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

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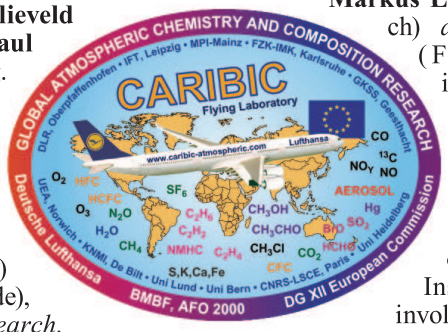

Introduction

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

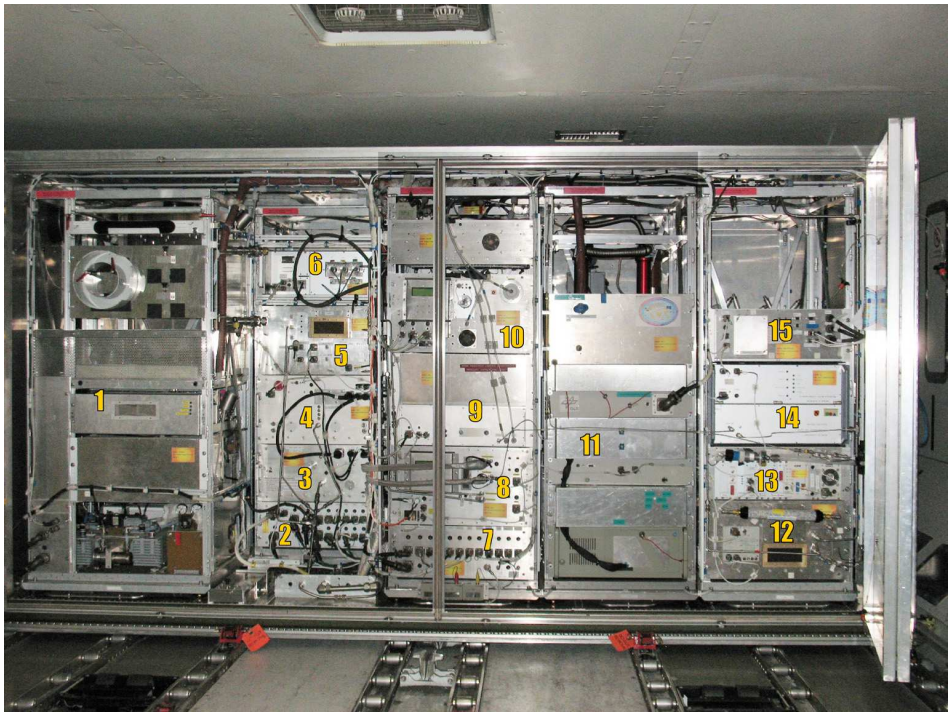

Figure 1. Frontal view of the container inside the forward cargo bay of the A340-600, facing the flight direction. Its width is 3 meters and its mass is 1.5 ton. In-flight its doors are obviously closed. 1. PTRMS. 2. Main power unit. 3 Aerosol, CPC and sampler. 4. Aerosol, 2 CPCs. 5. Aerosol OPC. 6. 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

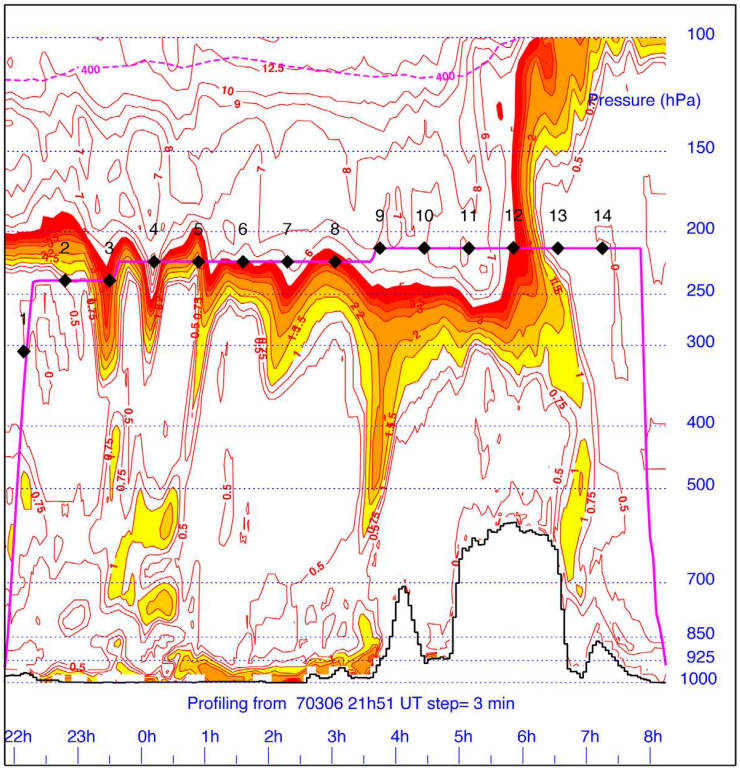

Figure 3. Vertical cross-section of the potential vorticity (PV) field along the aircraft's track for a flight from Frankfurt to Guangzhou (Cross section of 209 profiles, 7/3/2007, 0 UTC, from model level data; KNMI/ECMWF). The tropopause is highly structured, and in this particular case lowermost stratospheric 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.

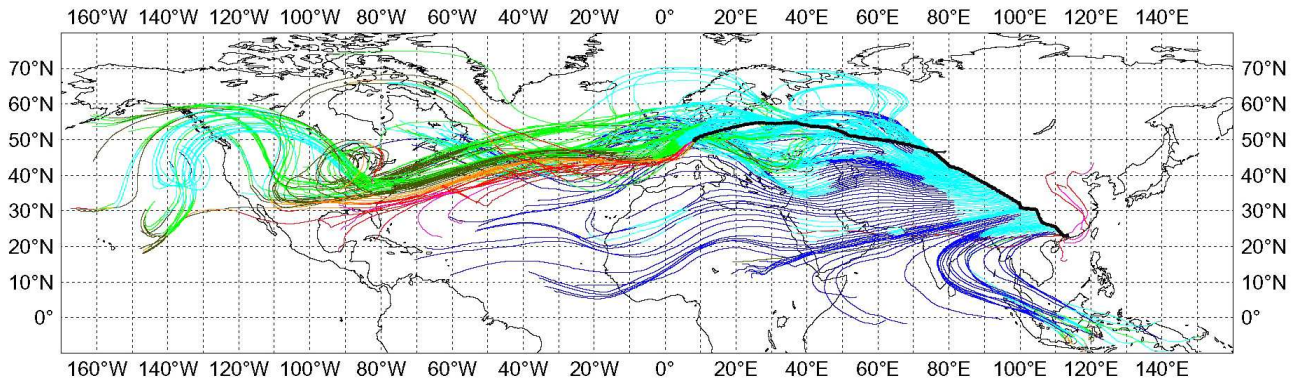

Figure 4. Five day back-trajectories for points along a single flight path (for more detail, like individual trajectories, we refer to the KNMI website for CARIBIC). Note the change in "origin" of the air upon approaching Guangzhou.

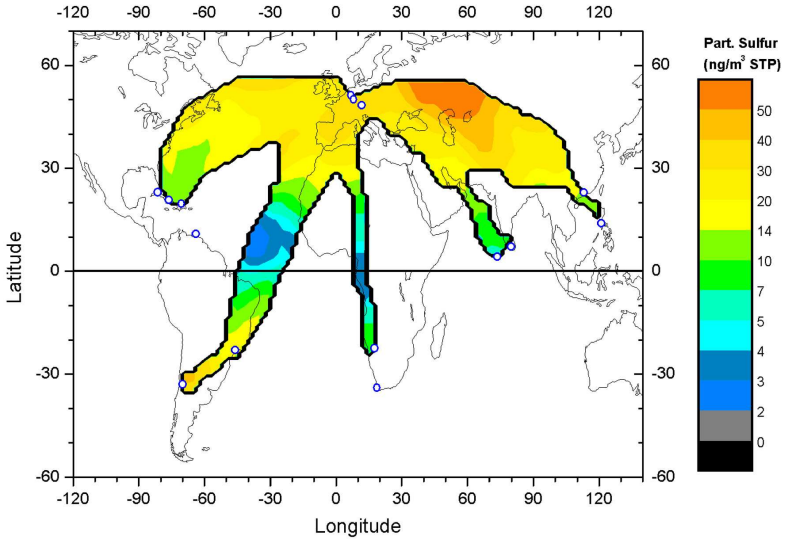

Figure 5. The concentration of sulfur (as present in aerosol particles) based on PIXE analyses carried out in Lund. Used are all data from the previous and present CARIBIC flights. At mid to higher latitudes this climatology is biased by the fraction of stratospheric air (more sulfate). For this a correction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

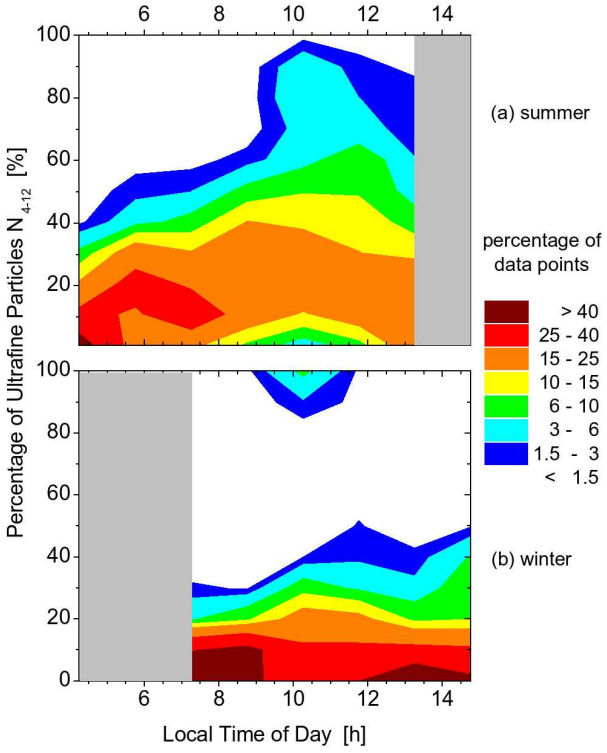

Figure 6. Probability distribution for the percentage of ultrafine particles (4 nm < diameter < 12 nm, $N_{4\text{-}12}$) at 8-12 km altitude, versus local time of day for tropical latitudes between 5° and 20°N over the Arabian Sea in (a) summer and (b) winter. Gray shading marks lack of data. For more 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

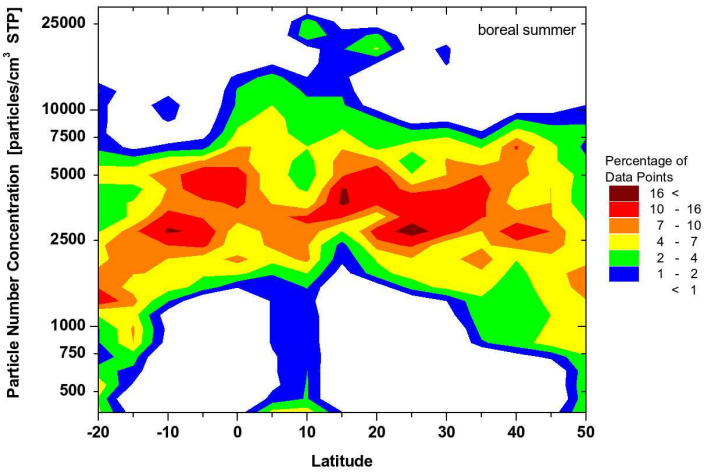

Figure 7. Probability distribution of Aitken mode particles (diameter >12 nm) in the UT in boreal summer along the South America route. Colors indicate the percentage of all particle measurements in a latitude band that fall into a particular concentration bin. Mean location of the ITCZ over the Atlantic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

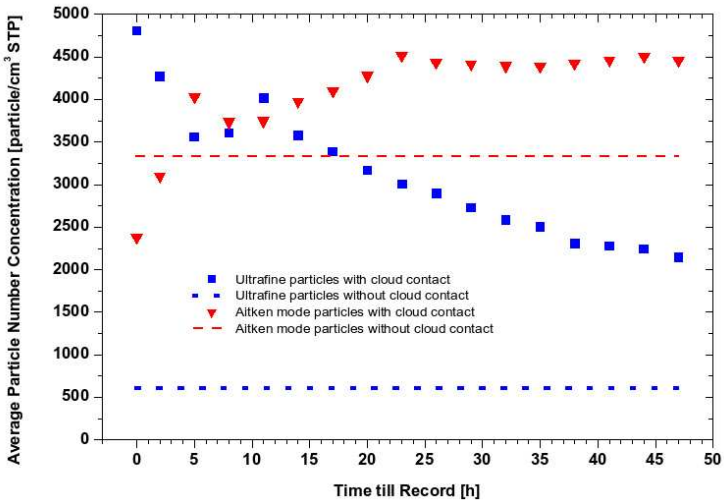

Figure 8. The dependency of particle number concentrations (over the Arabian Sea) on the time elapsed between the last cloud contact and the actual measurement by the CARIBIC aircraft. Each point includes all values for which the last cloud contact fell into the time period indicated at the x-axis. Blue 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.

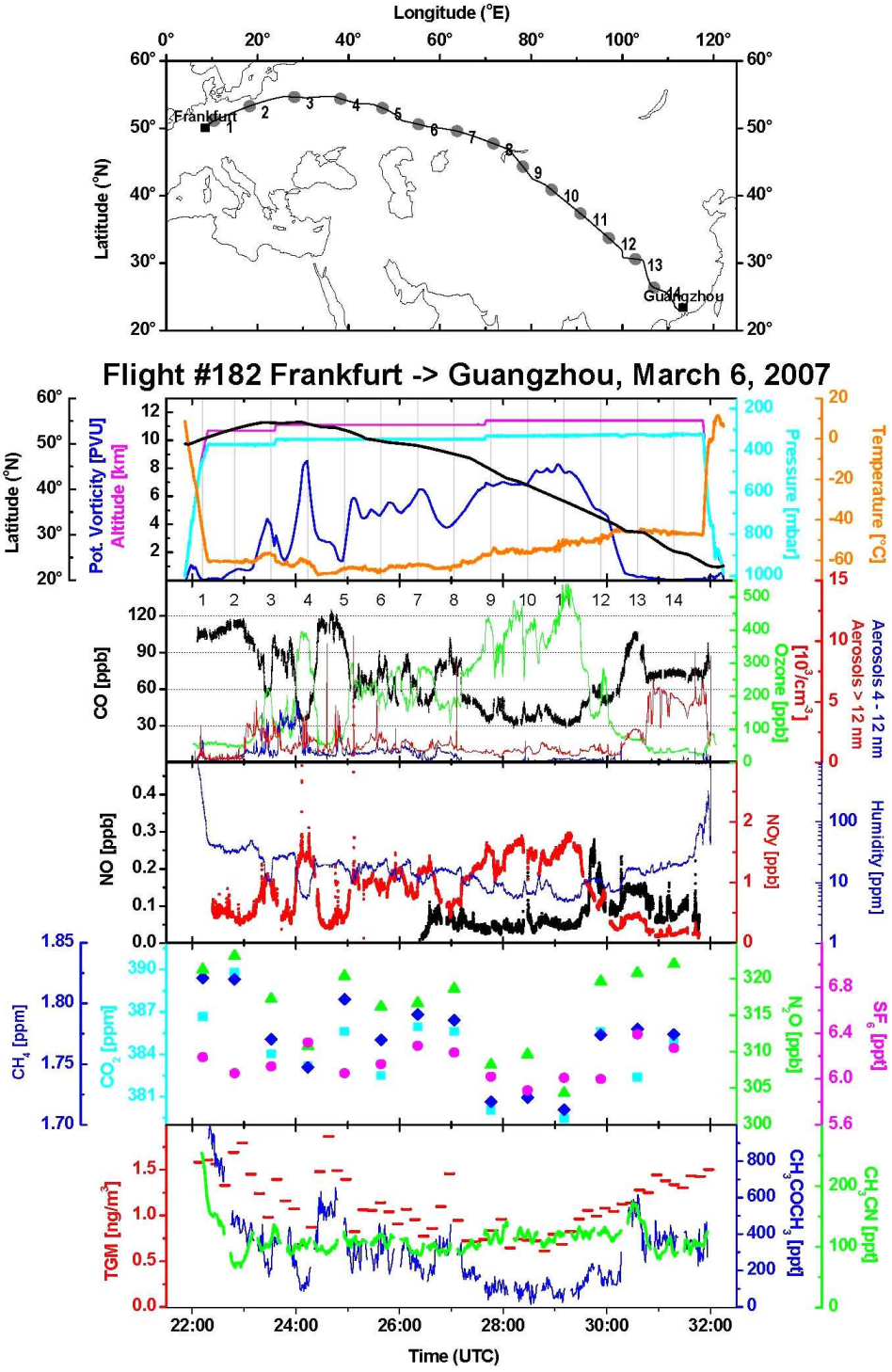

Figure 9. An overview of measurements obtained for a flight to Guangzhou (Meteo data in Figures 3 and 4). The grey dots on the map and the numbers on the x-axis of the top plot indicate locations of discrete air samples. Irregularities in the flight track are mostly due to the pilots avoiding lightning 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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