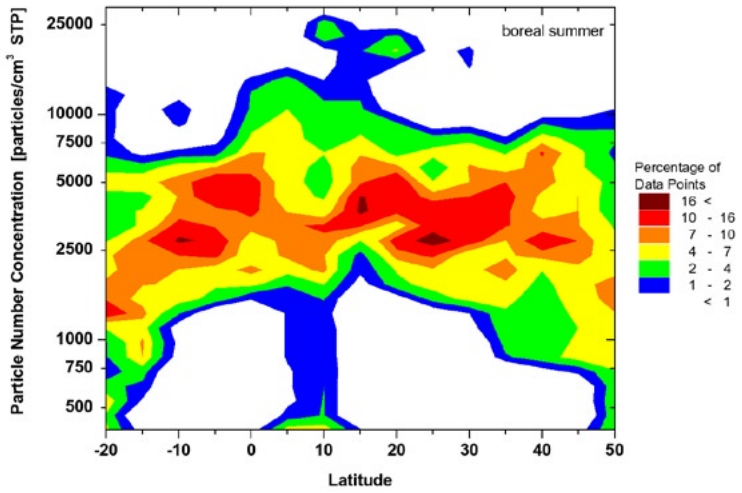

Figure 7. Probability distribution of Aitken mode particles (diameter >12 nm) in the UT in boreal summer along the South America route. Colors indicate the percentage of all particle measurements in a latitude band that fall into a particular concentration bin. Mean location of the ITCZ over the Atlantic is around 8° N.

deep convective clouds act as a sink for Aitken mode particles.

While Figures 5-8 demonstrate the capacity of CARIBIC for aerosol research, with the emphasis on systematic information in space and time, we use Figure 9 to elucidate details obtainable from even a single flight. Lack of space prevents display and discussion of the non-methane hydrocarbon and halocarbon measurement results for the air samples or to discuss the fine structure of many highly time resolved measurements (e.g. water vapor and ozone).

The vertical PV cross section and the 5 day back trajectories for this flight from Frankfurt to Guangzhou were already presented in Figure 3 and 4 respectively. The interested reader may wish to inspect the full meteorological information from CARIBIC flights using the KNMI website of Peter van Velthoven, http://www.knmi.nl/samenw/campaign_support/CARIBIC/. This particular flight of March 6, 2007, is characterized by its predominant cruising in the lowermost stratosphere (~60%). For the stratospheric sections of the flight a compact negative correlation between carbon monoxide and ozone, down to several highly resolved structures at PV > 6, is visible. In the sub-tropical lowermost stratosphere, air with up to 500 ppbv ozone and only 30 ppbv carbon monoxide was found. This shows that despite the limited range of cruising altitudes a useful range of airmasses can be intercepted .

Ultrafine particles show detailed features in their abundance but are virtually absent south of 45° N (after 27:00 UTC), in part because this is where the aircraft started to encounter lowermost stratospheric air (PV rising to 7 PVU), and subsequently free tropospheric air in the absence of convection. The Aitken mode aerosol is present throughout, partly correlating with CO, for instance north of 45°, and reaching high values in the tropical troposphere over China. The narrow spikes in the Aitken mode aerosol abundance are due to aircraft emissions.

NO_y correlates well with ozone, which is indicative of stratospheric HNO_3, whereas several of the aerosol spikes also correlate with high NO_y. Just before sample 5 was taken, NO_y, ultrafine and Aitken aerosol, and CO are elevated, showing considerable chemical activity. NO is also displayed, but is only detected after 26 UTC, corresponding to sunrise over eastern Asia. The anti-correlation between NO and NO_y – seen when flying in the lowermost stratosphere – changes to a positive correlation for the tropospheric part of the flight. Finally the same panel shows humidity varying from 100 down to about 5 ppmv.

The next panel displays the "greenhouse gases" (CH_4, CO_2, N_2O, SF_6) as measured in 14 of the 28 glass flasks on board (outward bound flight). Carbon dioxide ranges from 390 ppmv (early spring NH maximum) to 380 ppmv in the

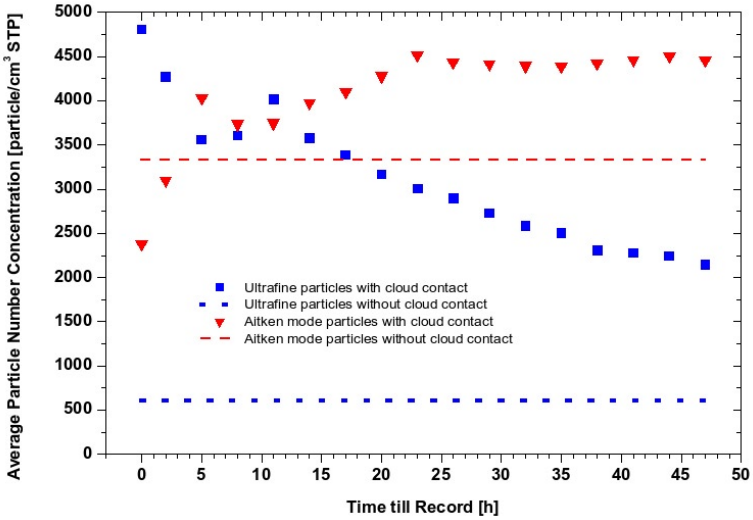

Figure 8. The dependency of particle number concentrations (over the Arabian Sea) on the time elapsed between the last cloud contact and the actual measurement by the CARIBIC aircraft. Each point includes all values for which the last cloud contact fell into the time period indicated at the x-axis. Blue squares represent the cloud-influenced averages of particles between 4 and 12 nm diameter (ultrafine particles), and red triangles of particles lager than 12 nm (Aitken mode). The corresponding values, which are not influenced by clouds, are indicated as dotted and dashed lines in the same color, respectively.

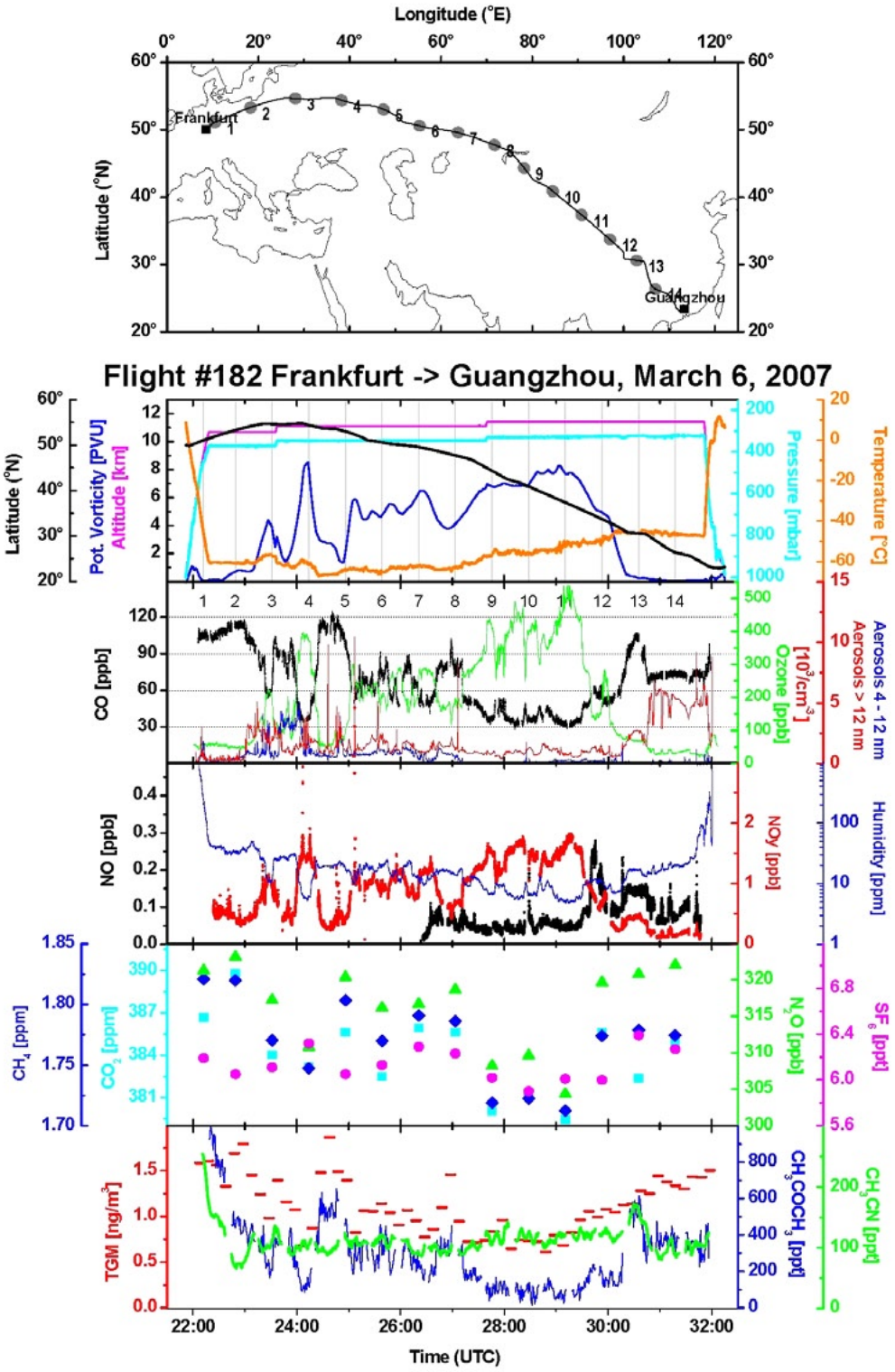

Figure 9. An overview of measurements obtained for a flight to Guangzhou (Meteo data in Figures 3 and 4). The grey dots on the map and the numbers on the x-axis of the top plot indicate locations of discrete air samples. Irregularities in the flight track are mostly due to the pilots avoiding lightning storms, turbulence or both. The top panel gives certain flight parameters obtained from the Arinc databus in the aircraft and the PV values from KNMI. The second panel gives carbon monoxide, ozone, and the aerosol data. The third panel gives NO, NOy and humidity. The fourth panel gives the "greenhouse gases" obtained for air sample measurments. The bottom panel gives TGM (Total Gaseous Mercury), acetonitrile and acetone..

lowermost subtropical stratosphere (sample 11) reflecting the seasonal time-lag. At first glance, methane variations (1.7 to 1.8 ppmv) track those of carbon dioxide. The chemical loss of methane in the stratosphere and the seasonal change in carbon dioxide are the main cause for this. Furthermore, the deviation for some samples from the somewhat fortuitous correlation, namely samples 1, 5, 6 and 13 is related to elevated methane levels in polluted air (e.g. see also elevated carbon monoxide). Note that sample 2 – taken over northeastern Europe -- has the highest level of carbon dioxide. This sample's trajectories indicated possible surface contact (500 hPa<p<850 hPa), over the USA five days prior to sampling. Furthermore, the sulfur hexafluoride data show its use as an indicator of the age of air masses in the stratosphere (e.g. lowest value here of 5.8 pptv for sample 11, at over 500 ppb ozone), whereas elevated levels (e.g. sample 13 over China) indicate polluted air. Nitrous oxide levels reached 322 ppbv, with a low value of ~305 ppbv, again for sample 11.

The lowermost panel gives total gaseous mercury (TGM), which correlates well with carbon monoxide. The combustion of coal and biomass burning are sources for both of these gases. The low values in the stratosphere (down to 0.5 ng/m^3) are caused by the loss of mercury on aerosols after oxidation to the less volatile form. The CARIBIC mercury measurement data already form a significant part of the globally available inventory of such data.

One of the most difficult experiments on board of CARIBIC is the proton transfer reaction mass spectrometer. The unattended operation and calibration over 2 days has been a major challenge, but the unique measurements appear worth the effort. For acetone, which is a source of OH in the upper troposphere, a tight correlation with carbon monoxide is visible, but the dynamic range in concentrations is almost a factor 10, in comparison to ~ 4 for carbon monoxide. We also show acetonitrile, a product of biomass burning, visible at about 100 ppt but with less overall variability (due to its lifetime about 1 year). Other compounds measured with the proton transfer mass spectrometer are acetaldehyde and methanol.

Conclusions

The CARIBIC community can confidently show a large amount of valuable data, of which we hope other colleagues (including modelers) will also benefit. Data from CARIBIC phase 1 are deposited on the CERA database, and those from the present phase are following (please see the CARIBIC website or contact the coordinator). Such a joint effort allows us to better understand the chemistry and transport of the atmosphere, the strength of natural and anthropogenic sources of a host of trace gases, the complex behavior of particles, and atmospheric change in general over the years to come.

We argue for similar systems to be deployed, for instance in the USA, China, India, and Brazil to provide a detailed picture of our atmosphere over a very large part the globe. We have developed CARIBIC twice (once on a Boeing aircraft and once on an Airbus aircraft) and the feasibility is more than adequately proven, we believe.

Acknowledgments

We acknowledge that the ever deepening awareness and veritable concern in Europe, especially in Germany, regarding global climate change has made it possible to pioneer this valuable project. LTU, Lufthansa and Lufthansa Technics, in particular all staff that helped with enthusiasm and great skill to implement CARIBIC in the complex aviation environment, are faithfully thanked.

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The MOZAIC Program (1994-2007)

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MOZAIC (Measurements of ozone and water vapour by Airbus in-service aircraft; Marenco et al., 1998) was launched in January 1993 under the support of the European Commission (Aeronautics and Environmental Programmes) by Airbus Industries, INSU-CNRS and CNRM in France, FZJ in Germany, and the University of Cambridge in the U.K., and several European airlines (Lufthansa, Air France, Austrian, and Sabena). Three phases took place between 1993 and 2004 under co-funding of the European Commission. The measurements started in August 1994, with the installation of ozone and water vapour sensors aboard five commercial aircraft. In 2001, the instrumentation was upgraded by installing carbon monoxide sensors on all aircraft and a total odd nitrogen instrument (NO_y) aboard one aircraft. Since 2004, the measurements have continued without EC funding, instead being supported by institutional and national resources (INSU-CNRS and FZJ). Presently, three of the MOZAIC aircraft (2 Lufthansa, 1 Air Namibia) are still in service. More than 25,000 flights (comprised of two vertical profiles and about 8 hours of data in the UTLS per flight) have been realised since 1994 on the routes shown in Figure 1.

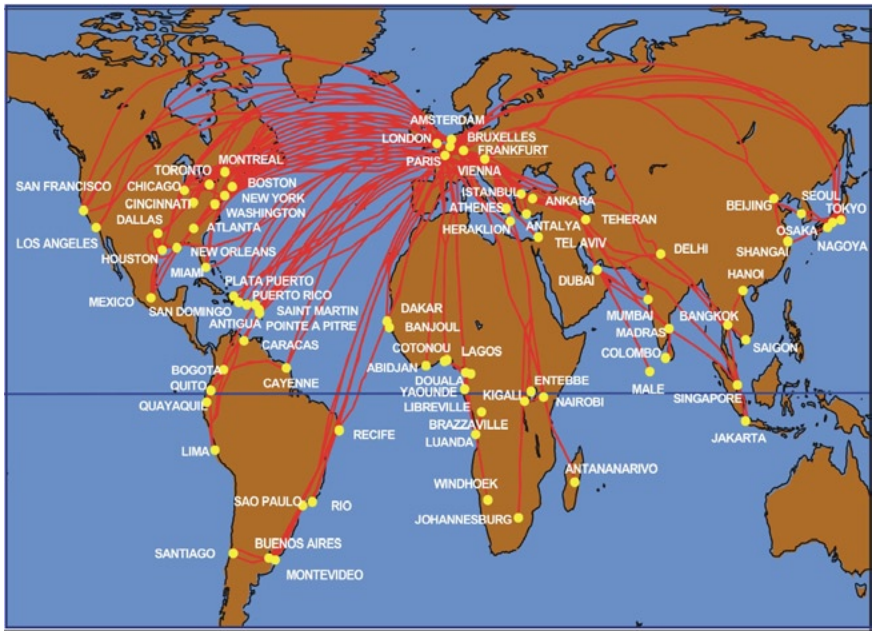

Figure 1 : Map showing coverage of the MOZAIC flights.

MOZAIC has provided novel information on the distributions of H_2O, O_3, CO and NO_y in the Upper Troposphere and Lower Stratosphere (UTLS) and on the vertical structure of these species in the troposphere. The data were exploited by the MOZAIC investigators and by co-investigators from research institutions worldwide, resulting in 13 PhD theses and 115 publications in peer-reviewed scientific journals (see http://mozaic.aero.obs-mip.fr/web/). Research topics include the seasonal, geographical and interannual variation of the trace gases in relation to their sources and atmosphere dynamics, the evaluation of global chemistry transport models and the evaluation of retrieval techniques for atmospheric chemistry products from space-borne observations.

Instrumentation and operation

Ozone is measured by UV absorption (Thermo Instruments, Model 49-103). The instruments are calibrated before and after each period of deployment (~every 12 months) and in-flight quality control is achieved, both for bias and calibration factor, with a built-in ozone generator and by comparing the data from aircraft that fly close to each other. The accuracy is estimated to be ± [2ppbv+2%]. Quality control procedures have not changed over the history of MOZAIC, in order to avoid instrumental artefacts in the time series (Thouret et al., 1998A). A comparison of the first 2 years of MOZAIC data with data of the ozone sounding network showed good agreement (Thouret et al., 1998B).

For the measurement of **relative humidity (RH) and temperature** a compact airborne sensing device is used (Vaisala, Humicap-H and Pt-100). The sensors are contained in a standard aeronautic TAT housing and are calibrated before and after deployment (ca. 500 flight hours) in the laboratory under simulated flight conditions. From these calibrations a 2σ-uncertainty of ±(5-10)% RH and ±(0.5-0.7) K for temperature was derived (Helten et al., 1998). The in-flight performance was confirmed by comparisons with reference instruments during dedicated aircraft missions (Helten et al., 1999). The response time of the humidity sensor is about 1-2 minutes.

For the measurement of **CO**, the IR gas filter correlation technique is employed (Thermo Environmental Instruments, Model 48CTL). Although less sensitive than more recent instruments (TDL or resonance fluorescence), the IR instrument is much less complex and provides excellent stability, which is important for continuous operation without frequent maintenance. The sensitivity of the instrument (normally 10 ppbv for 300 s integration) was improved by several modifcations (Nédélec et al., 2003), including periodic zero measurements, the use of a novel IR detector with temperature regulation, and by enhancing the pressure in the absorption cell. The precision achieved through this approach is ±5 ppbv or ±5% for a 30 s response time.

Figure 2: A340 aircraft of the MOZAIC program (Air France, Lufthansa, Air Namibia, and Austrian)

NO_y (the sum of NO and its atmospheric oxidation products) is measured by chemiluminescence of NO with O_3 after catalytic reduction of the different NOy compounds to NO by traces of H_2 on a hot gold surface (Volz-Thomas et al., 2005). The instrument was especially designed for routine operation aboard commercial aircraft, featuring a 50 kg package weight, unattended endurance of >5 weeks, and in-flight calibration. The sensitivity of 0.4–0.7 cps/ppt[1] gives a statistical detection limit of better than ±50 ppt for an integration time of 4 s and ±150–300 ppt at the maximum resolution of 10Hz. An in-flight comparison with a research instrument operated by ETH-Zurich (Pätz et al., 2006) showed agreement within the combined uncertainty of the two instruments (±7%).

All of these instruments are mounted in a 19" rack (134 kg) in the avionic compartment below the cockpit (see accompanying paper on IAGOS by Volz-Thomas), with the intake tubes mounted on the fuselage 7 m behind the aircraft's nose. The system operates automatically without help of the flight crew. The airlines (Figure 2) carry the equipment free of charge and perform the essential maintenance, i.e. functionality check, replacement of data disk and humidity sensor. Lufthansa has been operating 2 aircraft since 1994. Air France and Austrian have operated one aircraft each from 1994 to 2005. The former Sabena aircraft is operated since 2006 by Air Namibia with transportation costs provided by INSU-CNRS and FZJ. The three remaining aircraft carry the upgraded MOZAIC rack with O_3, H_2O and CO. In addition, NO_y is installed on one Lufthansa aircraft.

The observations are sampled at 4sec intervals between takeoff and landing (except NO_y, which is not measured during ascent and is switched off before landing). After processing and quality control by the responsible laboratories, the data are stored at Meteo France, together with metadata such as backward trajectories and the position of the potential-vorticity-based dynamical tropopause, calculated by Météo-France from ECMWF fields. Users have free access to the data through the MOZAIC data protocol (http://mozaic.aero.obs-mip.fr/web/).

The database is presently re-constructed with the support of CNES and INSU-CNRS under the Thematic Assembly Center ETHER. The new database will provide user-friendly access with the provision of data pre-selection according to, e.g., time, position. The new data base will also allow the introduction of new data from IAGOS.

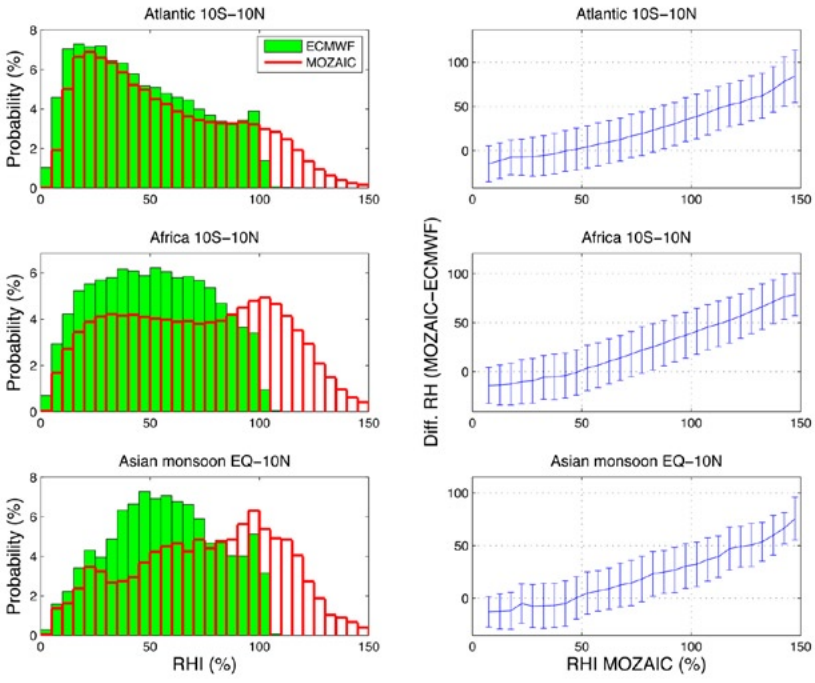

Figure 3: Probability density functions (PDFs) of RHi for the MOZAIC and ECMWF (left columns) and mean and standard deviation of the ECMWF biases (MOZAIC – ECMWF) stratified with respect to MOZAIC observations (right columns). Only observations equatorward of 10° are shown. From Luo et al. (2007B).

[1] cps = counts per second, photon counting

Selected Results

Upper tropospheric water vapor

Having provided the first climatology of upper tropospheric humidity, MOZAIC has shown that the UT is much wetter than previously assumed (e.g., Gierens et al., 1997 ; Luo et al., 2007A). This is an exiting finding because water vapour is the most important greenhouse gas and drives the engine of atmospheric dynamics. Luo et al. (2007B) show that the annual cycle of the ECMWF UT water vapour has an overall dry bias of about 10%-30% for relative humidity with respect to ice (Figure 3). The bimodal distribution of tropical UT water vapour seen by MOZAIC (Kley et al., 2007) has, in the ECMWF analyses, its moist mode abruptly cut off, due to the lack of ice supersaturation in the model. Ice supersaturation is associated with the existence of sub-visible cirrus clouds, which have important radiative impacts and which are important for contrail formation and persistency. MOZAIC humidity and temperature fluctuations, combined with local criteria for the formation and persistence of contrails, have allowed calculation of the maximum fractional coverage of contrails within a model grid (Gierens et al., 1997, 1999, 2000AB). Climate models have difficulties to reproduce the observed RH-distribution.

Tropospheric layers, stratification and mixing

A major outcome of MOZAIC was the documentation of the global and seasonal coverage of tropospheric layers with origins in the stratosphere or in the boundary layer. Approximately half of the layers are dry and ozone-rich layers, most likely originating from the stratosphere (Newell et al., 1999 ; Thouret et al., 2000). The seasonal cycle of the tropospheric ozone stratification exhibits a clear summer maximum (Colette et al., 2005ab). The persistence of stratospheric intrusions is most sensitive to the horizontal strain, while the mixing of layers coming from the boundary layer is enhanced by the summertime convection (Colette et Ancellet, 2006). The MOZAIC horizontal wind and temperature fluctuation data have been used to study gravity waves and two-dimensional turbulence and the atmospheric kinetic energy spectrum in general (Cho et al., 2000 ; Cho and Lindborg, 2001 ; Lindborg 1999, Lindborg and Cho, 2000 and 2001).

UTLS seasonal distributions (O_3, CO and NO_y)

Although MOZAIC data composites are not climatologies in the sense of a large temporal average, the frequent observations, the fine horizontal and vertical resolution of the data, and the 5 isobaric flight levels in the 9-12km altitude range provide a relevant picture of the global distribution

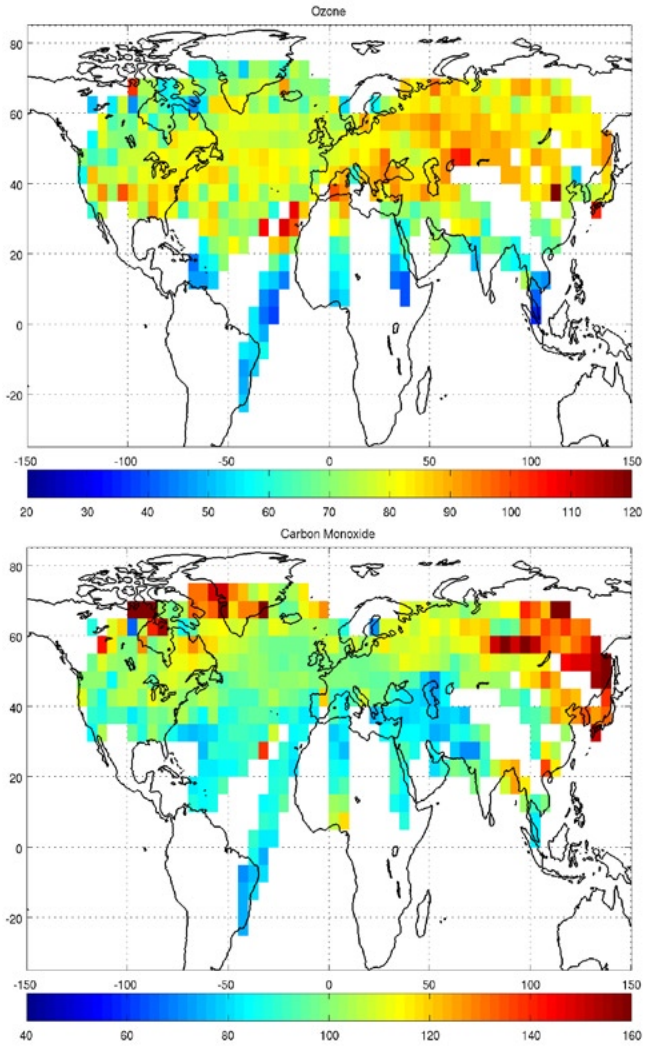

Figure 4: June to August distributions (in ppb) of ozone (top) and carbon monoxide (bottom) in the UT, established using 2001-2002 MOZAIC data. See Thouret et al. (2006) for the methodology.

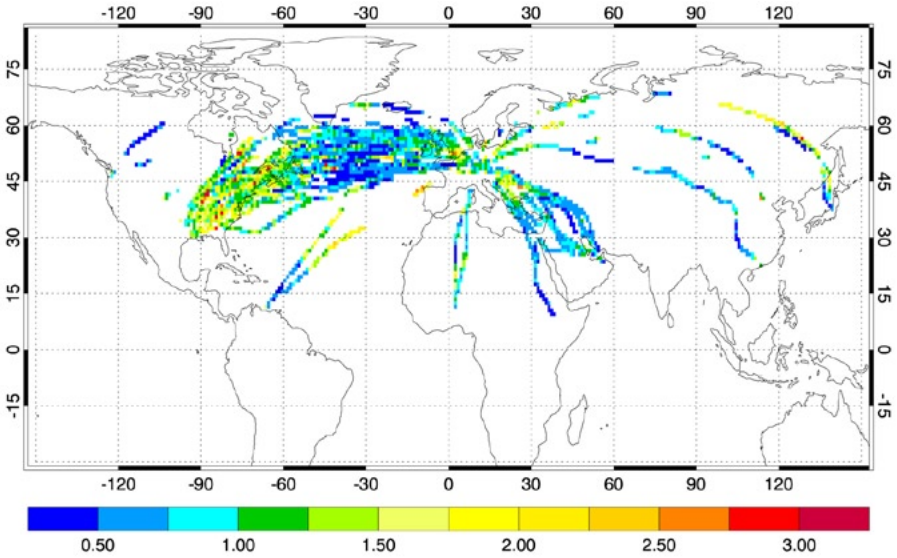

Figure 5: Mean summertime distribution of NO_y (ppb) in the UT (2001-2005, 1°x1° averages, for samples at altitude >8km and p > p(pv2)+ 15hPa – i.e. 15 mbar away from the PV=2 surface).

of species and allow us to monitor the seasonal cycles of tracers from the ground to the lower stratosphere. Seasonal distributions in the UTLS region (Figure 4) provide an in-depth description of the change of phase of seasonal cycles across the tropopause (Thouret et al., 1998a ; Emmons et al., 2000 ; Thouret et al., 2006) and over tropical areas (Bortz et al., 2006). Seasonal variations in vertical profiles have been established over Europe, USA and Japan (Zbinden et al., 2006), over Eastern Mediterranean (Kalabokas et al., 2007), over Beijing, China (Ding et al., 2007), over Africa (Diab et al., 2003 ; Sauvage et al., 2005) and over the Middle East (Li et al., 2001).

The climatology of NO_y in the UTLS exhibits a pronounced seasonal cycle generally in phase with that of O_3. During winter, NO_y mixing ratios in the UT are usually well below 0.5 ppb with no significant enhancement over the North Atlantic flight corridor. High NO_y mixing ratios are observed in spring and summer, particularly over the East coast of North America, but also over Europe and Asia (Figure 5). The correlation with O_3 and CO suggest that a large fraction of these enhancements is due to lightning, in addition to convective transport from the planetary boundary layer.

Model evaluation and assimilation of data

Present day global chemistry-transport models (CTMs), used to evaluate present and future impacts of human activities and to provide information to policy makers and industry, still have large uncertainties in the assessment of stratosphere-troposphere exchange, net photo-chemical production and surface deposition, all of which govern the tropospheric ozone budget. The MOZAIC dataset has been crucial for the identification of errors in emission source characterization, incorrect assumptions about the strength of convection, and missing chemistry in the models (Law et al., 1998, 2000 ; Emmons et al., 2000 ; Bregman et al., 2001); for the assessment of stratosphere-troposphere exchange (e.g., Clark et al., 2007); and for understanding of the net photo-chemical production (e.g. Crowther et al., 2002). Assimilation of MOZAIC ozone data in CTMs has resulted in improvements in the model up to 3-4 days after the data assimilation phase (Cathala et al., 2003) and have been shown to reduce the model bias (Chai et al., 2007). MOZAIC is also providing one important stream of data to evaluate the coupled simulations of the ECMWF dynamical model with global state-of-the-art CTMs in the European GEMS project.

Stratosphere–troposphere exchange

Over the tropics, MOZAIC has shown that stratosphere-troposphere exchange (STE) processes, previously not considered to be as important as at mid-latitudes, might enhance, via stratospheric intrusions, the oxidizing capacity of the UT (Cammas et al., 1998 ; Baray et al., 2003 ; Scott et al., 2001). Alternatively, STE can moisten the northern extratropical LS over the Asian summer monsoon anticyclone (Dethof et al., 1999). At mid-latitudes, tropopause fold and cut-off low processes associated with the polar jet stream dynamics are relatively well known exchange processes described from dedicated research experiments. The added value of MOZAIC is the abundance of tracer data (O_3, H_2O and CO) in the UTLS and in vertical soundings that allow us to conduct case studies (Gouget et al., 2000 ; Brioude et al., 2006), to validate budget studies in mesoscale and global models (Clark et al., 2007) and to build Lagrangian and statistical methodologies to assess the degree of irreversible exchange and mixing (Morgenstern and Carver, 2001 ; Borchi and Marenco, 2002 ; Borchi et al., 2005).

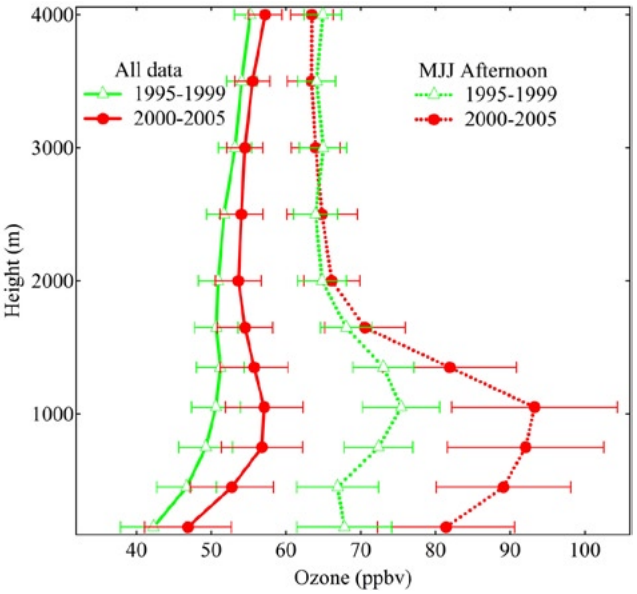

Figure 6: Comparison of mean ozone profiles in the lower tropopshere over Beijing between 1995-1999 and 2000-2005. Solid lines are annual mean values and dashed lines represent the data collected during summer in the afternoon (LT 15:00-16:00, May-July) (from Ding et al., 2007.)

Inter-annual variability and decadal tendencies

Although it is still too early for a significant trend assessment, some interesting results have emerged from the analysis of more than a decade of MOZAIC observations. A linear increase in ozone of about 1 ppb/yr is observed over the northern tropics (Bortz et al., 2006). Enhanced mid-latitude ozone concentrations in 1998-1999 in both the UT and the LS are likely attributed to a combination of different processes involving large scale modes of atmospheric variability like the North Atlantic Oscillation, and local or global pollution (Thouret et al., 2006 ; Zbinden et al., 2006). The marked inter-annual variability of four winter seasons was associated with stratosphere-troposphere exchanges (Morgenstern et al., 2000). Last but not least, ozone profiles obtained over Beijing between 1995 and 2005 (Figure 6) clearly indicate the deterioration of air quality over the region (Ding et al., 2007).

Transport pathways of ozone-rich air masses

According to international assessments (IPCC and TF HTAP), the influence of polluted air masses transported over long distances is among the largest uncertainties persisting in assessing air quality. The relevance of MOZAIC data has allowed us to improve our knowledge in linking features of the distribution of tracer species with emission sources, processes and transport mechanisms. Studies document (i) the supply of UT ozone by the North American and Eurasian continental boundary layers in spring and summer (Stohl et al., 2001 ; Cooper et al., 2005, 2006) and by the African boundary layer in fall and winter (Stohl et al., 2001), (ii) the transport of biomass burning plumes by convection and warm conveyor belts (Figure 7; Nédélec et al., 2005), (iii) the channelling of pollution by persistent atmospheric pressure centers (Kalabokas et al., 2007), (iv) the transport of pollution and biomass burning plumes by Harmattan and trades winds (Sauvage et al., 2005, 2007C) and by Hadley and Walker tropical cells, including lightning-NOx plumes (Sauvage et al., 2006, 2007AB).

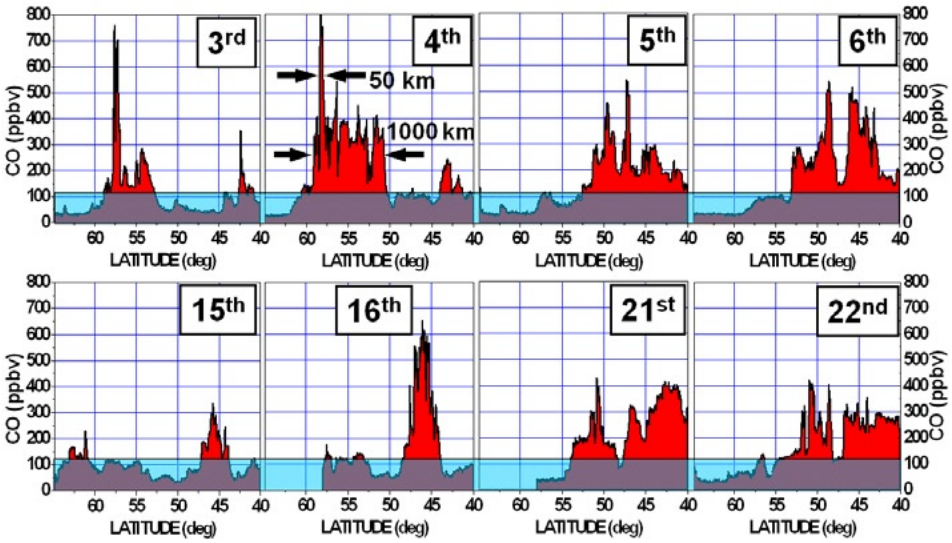

Figure 7: CO concentrations measured above 8 km over Asia between 40 and 65°N, for 8 days in June 2003. The light blue shading indicates the "background" CO level of 110 ppbv (from Nédélec et al., 2005).

Planetary Boundary Layer and Regional Air Quality

Although the MOZAIC program was not initially targeted to investigate processes governing the distribution of ozone in the boundary layer and regional air quality, the combination of data from MOZAIC and data from surface stations has been shown to be relevant in this respect. With a set of western European mountain stations covering a range of altitudes between 115 and 3580m, Chevalier et al. (2007) show that the surface measurements capture the climatological ozone stratification revealed by MOZAIC profiles in Frankfurt (Germany), thus pointing to the role of altitude and rapid vertical transport in the interpretation of ozone at mountain stations. The opportunity to get three-daily MOZAIC soundings in Frankfurt during the summer 2003 heat wave allowed Tressol et al. (2007) to accurately define the ozone and carbon monoxide anomalies within the boundary layer, which helps to size the difficulties that global CTMs face to reproduce such anomalies.

Satellite validation

Although satellite sensors provide quasi-global coverage, the data suffer from a lack of accuracy and vertical resolution and need to be validated against accurate in-situ datasets. MOZAIC data are used for the validation of trace gas retrievals from a growing number of satellite instruments, e.g., water vapor and ozone from the POAM III (Polar Ozone and Aerosol Measurement) instrument (Nedoluha et al., 2002; Prados et al., 2003), CO from the MOPITT intrument (Measurements of Pollution in the Troposphere) on the Terra satellite (Emmons et al., 2007), ozone and CO from the MLS instrument (Microwave Limb Sounder) onboard Aura (Livesey et al., 2007), CO from the ACE instrument onboard FTS (Clerbaux et al., 2007), and water vapour and temperature from GPS radio occultation (Heise et al., 2007). In the tropics, the consistency of measurements of lightning flashes and ozone precursors with satellite instruments (GOME, OTD and LIS) was evaluated using a CTM and MOZAIC measurements (Sauvage et al., 2007B).

Estimates of the impact of subsonic aircraft emissions on ozone

Ozone measurements from commercial aircraft programs such as MOZAIC are a reliable and representative source of information against which modelled impact of NO_x emissions from subsonic aviation upon the chemical composition of the atmosphere can be tested. This has been done in European projects dedicated to the impact of aircraft emissions, e.g. TRADEOFF (Isaksen et al., 2003) and SCENIC (Rogers et al., 2005), and continues in the on-going QUANTIFY project (http://www.pa.op.dlr.de/quantify/). During the nineties, the range of predicted perturbations on ozone decreased among the different modelling groups (Brasseur et al., 1998). The reference group of the University of Oslo (Gauss et al., 2006) predict aircraft-induced maximum increases of zonal-mean ozone between 3.1 ppbv in September and 7.7 ppbv in June in the tropopause region of the Northern Hemisphere and discusses the pros and cons of possible mitigation strategies, such as flight altitude and flying polar routes.

Summary

Over the period 1994-2007, the information from MOZAIC sensors flown on a fleet of initially five commercial aircraft has proven invaluable to improve our knowledge in the very sparsely sampled UTLS. The program has demonstrated the feasibility of routinely collecting high quality measurements from civil aircraft by delivering thirteen years of quality assured measurements of ozone and water vapour and seven years of quality assured measurements of carbon monoxide and of total odd-nitrogen. The costs of 500 Euro per flight (average of total costs over the entire period) compare favourably with those of ozone sondes, despite the large initial costs for aeronautical certification. With about 115 peer-reviewed articles having used MOZAIC data until now, the program has made an impact far beyond what had been anticipated at its inauguration. The main scientific achievements concern the UTLS seasonal distributions, stratosphere-troposphere exchange, tropospheric layering and the long-range transport of pollution, and begin to reflect the signs of interannual variability and decadal tendencies of trace gases. The MOZAIC data base provides a wealth of high quality information for the validation of global chemistry-transport models and the new generation of satellites and provides important data for estimating the impact of aircraft emissions. The dissemination of the results has contributed to a significant reduction in the uncertainties in assessing climate change, as acknowledged in international assessments (IPCC, WMO/UNEP, IGAC, SPARC and EUROTRAC).

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MOZAIC would not have happened without the enormous effort of its pioneer, Alain Marenco. We gratefully acknowledge the support for MOZAIC by the European Communities, Airbus, and INSU-CNRS. Particular thanks go to Lufthansa, Austrian, Air France and the former Sabena, who have carried the MOZAIC equipment free of charge since 1994.

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Acronyms

CLRTAP : Convention on Long Range Transport of Air Pollution
CNES: Centre National des Etudes Spatiales
CNRM : Centre National de la Recherche Météorologique, Météo-France
CNRS: Centre National de la Recherche Scientifique
ECMWF : European Center for Medium-Range Weather Forecasts
ETHER: Centre for Atmospheric Chemistry Products and Services, http://ether.ipsl.jussieu.fr
FZJ : Forschungszentrum Jülich, Germany
GEMS: Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data (http://www.ecmwf.int/research/EU_projects/GEMS/)
GMES: Global Monitoring of the Environment and Security
IAGOS: In-service Aircraft for a Global Observing System (http://www.fz-juelich.de/icg/icg-2/iagos)
INSU-CNRS: Institut National des Sciences de l'Univers - CNRS
IPCC : Inter-governmental Panel on Climate Change
MOZAIC (http://mozaic.aero.obs-mip.fr/web/)
TF HTAP: Task Force on Hemispheric Transport of Air Pollution (http://www.htap.org/10_2007/meeting.htm)

In-service Aircraft for Global Observations – the future

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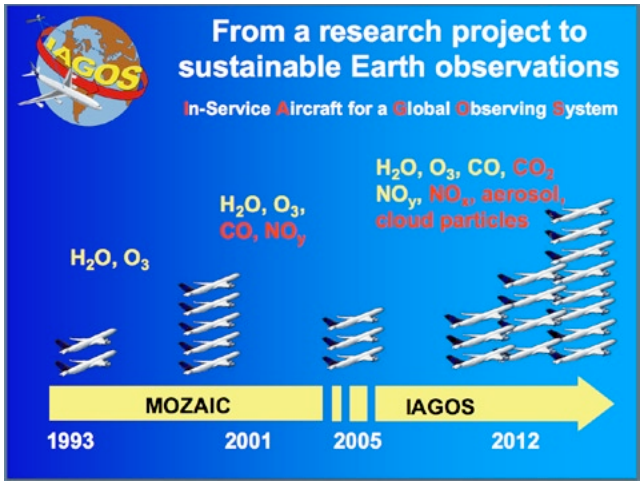
The European initiative IAGOS-ERI (In-service Aircraft for a Global Observing System – European Research Infrastructure) seeks to establish a sustainable distributed infrastructure for global observations of atmospheric composition from a large fleet of in-service aircraft. This will be achieved by installing autonomous instrument packages aboard, initially, 10-20 long-range aircraft of internationally operating airlines. IAGOS-ERI is one of the new research infrastructures proposed by the European Strategy Forum for new Research Infrastructures (ESFRI, http://cordis.europa.eu/esfri/) on the ESFRI Roadmap 2006. Whilst the initiative originated from the European MOZAIC project (see accompanying article by Cammas & Volz-Thomas), close links are already established with several other routine aircraft programmes. CARIBIC (see accompanying article by Brenninkmeijer et al.) has already become a member of IAGOS-ERI and it is the hope to establish formal links to other programmes in Japan (see article by Machida et al.) and North America within the next few years in order to build the aircraft component of the Integrated Global Atmospheric Chemistry Observations system (IGACO; Barrie et al., 2004).



IAGOS-ERI will provide high quality, in-situ observations of greenhouse gases and reactive gases, aerosols, and cloud particles in the tropopause region. This region is inadequately resolved by remote sensing from space and, on the other hand, is one of the most sensitive regions for climate change. At the same time, IAGOS-ERI will provide detailed vertical profiles in the troposphere, which are of paramount importance for predicting changes in local and regional air quality and its causes. The use of commercial aircraft allows the collection of highly relevant observations on a scale and in numbers impossible to achieve using research aircraft, and where other measurement methods (e.g., satellites) have technical limitations.

Table 1. Key areas defined by IGACO (air quality, oxidation efficiency, climate, and ozone depletion) with the species that need to be measured (from Barrie et al. 2004). The last column shows the species measured by IAGOS (green: less frequently).

Chemical Species	Air Quality	Oxidation Efficiency	Climate	Strat. O_3 Depletion	IAGOS
O_3	☑	☑	☑	☑	☑
CO	☑	☑	–	–	☑
$J(NO_2)$	☑	☑	–	–	
$J(O_1D)$	☑	☑	–	–	
H_2O (water vapour)	☑	☑	☑	☑	☑
HCHO	☑	☑	–	–	
VOCs	☑	☑	–	–	☑
Active nitrogen: NO_x Reservoir species: HNO_3	☑ ☑	☑ ☑	– –	☑ ☑	☑ ☑
N_2O	–	–	☑	☑	☑
SO_2	☑	–	☑	–	
Active halogen: BrO, ClO, OClO Reservoirs: HCl, $ClONO_2$ Sources: CH_3Br, CFC-12, HCFC-22, halons	☑ ☑ ☑	– – –	– – –	☑ ☑ ☑	☑ – ☑
Aerosol properties	☑	–	☑	☑	☑
CO_2	–	–	☑	–	☑
CH_4	–	☑	☑	☑	☑

The Rationale

Global climate change represents arguably the most serious environmental issue facing mankind today, with implications for global political stability and the global economy. Reliable predictions of the future climate using climate models are a central and fundamental requirement for determining future mitigation strategies.

According to the IPCC and other international assessments by WMO/UNEP, IGAC, SPARC and EUROTRAC-2, the largest uncertainties in assessing climate change and air quality persist in:

- The influence of water vapour, aerosol and clouds on climate, in particular in the UT;
- The unexplained apparent increase of H_2O in the UTLS;

Figure 1. The MOZAIC NO_y instrument in the avionic bay of a Lufthansa A340-300. The new IAGOS equipment will be installed at this location (see Fig. 2 for details). Photo: Udo Kröner, Deutsche Lufthansa.

- The indirect effect of aerosols on climate via influences on cloud formation and lifetime;
- The influence of ozone and aerosol which have been transported over long distances on regional air quality and climate;
- Climate-induced changes in the exchange between the stratosphere and troposphere, with implications for the budgets of O_3 and H_2O and with important feedbacks on air quality and climate.
- Possible changes in the oxidising efficiency of the atmosphere with consequences for the budgets of GHGs as well as CFC-substitutes involved in stratospheric ozone depletion;
- Feedback processes that control the atmosphere's carbon balance, such as potentially increased uptake by vegetation under elevated CO_2 levels.

IPCC (2001) emphasised the large uncertainties in the contributions of aerosols and clouds, in turn preventing an accurate prediction of future climate change and its causes. Specifically expressed was the need for continuation and extension of current measurement programmes.

The Integrated Global Atmospheric Chemistry Observations Theme of WMO (IGACO; Barrie et al., 2004), which was adopted by IGOS-P in 2005 and is now being implemented under GEO (Global Earth Observation), specifically recommends the continuation and extension of routine aircraft measurements as an essential complement to ground-based and satellite observations. IAGOS-ERI will serve to implement the aircraft component of IGACO. The measurements proposed in IAGOS closely match the priorities defined in the IGACO report, as highlighted in Table 1.

The Integrated Global Carbon Observations Theme (IGCO), also adopted by IGOS-P, recommends "atmospheric CO_2 concentration measured from in situ networks, including airborne" as core observations. Routine airborne CO_2 profile measurements are essential for the validation of satellite data of atmospheric CO_2 columns and possibly of vertical profiles obtained, e.g., from OCO, AIRS, SCIAMACHY and IASI. These would complement the CO_2 measurements being made on Japan Airlines commercial aircraft (see accompanying article by Machida et al.).

The importance of routine aircraft observations has been explicitly acknowledged by the GATO assessment for GMES (Braathen et al., 2004), the European contribution to Global Earth Observation.

The technical concept

The technical concept of IAGOS draws on the experience gained in MOZAIC (Measurement of OZone and Water vapour by Airbus In-service airCraft), which was funded between 1993 and 2004 by the European Commission as a series of three research projects. At the beginning of MOZAIC, airborne systems for ozone and water vapour, developed by Laboratoire d'Aerologie, Toulouse and FZ-Jülich, were installed in 1994 by Airbus on five A340 aircraft (see Cammas et al., this issue). The MOZAIC rack was modified in 2001 for additional measurements of CO (Nedelec et al., 2003), and an additional NO_y instrument (Volz-Thomas et al., 2005) was installed on one of the Lufthansa MOZAIC aircraft (see Fig.1) with the help of Lufthansa Technik.

As MOZAIC had been originally planned as a 3-year research project, several technical deficiencies later became apparent. Maintenance of the equipment was only possible in cooperation with the service departments of the participating airlines. An additional significant problem was that the original MOZAIC rack (see Fig. 2) was quite heavy (120 kg + 50 kg for the NO_y instrument) and space-consuming, and the original location of the MOZAIC rack is no longer available in newer models of the A340. After more than 10 years in service, the five MOZAIC aircraft are in danger of being replaced. No provision was foreseen in the Airbus service bulletins for installing the equipment on A340 aircraft already in service. Presently only three aircraft are still providing measurements, two operated by Lufthansa and one operated by Air Namibia.

At the end of the last MOZAIC project it was clear that for a sustainable long-term operation the instrumentation had to be redesigned completely in order to reduce weight and size by >50% and to obtain the general certification documents required for installation and maintenance aboard in-service aircraft. This situation was the starting point for IAGOS-DS, a European Design Study for new research infrastructures funded under the Sixth Framework Programme between 2005 and 2008 under the coordination of Research Centre Jülich (FZJ). A major task in IAGOS is the redesign of the former MOZAIC rack into a compact package of ca. 50kg . This task is being undertaken by the Laboratoire d'Aerologie, CNRS, Toulouse.

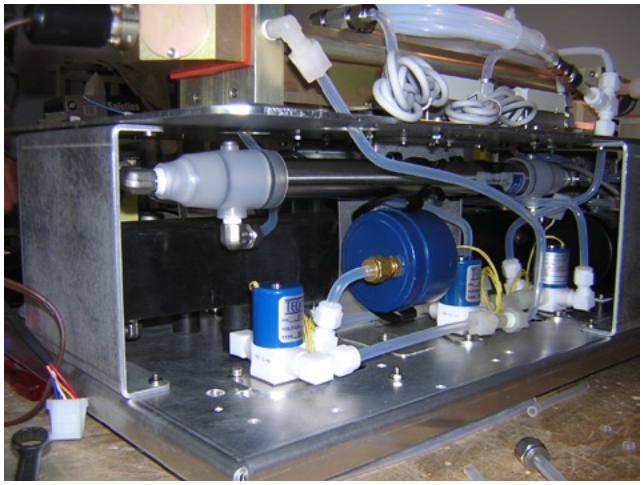


Figure 2. Original MOZAIC Rack in the avionic bay of an A340 with (from top) O_3-analyser, data acquisition system, CO-analyser, pumps, power supply). Photo: Udo Kröner, Deutsche Lufthansa. The photo below shows the prototype of IAGOS-Package 1, which contains the same components. Photo: Philippe Nedelec, Laboratoire d'Aerologie

Because of the scientific requirements outlined above, the MOZAIC partnership was expanded in IAGOS-DS in order to enhance the measurement capabilities with new instruments for cloud particles (University of Manchester), aerosols (Institute for Physics of the Atmosphere, DLR), and CO_2 (MPI-BGC, Jena).

The technical concept for the new IAGOS instrumentation and its installation into the avionic bay of an Airbus A340 is outlined in Figure 3. It comprises two instrument packages:

Package 1: O_3, H_2O, CO, cloud particles, data acquisition, and data transmission (near realtime for selected parameters)

Package 2a: NO_y (the sum of NO_x and its atmospheric oxidation products)

Package 2b: NO_x

Package 2c: Aerosol

Package 2d: CO_2

Package 1 will be installed on all aircraft, together with one option of Package 2. The instruments will have the full certification for installation and operation on Airbus A340 in-service aircraft, including the EASA Form One for instrument re-installation after maintenance.

The aircraft modification (Supplementary Type Certificate) is being achieved by CNRS together with an industrial partner, allowed to design and perform modifications on Airbus aircraft. IAGOS-DS will provide an Airbus Service Bulletin for the installation of the scientific instruments on in-service A340-300 aircraft. The required aircraft modification for installation of the systems will be performed during scheduled layovers of the aircraft, thereby avoiding immobilisation costs for the airlines.

IAGOS will develop certified maintenance procedures for periodic system replacements and quality assurance. The maintenance schedule for the instruments will be kept as minimal as possible, about every 6 months for Package 1 and 2-3 months for Package 2 (due to the need for exchanging gas cylinders and more frequent laboratory checks and calibrations). The automatic data transmission will allow daily surveys of the proper operation of the systems.

Automatic data transmission will be performed in two steps:

- fast transmission (resolution of ~15 minutes) for temperature, wind, O_3, H_2O and other suitable parameters for operational models. Here, a close collaboration is planned with AMDAR, which is already installed on many aircraft.
- slow transmission (on ground at the airport) of all data recorded during flight, using established ground

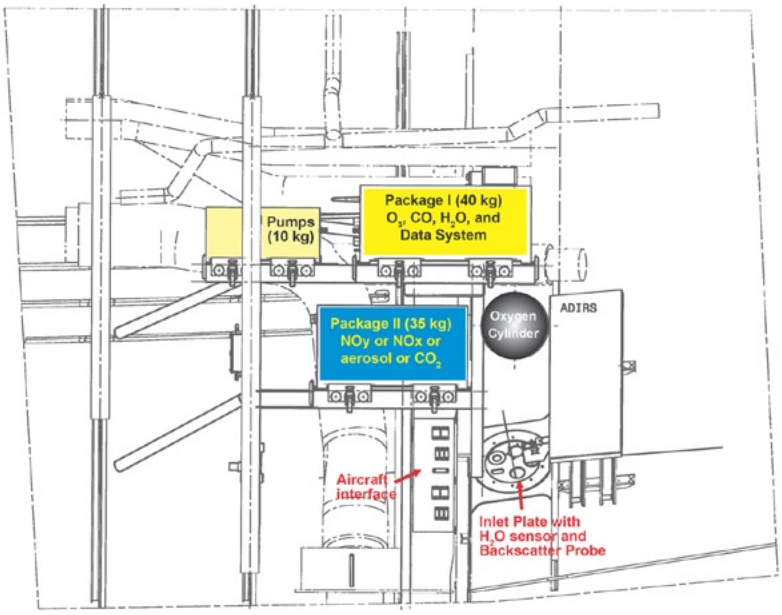


Figure 3. Technical concept for installation of IAGOS-instruments in A340

communication networks, such as GPRS or WiFi.

With the fast transmission, operational models will be able to receive the data for near real-time validation and assimilation. The data availability and usability of the database will be much improved in comparison to MOZAIC.

The second component of IAGOS-ERI is provided by the CARIBIC project (see Brenninkmeijer et al., this issue), which has joined the partnership for the new infrastructure. The CARIBIC aircraft (an A340-600 operated by Lufthansa) is equipped with an instrumented cargo container and a specially designed inlet system. It will provide a large suite of measurements, including those measured on all aircraft but also methane, N_2O, a suite of hydrocarbons and fluorocarbons, as well as detailed information on aerosol, and a MAX-DOAS system for remote sensing of atmospheric species. Isotopic information on trace constituents includes ^{14}CO, an excellent tracer for estimating global OH (e.g., Volz et al., 1981; Brenninkmeijer et al., 1992). Because of the larger technical complexity and the high transportation costs, the CARIBIC container will be deployed only during a few flights per month.

Where are we?

In the ongoing project IAGOS-DS, the aeronautic certification for the revised MOZAIC instruments will be obtained and the new technical development for aerosol and cloud particles are almost completed. For the autonomous measurement of CO_2 several solutions are currently under investigation. Certification of the new components will be achieved in the next phase (IAGOS-ERI), which will start in 2008 under FP7 for a duration of 4 years and serves to prepare the legal and logistical boundary conditions for operation of the new infrastructure in Europe. The new project will also provide some technical work necessary to bring the CARIBIC container (see above) into routine operation and to complete the work for the small instrument package.

Several airlines operating long-range Airbus aircraft have already expressed their interest in participating in the project – i.e., Lufthansa, Iberia, South African Airways, China Airlines Taiwan – ensuring global data coverage, including the Southern Hemisphere and the Pacific.

Major efforts are still required to secure the financial support for the initial investments and the operation of the systems. Figure 4 shows an estimate of the total cost for deploying instruments on 20 long-haul aircraft. The initial investment for new instruments, certification and aircraft modification are around 5 million Euro. Operation of the systems, including maintenance work, quality assurance and data base development/maintenance, as well as fuel costs for transportation of the additional weight, are estimated to approximately 250 thousand Euro per aircraft per year (see Figure 3).

Although a significant fraction of these cost will be provided by the participating institutions in the form of technical and scientific personnel, laboratory space and test equipment, it is envisaged that about 50 percent of the operational cost must be covered from additional resources, provided through national and international programmes, such as the GMES Atmospheric Service in Europe. The availability of long-term funding will be the limiting factor for the size of the fleet and securing the necessary resources will constitute an important part of the next phase of IAGOS.

The vision

IAGOS will make a strong effort to integrate the

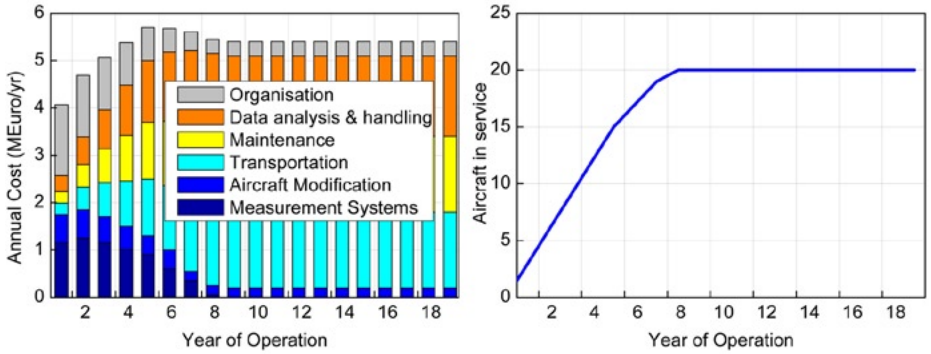


Figure 4. Projection for number of aircraft in service (right) and estimated annual cost for establishment and operation of 20 longhaul aircraft by cost categories (from IAGOS proposal to ESFRI). The figures do not include the cost for CARIBIC, which are estimated to approximately 1 Million Euro per year.

user communities and their needs in order to create a sustainable monitoring system for the global environment. Thirty years after the termination of the Global Atmospheric Sampling Program (GASP), the first programme using in-service aircraft (Nastrom, 1979), IAGOS opens the vision for routine monitoring of the atmosphere, where atmospheric observation systems, built and certified by aeronautical manufacturers, can be installed on a range of aircraft models. Such a system would ensure the largest degree of flexibility and would help even developing countries to install aircraft-based observations at moderate cost. Quality assurance is foreseen to be linked into the QA/QC-programmes of, for example, WMO-GAW. If this vision became true, IAGOS-ERI would enable the atmospheric community to capitalise on 10 years of investment into a number of research projects, leading to a new-generation global in-situ aircraft observation system. In this context, a very small instrument package (~20kg) is being developed in IAGOS under the lead of the University of Cambridge. This would, in the future, allow an even wider distribution at relatively moderate cost and would also allow access to smaller aircraft.

Acknowledgments

IAGOS is supported by the European Communities under FP7-Infrastructures. The strong support by Lufthansa and Lufthansa Technik for CARIBIC and MOZAIC is gratefully acknowledged. We would also like to thank our colleagues who have so emphatically supported the IAGOS initiative at a time when MOZAIC was in danger to come to an end. This support was crucial for the success of the IAGOS initiative.

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A new JAL project: CONTRAIL *Comprehensive Observation Network for TRace gases by AIrLiner*

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Introduction

Carbon dioxide (CO_2) in the atmosphere has been increasing since the beginning of the industrial age due to the rapid growth of human activities. There is now sufficient scientific evidence to show that this increase in atmospheric CO_2 is the main cause of the observed global warming and changes in climate (IPCC, 2007). How well we can predict the future states of climate, its change and variability depends greatly therefore on how well we can predict the future levels of CO_2 and other greenhouse gases in the atmosphere. The amount of CO_2 in the atmosphere is influenced not only by the anthropogenic emissions of CO_2 but also by exchanges of CO_2 with its two other major carbon reservoirs, the land biosphere and the oceans (WMO, 2006). The spatial and temporal variations of atmospheric CO_2 contain information about the nature and characteristics of the CO_2 exchange processes between the atmosphere and the land biosphere and the oceans. In the carbon cycle community, various top-down inverse calculation methodologies that use atmospheric transport models to exploit this property of atmospheric CO_2 have been developed to estimate the magnitude of CO_2 sources and sinks worldwide. However, right now there is an acute shortage of observations of CO_2 in the atmosphere to constrain and reduce large uncertainties in these inverse CO_2 flux estimates; the severity of this shortage is particularly critical in the portion of the atmosphere above the planetary boundary layer. It has been noted recently that a significant expansion of the existing aircraft observation programs and implementation of new ones will go a long way in improving the global CO_2 flux estimates (Stephens et al., 2007).

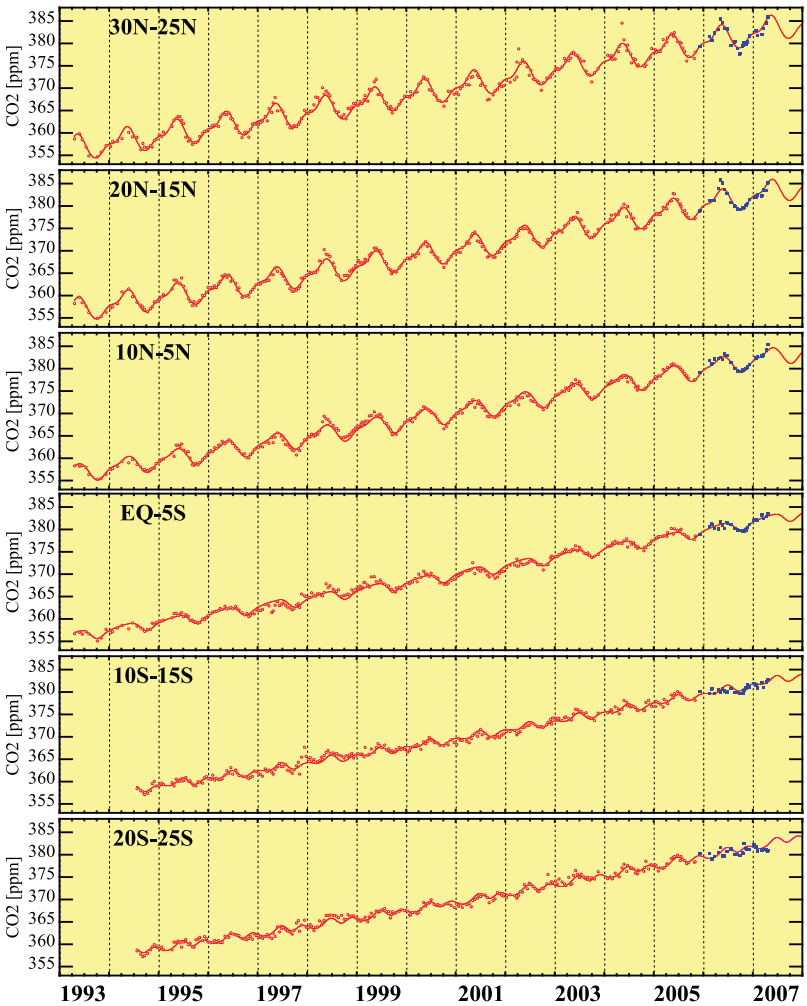

Figure 1. Time variations of CO_2 at about 10 km for six latitudinal bands between 30°N and 25°S over the western Pacific. The red and blue dots represent the observed data from the first and second phases of the JAL project, respectively. The red curve represents the climatology from the first phase of the JAL project during 1993 – 2005.

Until relatively recently, measurements of CO_2 in the "free troposphere" (i.e., above the planetary boundary layer) have been sporadic due to limited opportunities and campaign-style aircraft observations with limited spatial and temporal coverage (e.g. Machida et al., 2003; Sawa et al., 2004). However, commercial airlines provide a significantly more powerful observational platform for obtaining free tropospheric CO_2 systematically for long periods of time over a large geographical space. During the last 45 years, airline observations of CO_2 have been successfully implemented over several flight routes (e.g. Matsueda et al., 2002a, and references therein). 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 (Machida et al, 2007).

The first phase of the JAL project from 1993 to 2005 was carried out using an automated flask sampling system to obtain a long-term record of CO_2 and other trace gases. As shown in Figure 1, the CO_2 record over the western Pacific has provided valuable information on the latitudinal distribution of the atmospheric CO_2 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., 2002a, 2007). The JAL CO_2 data were employed to

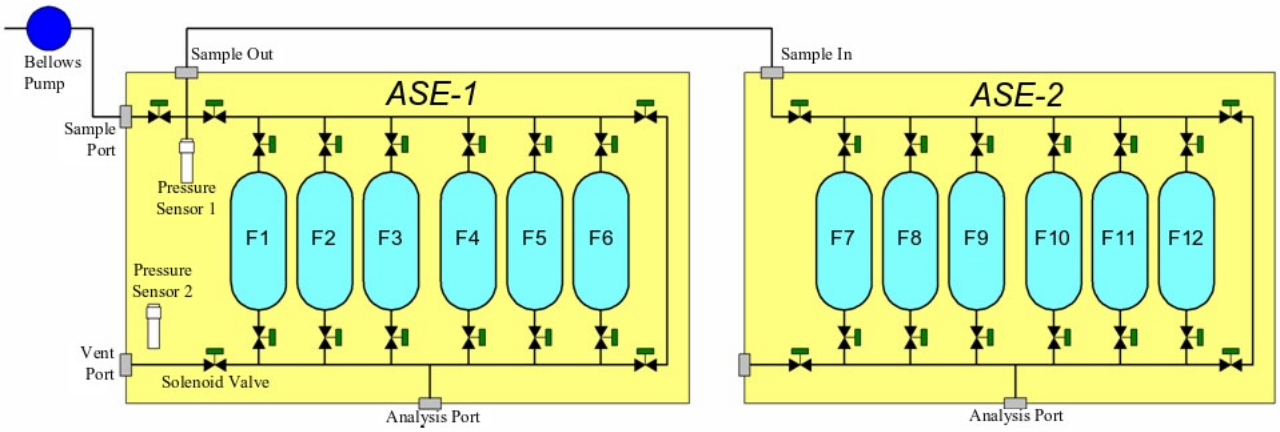

Figure 2. A schematic of ASE separated into two packages of ASE-1 and ASE-2. The 12 titanium air sample flasks are connected to the airflow via solenoid valves..

constrain an inverse estimate of the Southeast Asia source (Maksyutov et al., 2003) as well as for validating model transport (Shia et al., 2006) and satellite-based CO_2 observations (Chédin et al., 2003; Chahine et al., 2005; Engelen et al., 2005). In addition, a unique record of carbon monoxide measurements from the first phase of the JAL project captured well the significance of the dispersion of the biomass-burning emissions associated with the strong El Niño in 1997 (Matsueda et al., 1999, 2002b; Taguchi et al., 2002; Duncan et al., 2003).

For the second phase of the JAL project (*C*omprehensive *O*bservation *N*etwork for *TR*ace gases by *AI*r*L*iner: CONTRAIL; http://www.jal-foundation.or.jp/html/shintaikikansokue/Contrail_index(E).htm), a new *A*utomatic air *S*ampling *E*quipment (ASE) for flask sampling and a new *C*ontinuous CO_2 *M*easuring *E*quipment (CME) for in-situ CO_2 measurements were installed on Boeing 747-400 and Boeing 777-200ER aircraft (Machida et al., 2007). In all, one or both of these instruments have been installed on five Boeing aircraft operated by JAL with regular flights from Japan to Australia, Europe, East and Southeast Asia, Hawaii, and North America, providing significant spatial coverage, particularly in the Northern Hemisphere. Shown in Figure 1 are the ASE flask CO_2 data obtained since December 2005 over the same Japan-Australia JAL route as before, confirming the consistency in the continuity of the observational record between the first and second phases of the JAL project. In the following sections, details of the new observation systems and first preliminary measurement results from the CONTRAIL project are presented.

Figure 3. Photo of ASE-1 and ASE-2. The surface panels are detached.

Measurement Equipment

Automatic Air Sampling Equipment (ASE)

The basic design of the new ASE instrument for flask air sampling is similar to that of the previous ASE used in the first phase of the project (Matsueda and Inoue, 1996), but all of the components have been newly replaced. One of the most significant advancements in the new ASE is a sampling operation system, which can be automatically controlled based on the real-time monitoring of the flight navigation data from the ARINC 429 data bus of the aircraft.

Figure 2 shows a schematic of the new ASE instrument, which is separated into two packages (ASE-1 and ASE-2) for easy handling of each package (430 mm length x 215 mm width x 531 mm height); ASE-1 and ASE-2 weigh 17.9 kg and 15.6 kg, respectively. The main components of each ASE package are six flasks, solenoid valves, and pressure sensors (Figure 3), so that 12 air samples in total can be collected during one flight. The cylindrical flasks with an internal volume of about 1.7 L are made of titanium.

The automated air sampling is performed by a specially designed control board attached to each of the ASE-1 and ASE-2 packages. These boards control the pump and solenoid valves for air sample collection based on inputs from the ARINC429 data bus on the aircraft and the pressure sensors. Before each flight, sampling operation parameters (such as sampling locations, altitudes and air pressure) in the control board are set by a laboratory computer. During the flight, the control board records the ARINC parameters at 12 air

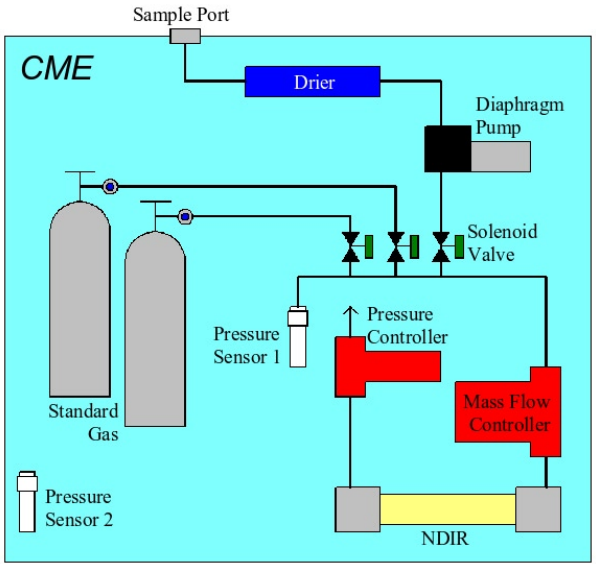

Figure 4. A schematic showing air flow in the Continuous CO_2 Measuring Equipment (CME). The main components of drier, pump, solenoid valves, two standard gas cylinders, and NDIR are shown.

sampling locations, as well as information about the valve and pump operations, so that we can check the sampling procedure after the flight. Thus, the ASE design enables us to operate automatically without any assistance of crew members onboard the aircraft.

After the flight, the ASE-1 and ASE-2 are unloaded from the aircraft and transported to the National Institute for Environmental Studies (NIES) for high-precision analysis of the mixing ratios of CO_2, methane (CH_4), carbon monoxide (CO), molecular hydrogen (H_2), nitrous oxide (N_2O), and sulfur hexafluoride (SF_6) and stable isotope ratios of CO_2 and CH_4.

Continuous CO_2 Measuring Equipment (CME)

Figure 4 shows a schematic diagram of the CME package (264 mm length x 330 mm width x 570 mm height), which weighs about 25 kg. The main components are a drier, a pump, solenoid valves, a flow controller, a pressure controller, NDIR (LI-COR, LI-840) analyzer, two standard gas cylinders, and a programmable controller/datalogger device (Figure 5). Air samples are drawn by a diaphragm pump from the intake port mounted on the air conditioning duct and then introduced into the analyzer. In front of the pump, a drier tube packed with CO_2-saturated magnesium perchlorate is used to remove water vapor. The flow rate of the sampled air into the NDIR cell is kept constant at 150 standard cc per minute (sccm) by a mass flow controller. In addition, the absolute pressure of the sampled air in the NDIR cell is maintained at 0.110 MPa by an auto pressure controller to avoid signal drift in the NDIR associated with changes in cabin pressure.

The solenoid valves switch flows from sampled air to standard gases for regular calibration of the measurements by the NDIR at an interval of 10 minutes during the climb and descent, and 20 minutes during the level flight. Two CO_2-in-air standard gases of about 340 ppm and 390 ppm in 2.3-L high-pressure aluminum cylinders are pressurized to about 12 MPa. The automated operation is performed by the programmable controller/datalogger device. This device controls the pump, the solenoid valves for the flow selection (either air sample or standard gas), and the NDIR on the basis of the information received from the flight data system on the aircraft. To obtain the required flight parameters, an ARINC 429 Serial Port Adapter interface is connected to three channels of the ARINC buses of the aircraft data system. The parameters received by the controller/datalogger device are date, time, pressure altitude, radio altitude, latitude, longitude, ground speed, static air temperature, wind direction, and velocity. In order to avoid sampling of polluted air (high CO_2 and H_2O) in the lower layers of the atmosphere, air sampling does not start until the radio altitude registers 1200 feet during the climb, and stops at 1200 feet during the descent. All of the data from the aircraft data system, the NDIR signal, flow rate, cell pressure and supplied air pressure measured by pressure sensor 1 are recorded every 10 seconds during the climb and descent and every 1 minute during the level flight. The time interval of 10 seconds corresponds to about 100m in altitude, while that of 1 minute corresponds to 10-20 km in the horizontal. The measurement precision is estimated to be 0.1ppm (one standard deviation) when using 10-second averaged data. Thus, CME enables us to obtain the fine structures of the atmospheric CO_2 distribution from all of the flights.

Figure 5. Photo of CME. The surface panels are detached.

Figure 6. Forward cargo compartment of Boeing 747-400. ASE and CME are installed beside the water tanks in the forward cargo compartment.

Figure 7. Forward cargo compartment of Boeing 777-200ER. Only the CME is installed on the sidewall of the forward cargo compartment.

Instrument Installation and STC Issuance

The ASE and CME instrument packages are mounted on two aviation-approved racks installed at the back of the forward cargo compartment of a Boeing 747-400 (Figure 6). A pump for the ASE is also located in the same area where all the instruments are easily accessible for maintenance and wiring of electric power supply and aircraft data buses. On a Boeing 777-200ER, only the CME instrument is installed (Figure 7). To comply with the safe operation of the aircraft during flight, emergency shutdown of both the ASE and CME operations is implemented (Machida et al., 2007).

After various tests, a Supplemental Type Certificate (STC) to install ASE and CME on Boeing 747-400 series aircraft was issued by the Federal Aviation Administration (FAA) on October 26, 2005. A similar STC document was issued by the Japanese Civil Aviation Bureau (JCAB) on November 2, 2005. Thus, ASE/CME instrument package can be installed on any Boeing 747-400 airplane operated by other airlines with only minor adjustments/changes. A certificate to install CME on Boeing 777-200 series was obtained from the FAA on March 22, 2006, followed by JCAB approval on March 31, 2006.

Observation and preliminary results

Flight area and frequency

We prepared five aircraft operated by JAL, two Boeing 747-400 and three Boeing 777-200ER, to be fitted with the instruments. The modifications to the last plane were completed in November 2006. The CME is installed on all five planes, but the ASE is installed only on the Boeing 747-400 planes that fly twice a month between Sydney, Australia and Narita, Japan to perform flask measurements of CO_2 and other trace gases. Maintenance of the CME is performed every 1-1.5 months, with replacement of the standard gas cylinders and the chemical drier, as well as data retrieval. This relatively long-term maintenance-free operation allows frequent global-scale coverage by two or three commercial airplanes fitted with the CME.

Typical flight routes for CME observations from November 2005 to April 2007, along with airport code and flight frequency (as represented by the number of vertical profiles taken), are shown in Figure 8. More than half of the flights from Narita, Japan have been to East and Southeast Asia, and about 21% to Europe via Siberia. Other flights to North America and Australia are also shown in the diagram.

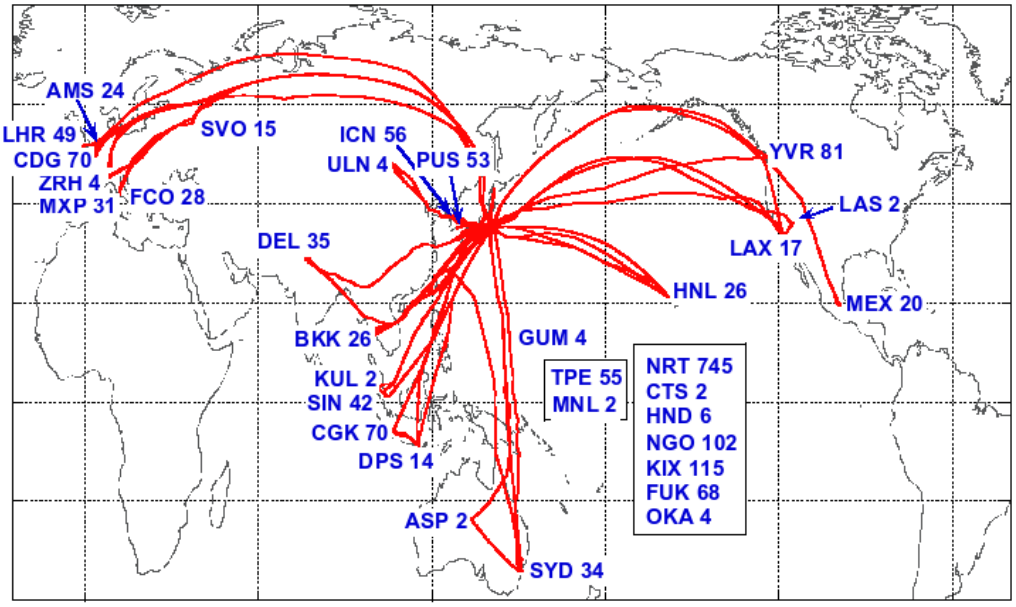

Figure 8. Flight routes of CME observation and destination airport. The numbers indicate the number of vertical profiles obtained over each airport from November 2005 to April 2007.

CO_2 data consistency check between CME and ASE

Latitudinal distributions of CO_2 in the upper troposphere between Sydney and Narita measured by CME are compared in Figure 9 with those obtained by ASE during the December 8, 2005 flight. There is a relatively good agreement, i.e. within ±0.2ppm. Data from other flights carrying ASE and CME show similar agreement. Analytical precision and accuracy are relatively better for ASE since flask samples are analyzed by a LI-6252 (LI-COR) in our laboratory under stable temperature and pressure conditions. Therefore, the data agreement indicates a high reliability in CO_2 measurements by a small NDIR inside the CME.

Horizontal distribution

In the CONTRAIL project that started in late 2005, measurements from CME in particular have provided detailed spatial and temporal distributions of CO_2 at 9-12 km heights, encompassing upper tropospheric and lower stratospheric regions in the extratropics. The observations in these regions have significant scientific importance in the understanding of the global carbon cycle and atmospheric transport. In this context, the JAL flights between Japan and Europe are of special interest because their flight levels take the planes quite often into the lower stratosphere.

Although gradients of CO_2 across the tropopause have been observed previously during several aircraft campaigns (e.g., Hoor et al., 2004; Sawa et al., 2004) and commercial flights (Nakazawa et al., 1991), these measurements have been spotty and give only a very limited picture of the CO_2 variation in regions near the tropopause. Understanding of the exchange processes across the tropopause can be enhanced by detailed analyses of CO_2 measurements with high spatial and temporal resolutions. Our measurements taken on commercial aircraft can provide such data. Since we know with a sufficient degree of understanding the seasonal variation of CO_2 in the lower troposphere, a detailed characterization of the CO_2 seasonal cycle near the tropopause will allow us to diagnose the dynamics of the transport and mixing processes across the tropopause,

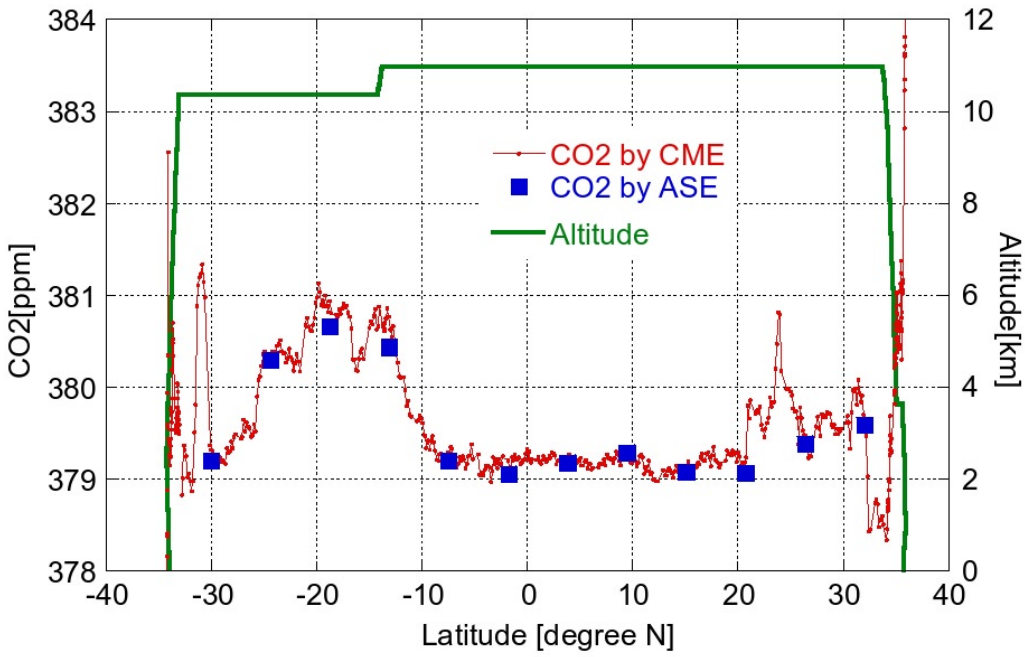

Figure 9. Latitudinal distributions of CO_2 mixing ratio in the upper troposphere measured on December 8, 2005 by CME and ASE. The green line indicates flight altitude.

as well as the transit time scale (Nakazawa et al., 1991; Boering et al., 1996; Strahan et al., 2007). In addition, the measurements taken between Japan and Europe will allow us to enhance our knowledge of sources and sinks of CO_2 over Siberia.

The potential scientific significance of our measurements taken with the JAL aircraft is demonstrated in Figure 10. Data from two JAL flights, one from Narita to Amsterdam and another one from Narita to Vancouver, on 22 December 2006 are pieced together to form a single snapshot of a long longitudinal profile of CO_2 influenced by different airmasses. Lower CO_2 values at altitudes of 10-12 km over the eastern North Pacific and the eastern Siberian regions are associated with high potential vorticity (PV) areas north of the polar jet (i.e., indicating that these samples are influenced by lower stratospheric air). On the other hand, higher CO_2 values over the western North Pacific and the western Eurasian continental regions correspond to upper tropospheric air with lower PV values. Thus, regular and frequent commercial flights conducted with five aircraft will provide high resolution temporal and spatial CO_2 data that will help us increase our understanding of the transport of CO_2 and constrain the source and sink estimates by validating atmospheric transport models.

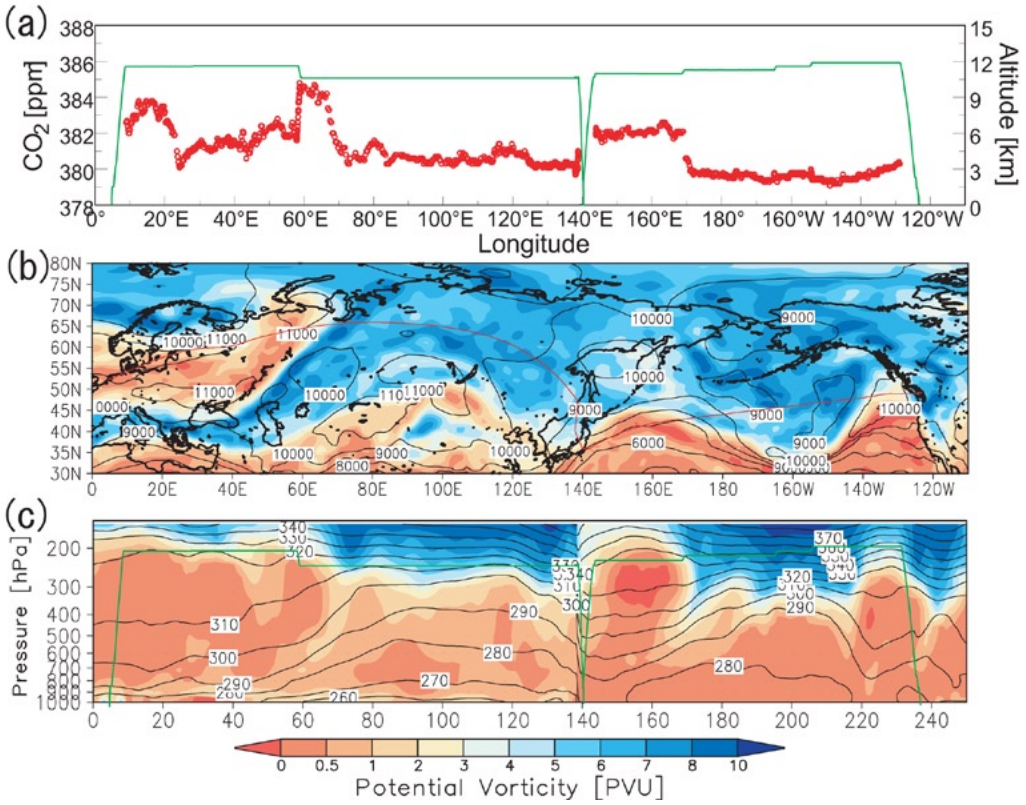

Figure 10. CO_2 mixing ratios (red circles) observed by CME during the level flights from Narita to Amsterdam and from Narita to Vancouver on 22 December 2006 with flight altitudes denoted by the green lines (a). Potential vorticity (shaded) and geopotential height (black contours; units in m) on the 320 K potential temperature surface at 12 UTC on 22 December 2006, with flight routes indicated by red lines (b). Cross section along the flight routes of potential vorticity (shaded) and potential temperature (black contours; units in K) with flight levels denoted by the green lines (c). Meteorological data obtained from the Japan Meteorological Agency Climate Data Assimilation System (JCDAS) reanalysis data (Onogi et al., 2007). Regions of high potential vorticity (blue) are indicative of stratospheric air and lower PV of tropospheric air.

Vertical distribution

More than 2000 vertical profiles of CO_2 have been obtained worldwide from November 2005 to April 2007. Although it is too soon to characterize the seasonal and geographical variations of these profiles, sample profiles are depicted in Figure 11. The Figure shows vertical CO_2 distributions obtained over Narita (NRT) and Jakarta, Indonesia (CGK) on 5-8 November 2005. High CO_2 values near the surface reflect CO_2 contaminated by human activities around the airport. Vertical profiles in the free troposphere above the planetary boundary layer are tightly bound over both airports, indicating high reproducibility of CME observation.

In the context of what vertical profiles tell us about the nature of atmospheric transport and various sources and sinks of CO_2, it is interesting to note the difference in the vertical profiles taken over NRT and CGK. A positive vertical gradient with higher CO_2 mixing ratios in the lower altitudes over NRT (Figure 11a) is caused by CO_2 release from land vegetation and fossil fuel use in this area. On the other hand, relatively constant CO_2 with height over CGK is reflective of the tropical region, with weak surface CO_2 emission and efficient vertical air mixing; this is similar to the results from the aircraft campaign observations obtained by Machida et al. (2003). The preliminary results shown in Figure 11 complement the horizontal distribution of CO_2 already mentioned above, and together they demonstrate the potential scientific benefit of detailed vertical and horizontal measurements of CO_2 at different locations to obtain a consistent 3-dimensional spatial and temporal distribution of CO_2 throughout the depth of the atmosphere, for a better understanding of the transport processes on a global scale and their interactions with CO_2 fluxes.

Conclusion and future prospect

With cooperation and encouragement from Japan Airlines, we have been actively carrying out systematic measurements of CO_2 and other trace gases in the

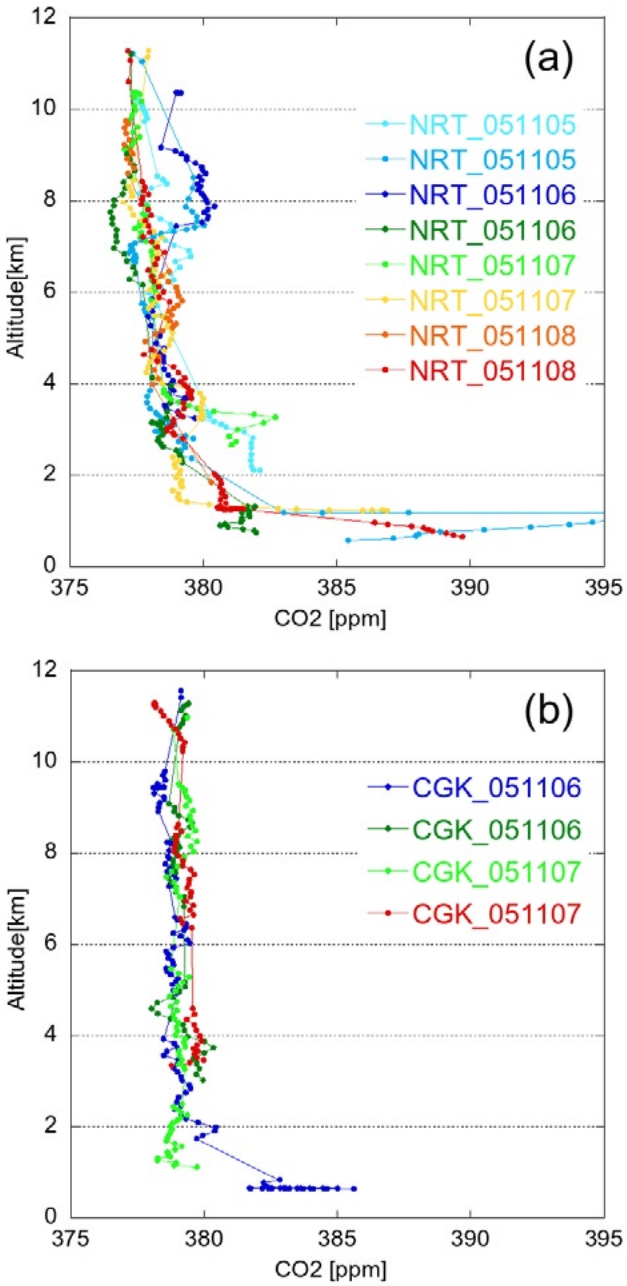

Figure 11. Vertical distributions of CO_2 mixing ratio observed on 5-8 November 2005 over Narita, Japan (a) and Jakarta, Indonesia (b).

atmosphere using commercial aircraft since 1993. During the first phase of the JAL project (1993-2005), an automated flask sampling system was used to obtain measurements of CO_2 and other trace gases over the western Pacific between Japan and Australia. In the second phase we have significantly expanded the program, to cover additional regions from Japan to Europe, East and Southeast Asia, Hawaii and North America, using five JAL aircraft. We have made substantial improvements in the ASE flask sampling system and have developed a new instrument, CME, to obtain continuous in-situ measurements of CO_2. The CME and ASE instrument packages have been certified by the FAA and JCAB to operate on Boeing 747-400 series, while only the CME has been certified to operate on Boeing 777-200 series. Thus these instruments can be used by any airline that employs any of these two Boeing series aircraft, providing a very powerful observational platform for making precise and accurate measurements of trace gases deep in the atmosphere on a global scale.

The ASE can collect 12 flask samples for precise analyses of CO_2 and other trace gases, while the CME allows measurements of high-resolution CO_2 that can provide wide spatial coverage. Data from these instruments provide detailed horizontal and vertical variations of atmospheric trace gases at spatial and temporal resolutions appropriate for enhanced quantitative understanding of the global carbon cycle and atmospheric transport and of mixing processes particularly across the tropopause and between the Northern and Southern Hemispheres. The data can also be used to validate atmospheric transport models and to provide a very powerful constraint to inverse CO_2 flux estimates, as well as to validate CO_2 measurements by the satellites GOSAT and OCO, to be launched soon.

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Observations of the atmospheric composition over Russia using a mobile laboratory: the TROICA experiments

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The mobile laboratory of the TROICA-10 experiment

Variations in the Earth's climate and in atmospheric composition have pronounced regional distinctions. The IPCC AR4 (2007) report demonstrates that, for the most recent 100-year period, the most intensive warming occurred in Siberia. Such changes in climate are accompanied by changes in the atmospheric gas and aerosol composition and in terrestrial ecosystems. A rapid development of the global atmospheric monitoring system in the 1990s, unfortunately, didn't cover Russia. Moreover, the efficiency of the Russian meteorological network in this period degraded; the stations to monitor background levels of O_3, NO_x, SO_2, and aerosol were terminated. In this crucial period, Dr. Prof. P.J. Crutzen proposed to the Obukhov Institute of Atmospheric Physics (RAS) that he organize observations of surface ozone and its precursors from trains traversing the Moscow–Vladivostok Trans-Siberian Railroad. In November–December 1995, the first experiment of such a kind was performed utilizing a specialized car-laboratory equipped with financial support from the Max Planck Institute of Chemistry (MPIC). It was shown that reliable information on the regional surface-air composition can be obtained if the car-laboratory is coupled at the head of a train moving along electrified railroads.

Thereafter, similar experiments were performed yearly up to 2001. These measurements now comprise the Trans-Siberian (or Trans-continental) Observations Into the Chemistry of the Atmosphere (TROICA). Owing to the active participation of Dr. C.A.M. Brenninkmeijer and his group, the set of monitored gaseous pollutants was substantially extended and studies of the isotope composition of CO and CH_4 were initiated. Cooperation with the Karpov Institute of Physical Chemistry allowed expansion of the scope of aerosol studies. With time, researchers from the NOAA Earth System Research Laboratory (ESRL), the University of Helsinki (UH), and the Finnish Meteorological Institute (FMI) joined the project.

In 2003, a new mobile laboratory (Foto) was constructed and equipped. It consists of two cars intended for continuous measurements of gas and aerosol concentrations and of radiative and meteorological parameters; it also housed equipment for chemical analysis of radioactive pollutants and of air, water, soil, and vegetation samples, including those in extreme ecological situations. The chemical laboratory is equipped with a specialized motor-car allowing for measurements and sampling over remote regions.

The laboratory is equipped with instruments allowing for measurement of O_3, NO, NO_2, CO, CO_2, CH_4, SO_2, NH_3, THC, ^{222}Rn, and 3-10μm aerosol (including soot); quantitative monitoring of surface air and measurements of the vertical O_3 and NO_2 profiles, solar radiation, meteorological parameters, 0-600m temperature vertical profiles; and some other parameters and atmospheric characteristics (Table 1). Further instruments allow

Table 1. TROICA observation system

Concentration of surface gases	O_3, NO, NO_2, CO, CO_2, SO_2, CH_4, NH_3, THC
Surface aerosols	Size distribution (2 nm-10 μm), scattering coefficient, mass concentration; black carbon
Remote sensing	O_3 and NO_2 total content and vertical profile (express Umkehr technique), O_3 stratospheric and mesospheric profile (microwave technique), NO_2 in boundary layer (DOAS)
Solar radiation	Integral, UV-A, UV-B, photodissociation rate J(NO_2)
Meteorology	Pressure, temperature, humidity, wind (speed and direction), temperature profile (0-600 m)
Sampling	Greenhouse gases and VOC; chemical, elements and morphological composition of aerosol, isotope composition of CO_2CO_2, and CH_4
Other	Navigation parameters (GPS), ^{222}Rn, radionuclides, TV picture of road-side territory (both sides), TV picture of cloudiness, samples of water, soil, vegetation

for air and aerosol sampling for subsequent chemical, isotope, and morphology analyses.

The following instruments belonging to other institutes have also been deployed at various times during the TROICA experiments: multi-channel chromatograph ACAT-IV, measuring ozone-destroying and greenhouse gases (NOAA-ESRL); proton-transfer-reaction mass spectrometer (MPIC); different mobility particle sizer (UH); particle into liquid sampler with ion chromatograph (FMI); and some others.

The entire set of the observational data and navigation parameters are collected in a single database. This database also contains information on all objects and observation conditions. The operators collected this information via PC almost continuously during each of the expeditions. This database also contains the roadside-territory photos taken in motion at 5s intervals by the TV cameras installed on both sides of the car.

To simplify working with a database of such a large volume, a specialized interface is utilized. Using data on the ratios of the concentrations of different pollutants allows identification of the polluted (urban or industrial), rural, and background route sections. Such studies showed that the leading electric locomotive and opposing trains influence the monitoring data only slightly. The latter effect in particular is negligible also because the time intervals between opposing trains are as long as 25–35 min.

Since 1995, ten TROICA experiments have been performed (Table 2 and Fig. 1). Of the several routes taken, the expeditions along the Trans-Siberian Railroad were most numerous. In 2000, several meridional expeditions and a three-week period of stationary monitoring at each of four climatic regions were performed during the Spring season (TROICA-6). In October 2006, the Moscow megapolis was circumnavigated (mean radius ~70km) three times via the electrified circuit railroad and Moscow city proper was traversed twice (TROICA-10). In the course of TROICA-5, shipboard atmospheric monitoring was performed while traveling down the 2000 km Ob River, starting from the city of Novosibirsk.

Table 2. TROICA experiments: dates and routes.

Experiment	Work period	Route
TROICA-1	1995 Nov 17 – Dec 2	N.Novgorod-Khabarovsk-Moscow
TROICA-2	1996 Jul 26 – Aug 13	N.Novgorod-Vladivostok-Moscow
TROICA-3	1997 Apr 1 – Apr 14	N.Novgorod-Khabarovsk-Moscow
TROICA-4	1998 Feb 17 – Mar 7	N.Novgorod-Khabarovsk-N.Novgorod
TROICA-5	1999 Jun 26 – Jul 13	N.Novgorod-Khabarovsk-Moscow/Ob river boating
TROICA-6	2000 Apr 6 – Jun 25	Moscow-Murmansk-Kislovodsk-Murmansk-Moscow/Observations at 4 stations
TROICA-7	2001 Jun 27 – Jul 10	Moscow-Khabarovsk-Moscow
TROICA-8	2004 Mar 19 – Apr 1	Moscow-Khabarovsk-Moscow
TROICA-9	2005 Oct 4 – Oct 18	Moscow-Vladivostok-Moscow
TROICA-10	2006 Oct 4 - Oct 7	Around and cross section Moscow megapolis
TROICA-11	2007 July 22-Aug. 5	Moscow-Vladivostok-Moscow

Figure 1. Routes of the mobile laboratory.

Most of these measurements had a frequency of 6 points per minute; thus, it is obvious that the mobile laboratory can be utilized to measure features of the atmospheric composition and state on scales ranging from continental to ten meters. A great number of these features have been discussed in the published literature. Here we note the most important features and discuss some more recent results.

The following characteristic features of the surface ozone concentration distribution over the continent are revealed: the ozone concentration is almost uniform in cold periods and increases eastward in warm periods. The surface ozone concentration temporal variations are controlled by Arctic-air advection and by temperature inversions, which are most frequent and intense over the central region of the continent. The ozone-concentration field is significantly affected by stratospheric injections of ozone-rich air and by photochemical processes in polluted air and in plumes of forest and steppe fires. The ozone concentration increase in the eastward direction across the continent is the result of the combined effect of these factors (Elansky et al., 2001; Shakina et al., 2001).

The CO and CH_4 isotope composition monitoring performed under the guidance of C.A.M. Brenninkmeijer provided important information on the sources of these gases. It was shown that the significant increase in the ozone concentration over background levels in western Siberia is of biogenic origin. Meanwhile, natural-gas leakages during the production, processing, and transportation of natural gas contribute to the atmospheric methane concentration. Shipboard measurements showed increases in thermogenic methane concentration of up to 50% above background levels at a distance of 0.5 km from gas-production points. The forest fires occurring in eastern Siberia and biomass burning occurring in China represent significant sources of atmospheric CO and CH_4 (Bergamaschi et al., 1998; Oberlander et al., 2002; Rockmann et al., 1999; Tarasova et al., 2006).

The $^{14}CO_2$ spatial distribution across Eurasia is determined from air analyses performed in the University of Colorado (Turnbull et al., 2007). Local perturbations are caused by easily identifiable sources associated with a nuclear power plant and urban pollution. Other data show an increase in the $^{14}CO_2$ concentration when progressing from western Russia to eastern Siberia. This is consistent with the gradual dispersion of fossil fuel plumes from the heavily populated European region, across northern Asia (Turnbull et al., 2007).

J. Elkins, D.F. Hurst, and P.A. Romashkin measured the atmospheric concentrations of ozone-depleting substances and estimated their emissions from the Russian territory. These emissions are insufficient in magnitude to play a major role in recent global emission shortfalls. On the other hand, based on the rather intense CFC-12 emissions estimated for the Russian territory from their observational data of 2001, recent estimates of the global CFC-12 reserve appear to be too low (Hurst et al., 2004).

Particular features in the aerosol (including black carbon) distribution over the continent were revealed. Each of the expeditions measured aerosol concentration, size distribution, and chemical composition; therewith, the most powerful local and regional aerosol sources were revealed and the contribution of long-range transport to the atmospheric aerosol content was estimated. It has been noted that vertical transport in mountain regions plays an important role in the balance of super-fine particles in surface air. During the TROICA-9 experiment, S.Kuokka and E.Vartiainen measured atmospheric ions and the size distributions of aerosol particles and showed that these parameters are dependent on the concentrations of such atmospheric pollutants as nitrogen oxides and carbon monoxide. Elevated concentrations of such burning-biomass tracers as oxalate, potassium, and levoglucosan allowed identification of some episodes of forest fires. Cluster ions below 1.5-nm size were detected. Their concentrations varied between 100 and 5000 cm^{-3}, and the negative cluster ions were, as a rule, much more abundant than the positive ones. A few events of particle formation were registered (Vartiainen et al., 2007).

Recently we have worked out a method for classifiying data into three groups corresponding to the surface-air composition in industrial, rural, and remote areas. This allows us to study the characteristic temporal and spatial variations of pollutants within these regions. This problem is quite soluble, because the body of information for each group is sufficiently great. Such a classification is very important for a country having no atmospheric monitoring network and which is characterized by a long history of disproportionate economic development across different regions. Practically, half of the Russian population resides on 15% of the land, and the concentrations of pollutants in these regions

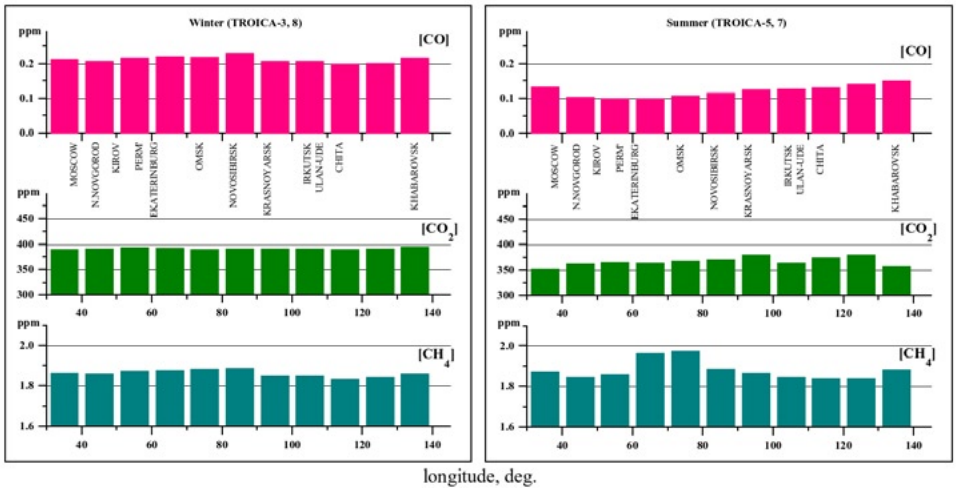

Figure 2. Spatial distribution of CO, CO_2 and CH_4 concentrations in background conditions between Moscow and Khabarovsk for winter and summer experiments.

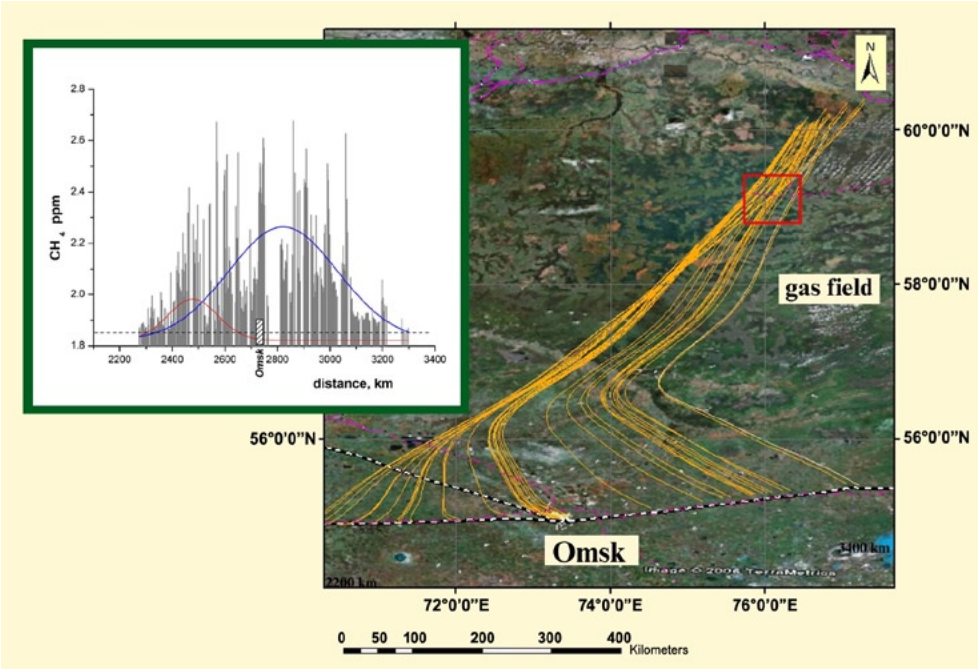

Figure 3. Transport of a methane plume from a West Siberia gas field to the Trans-Siberian railway (TROICA-5).

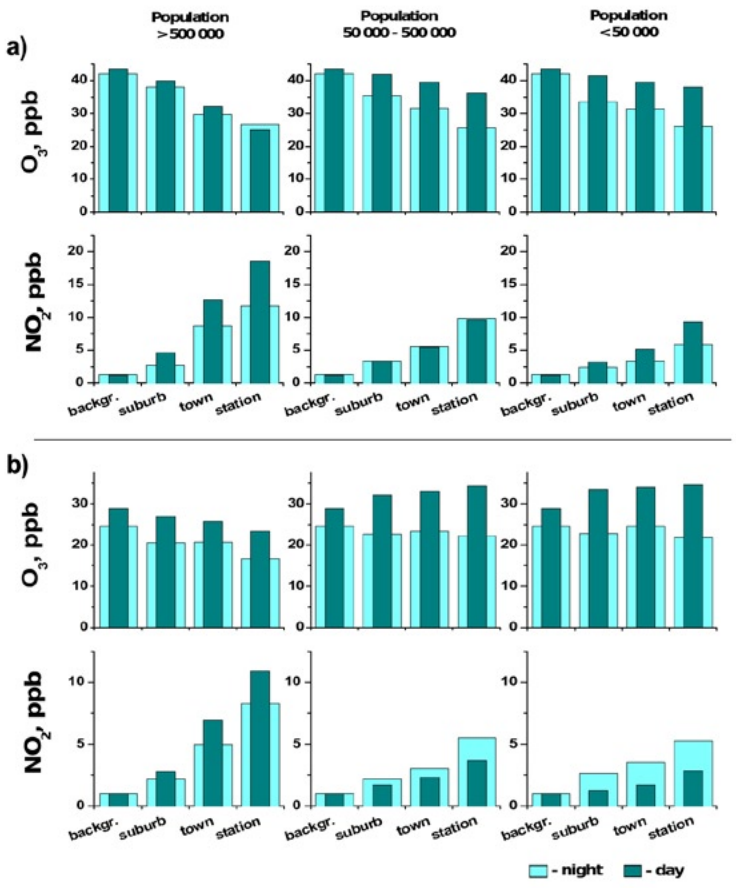

Figure 4. Average surface O_3 and NO_2 concentrations at the suburb, town and center of town (station) in comparison with background levels for three groups of cities during day and night: a) winter; b) summer.

exceeds the established maximum permissible levels. Meanwhile, effectively no economic activity occurs in over 65% of the Russian territory, and ecosystems in these regions are nearly in their natural state.

While moving along the Moscow-Vladivostok railroad (about 9300 km), the car-laboratory crosses the background regions for about 45–65% of the entire travel time. For these regions, spatial distributions and daily cycles of different pollutants are studied. The corresponding plots characterize the mean state of the atmosphere over the continent for the latitude belt 50°–57° N. An example of the longitudinal distribution of the CO, CO_2, and CH_4 background surface concentrations for the summer and winter seasons are shown in Figure 2. In cold periods (left panels of Fig. 2) the distribution of pollutants is almost uniform. A small increase in the CO and CH_4 concentrations seen in the central regions of Siberia is attributable to the Kuznetsk industrial enterprises producing great amounts of coal and burning it in electric power stations. In the summer season (right panels of Fig. 2), a positive eastward gradient in the CO and CO_2 concentrations is observed. Isotope analysis and nocturnal respiration flux estimates made on the basis of the ^{222}Rn concentration measurements provide an explanation of this gradient, via enhanced biogenic emissions and emissions from forest fires in the boreal zone of eastern Siberia. The enhanced CH_4 concentrations observed in western Siberia are mainly caused by biogenic methane emissions from the swamps and moistened soils [Belikov et al., 2006; TROICA, 2006].

Along those western-Siberian railroad sections where the car-laboratory crosses plumes propagating from gas-producing enterprises, the peak CH_4 concentrations are revealed against the background of the general positive trend of its concentration. Figure 3 illustrates an example of crossing one such plume, which shifts eastward as the train moves. The plume structure reflects the combined effects of biogenic and thermogenic (anthropogenic) methane emissions. The peak methane concentrations are also seen at some points where gas-main pipelines are crossed [TROICA, 2006].

Repeated passes through a great number of cities by the Trans-Siberian Railroad allows study of the principal features of the pollutant distributions over these urban areas as a function of their sizes,

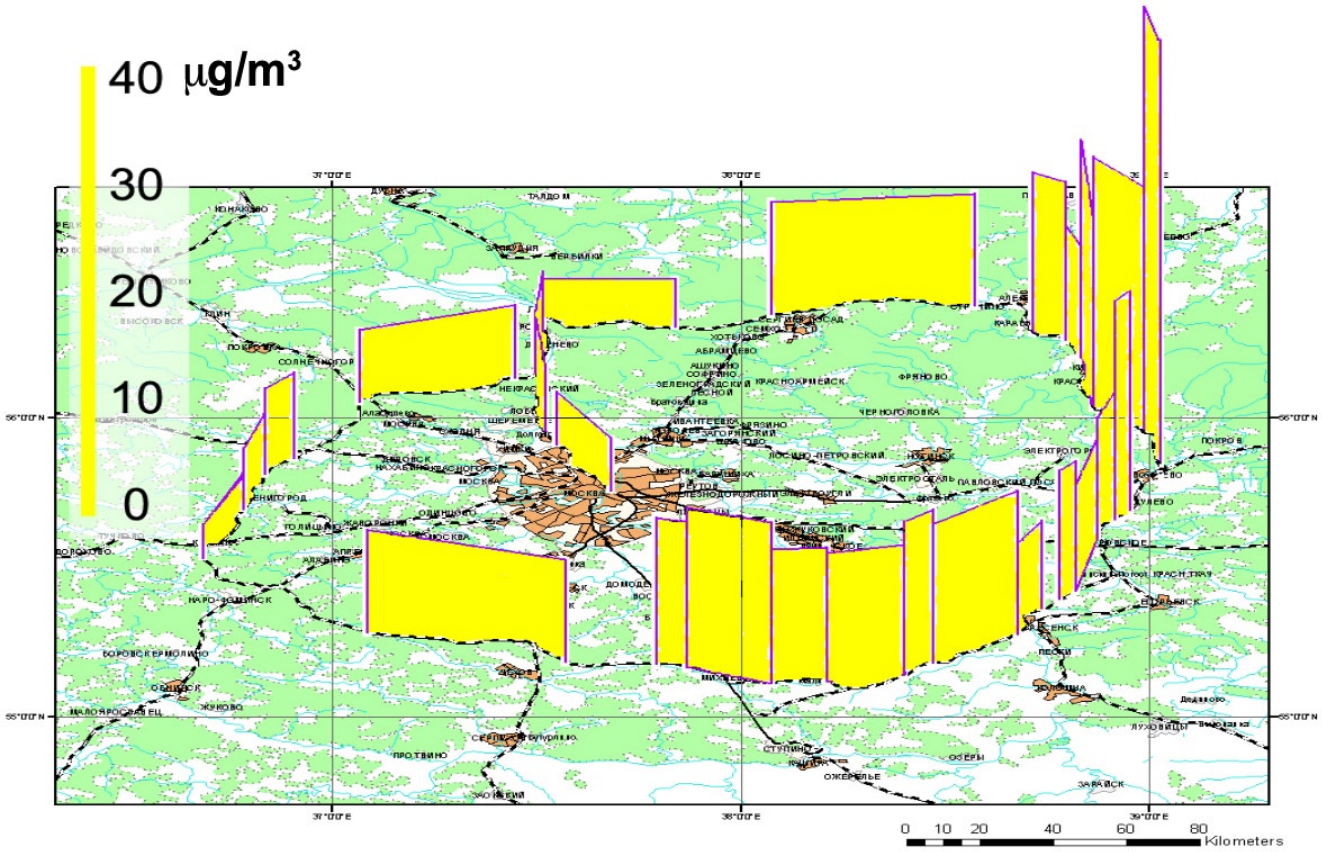

Figure 5. Spatial distribution of the integral surface concentrations for the sum of benzene, toluene, etylbenzene, m,p-xylene, o-xylene.

degree of saturation by industrial enterprises, season, time of day, and meteorological conditions. An example of the variability in the O_3 and NO_2 concentrations near and in cities of three sizes as a function of season and time of day is shown in Figure 4. The concentrations shown are averages from measurements made in the course of the TROICA-4 and -8 (winter) and TROICA-5 and -7 (summer) expeditions over the following territories: city vicinities, city, and city centre (where the railroad station is, as a rule, located). For comparison, the background O_3 and NO_2 concentrations are shown in the figure. In the mid-size and small cities, the O_3 concentration in summer exceeds its background level. In megapolises, the O_3 concentration is very low, because of its decomposition. This peculiarity is caused by climatic conditions (low illumination and air temperature), high level of NO_x emissions, and good ventilation of most of the cities. Detailed information on how the megalopolis of Moscow influences the spatial distribution of gas and aerosol constituents was obtained during the TROICA-10 experiment. Figure 5 shows an example of the integral for three circles around the city, measuring concentrations of the main four component aromatic hydrocarbons.

The sampling frequency of the TROICA measurements allows for the study the influence of individual objects on the atmospheric composition. Figure 6 demonstrates the mean variations of the surface ozone concentration during the crossing of high-voltage transmission lines. Ozone is generated under the influence of corona discharges,

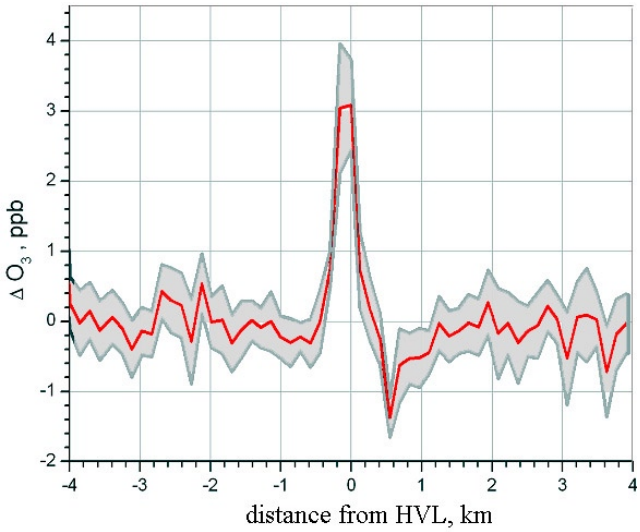

Figure 6. Ozone concentration changes under high voltage (500 kV) lines, located in unpolluted areas.

which form O, NO, and OH radicals. In the course of the TROICA-2 to TROICA-9 expeditions, more than 1000 events of crossing of high-voltage transmission lines were tracked. Of all these events, only those which occurred in unpolluted areas at points with coordinates determined to be within several meters of the 220–500-kV high-voltage transmission lines were taken into consideration. The measured increase in the O_3 concentration (3.1±0.6

ppb for 500-kV high-voltage transmission lines), though small, is always present and may influence local ecosystems. New results have corroborated previous ones published by N.F. Elansky et al. (2001). In polluted air, high-voltage transmission lines, as a source of radicals, heighten the oxidative activity of the atmosphere and can affect its cleansing capacity.

The principal conclusion is that a mobile laboratory can be a highly efficient means of studying the sources, transport, chemical transformation, and sinks of gaseous and aerosol atmospheric constituents. The potential utility of such a laboratory is not limited to observations of the surface-air composition. Tropospheric and stratospheric vertical profiles of temperature, gas species (O_3 and NO_2), and aerosols can be reliably retrieved from such a platform via remote sensing. Encouraging results were also obtained in the first attempts to use the observational data on scattered solar radiation in the UV and visible spectral regions for estimation of the NO_2 and SO_2 emissions by industrial sources located 6-8 km away [TROICA, 2006].

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Report on the joint NIES, NIWA, NOAA-ESRL sampling project aboard ships of the Toyofuji shipping company between Nelson, New Zealand and Osaka, Japan

A Brief Summary

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For many years "ships of opportunity" have been used to supplement regular measurements of atmospheric trace gases near the surface made at fixed land based sites and aboard oceanographic vessels. The ships that have been used are typically cargo and container ships on major ocean trade routes.

The data obtained from these programmes have been extraordinarily valuable, providing vital data from the mid-Pacific and other Oceanic regions. For example NOAA-ESRL (National Oceanic and Atmospheric Administration–Earth System Research Laboratory) in Boulder, Colorado and NIWA (National Institute of Oceanic and Atmospheric Research), Wellington, New Zealand have used ships originally from the Blue Star lines shipping company (more recently P&O NedLoyd) to obtain atmospheric trace gas mixing ratio and isotopic data on routes between Auckland, New Zealand and the West Coast of the USA. Many of these data sets now extend back almost 20 years.

More recently (2002) NIWA began a joint project with NIES (National Institute of Environmental Studies) based in Tsukuba, Japan. This project uses large Japanese car carriers run by the Toyofuji shipping company of Nagoya, Japan to collect very large clean air samples for the measurement of ^{14}C and ^{13}C in atmospheric CO and methane respectively. The ships cross both the South Pacific Convergence (SPCZ) and Inter-tropical Convergence (ITCZ) zones on a track between Nelson, New Zealand (41°S) and Osaka, Japan (35°N) in the Western Pacific. In this part of the Pacific the behaviour of the convergence zones is quite different from that observed at points further east in the mid Pacific. In the mid Pacific the zones are usually well defined and separated by typically 5-10 degrees of latitude. In the Western Pacific the situation is much more complex with the zones often forming a wedge and the disappearance of the SPCZ altogether. This has a profound effect on the transport of trace gases between the hemispheres in this region and is one of the major study points of the joint project. Because of the importance of this region NOAA-ESRL are supplementing their mid Pacific data with data obtained from 2L air samples collected aboard the Toyofuji shipping company ships. They measure a suite of compounds including CO_2, CH_4, CO, H_2, N_2O, and SF_6. Through a collaboration with the University of Colorado, Institute for Arctic and Alpine Research, stable isotopes of CO_2 (C and O) and CH_4 (C and H) are also analyzed in the samples.

In 2005 the Toyofuji shipping company introduced a new ship, the Transfuture 5, for the Nelson to Osaka voyages replacing their original vessel, the Fujitransworld. The new ship has a purpose built laboratory aboard containing gas chromatographs and a non dispersive infra-red analyser for trace gas mixing ratio analyses. This equipment is operated by NIES under the directorship of Professor Yukihiro Nojiri. On this ship NIWA has a second container laboratory which is mounted on the upper decks and houses the NOAA-ESRL sampling equipment as well as NIWA compressors and sampling equipment for large volume samples for isotopic analyses.

Dave Lowe of NIWA works with flask samples from one of the cargo "ships of opportunity" used to monitor atmospheric trace gases in the Pacific.

Data obtained from the voyages so far show the profound impact of the convergence zones on the inter-hemispheric gradients of the trace species measured. However, due to the continual movement and formation of the zones and other complex tropical meteorology, data obtained from each voyage has to be interpreted on a case by case basis.

Recent NIWA data, for example from December 2005, show the distinct "zoning" caused by an active SPCZ

and ITCZ. In addition, measurements of the stable isotopes in methane have demonstrated the convective vertical motion around the ITCZ "pulling down" stratospheric methane into the marine boundary layer. Other data from NOAA-ESRL show the impacts of Asian outflow north of the convergence zones.

This valuable project would not have been possible without the vision of Yukihiro Nojiri who initiated the collaborative project with Dave Lowe at NIWA in New Zealand, joined by Ed Dlugokencky from NOAA-ESRL. "Ships of opportunity" sampling programmes are notorious for sudden changes in shipping schedules which make planning of long term monitoring projects very difficult. This programme, based on the car carrying ships of the Toyofuji shipping company between New Zealand and Japan, has been made possible by their generosity, commitment to environmental research and a close relationship with the scientific organisations making the measurements.

2007 IGAC Scientific Steering Committee Meeting

The 2007 IGAC Scientific Steering Committee held its annual meeting at the Sleeping Lady resort in Leavenworth, Washington, September 3-6. In attendance were:

(L-R, front row) Graciela Raga, Phil Rasch (IGAC Co-chair), Kathy Law (IGAC Co-chair), Celine Mari, Laura Gallardo-Klenner, Stuart Piketh, Eric Wolff,

(L-R, back row) David Griffith, Sarah Doherty (Exec. Officer), David Parrish, Laurens Ganzeveld (iLEAPS liaison), Kevin Noone (IGBP Director), John Burrows, Randall Martin, Jen-Ping Chen, Tong Zhu, Maria Kanakidou, and Yutaka Kondo.

For more on the IGAC SSC, visit http://www.igac.noaa.gov/structure.php

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Bridging the Scale in Atmospheric Chemistry: Local to Global

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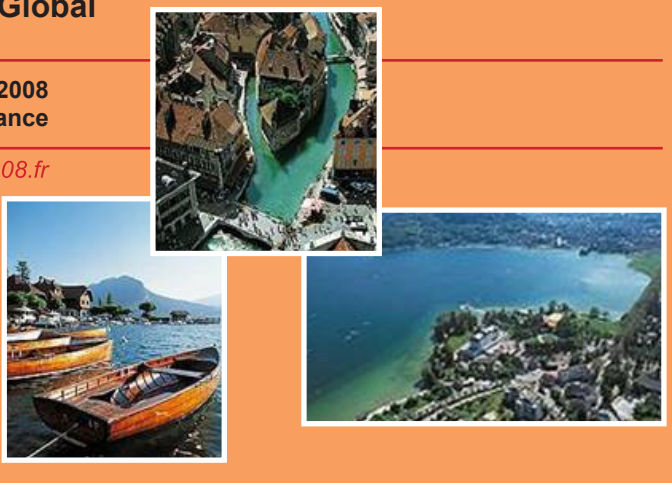
http://www.igacfrance2008.fr



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