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Civil aircraft for the regular investigation of the atmosphere based on an instrumented container: the new CARIBIC system

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Abstract

A large airfreight container with automated instruments for measurement of atmospheric gases and trace compounds was operated on a monthly basis onboard a Boeing 767-300 ER of LTU International Airways during long-distance flights from 1997 to 2002 (CARIBIC, Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container, http://www.caribic-atmospheric.com). Subsequently a more advanced system has been developed, using a larger capacity container with additional equipment and an improved inlet system. CARIBIC phase #2 was implemented on a new long-range aircraft type Airbus A340-600 of the Lufthansa German Airlines (Star Alliance) in December 2004, creating a powerful flying observatory. The instrument package comprises detectors for the measurement of O_3, total and gaseous H_2O, NO and NO_y, CO, CO_2, O_2, Hg, and number concentrations of sub-micrometer particles (>4 nm, >12 nm, and >18 nm diameter). Furthermore, an optical particle counter and a proton transfer mass spectrometer (PTR-MS) are installed. Aerosol samples are collected for analyses of elemental composition and particle morphology after flight. Air samples are taken in glass containers for laboratory analyses of hydrocarbons, halocarbons and greenhouse gases in several laboratories. Absorption tubes collect oxygenated volatile organic compounds. Three differential optical absorption spectrometers (DOAS) with their telescopes mounted in the inlet system measure atmospheric trace gases such as BrO, HONO, and NO_2. A video camera mounted in the inlet provides information about clouds along the flight track. Here we describe the flying observatory and report examples of measurement results.

1 Introduction

In view of the ever more detailed questions about the myriad of coupled chemical and physical processes in the global atmosphere, while witnessing human induced disturbances of this system that affect climate within a human's lifetime, the need for ad-

vancing observational capacities is evident. Besides intensive atmospheric chemistry research largely based on national and international measurement campaigns using ship, aircraft, satellites and ground stations, there is a need for systematic approaches in observing the chemistry and composition of the atmosphere with sufficient detail. To some degree the making of "observations" is moving from the realm of science to that of technology and engineering. Our understanding of atmospheric processes, be it chemical or physical, has grown enormously, and further growth is based on improved models and an adequate supply of sufficiently detailed, relevant data. Next to the understanding of processes, there is a clear need for monitoring to comprehend the entire system. Besides this, for systems as complex and variable as the earth's atmosphere (not to mention its coupling with the other complex components of the planetary system, like the biosphere) the need for monitoring in terms of meteorology and chemistry, and detecting changes is evident (http://www.ipcc.ch).

One has indeed seen an increasing number of surface based monitoring sites (e.g., ESRL-GMD, http://www.cmdl.noaa.gov/, GAW, http://www.wmo.ch (Montzka et al., 2003; WMO, 2004)), and the detailed information we have about H_2, CH_4, halocarbons and CO_2 is by virtue of this excellent work of monitoring. At several of such observatories the number of trace gases measured has been increased by installing sophisticated systems for measuring a host of trace gases in real time (the ALE/GAGE/AGAGE network http://cdiac.ornl.gov/ndps/alegage.html (Prinn et al., 2000)). Thus, next to the basic, yet powerful process of collecting air samples on a weekly to monthly basis, highly detailed high frequency information becomes available. This all enables a better understanding of trace gas sources, transport, chemistry, and mixing. Generally, observatories are on mountains or in coastal regions for the purpose of obtaining data representing air masses that are not affected by immediate input from nearby sources of air pollution. Next to these stationary and dedicated sites, platforms of opportunity such as ships are used by which the marine boundary layer can be probed (Lelieveld et al., 2004). Even the Trans-Siberian railroad with an instrumented observation wagon transported over the 9000 km long track from Moscow to Vladivostok and back is used

(http://www.troica-environmental.com; Crutzen et al., 1998; Hurst et al., 2004).

Remote optical sensing from the surface is a powerful technique that can deliver vertical profiles or column integrated data for aerosols and certain trace gases continuously (e.g., NDSC, EARLINET, http://lidarb.dkrz.de/earlinet/index.html). Remote sensing from satellites for tropospheric composition has been increasing strongly and its near global coverage is its major advantage.

Although the need for monitoring can be made clear with relative ease, the idea has really never thrilled science funding agencies since long-term commitments are involved. This forces us to be creative in developing affordable technologies. Atmospheric monitoring using civil aircraft is inherently a logical concept and in the mean time constitutes a valuable component in the colorful spectrum of global observation systems (see also IGACO, Integrated Global Chemistry Observations, http://www.igospartners.org/). The civil aircraft based projects offer the advantage of regular, long-distance, and long-term coverage. In particular the 10 to 12 km cruising altitude range which interestingly coincides with the upper troposphere/lower stratosphere region (UT/LS) at mid- to high latitudes is of high relevance. The UT/LS plays an important role in terms of radiative forcing by infrared-active species, such as water vapor, ozone, cirrus cloud particles which are strong absorbers of outgoing long-wave radiation. At the same time, the complex UT/LS is still one of the less well documented and understood regions of the atmosphere. Main reasons for this gap in our knowledge are the extreme dynamical and chemical complexity of the UT/LS and of course the difficulty to regularly monitor the UT/LS at affordable cost. Although long-range aircraft spent most time cruising, vertical profiles of trace gases near airports are also obtained.

Of course, the use of civil aircraft also involves some restrictions. As elucidated, the cruising altitude is limited, giving UT/LS information at mid-latitudes, and free troposphere information in the tropics. Furthermore, having a single home airport, such as Frankfurt as a basis, only a limited, albeit large region of the globe can be covered. Figure 1 shows the CARIBIC flight patterns which obviously are somewhat Eurocen-

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tric. A more global coverage can be obtained when combining such projects stationed in different continents. Since there is some spread in actual trajectories, the coverage is not limited to narrow corridors. Local peaks of aerosols and trace gases indicate that exhaust plumes of other aircraft are occasionally encountered, but most air inter-
5 cepted is representative for the background air. The effects of other aircraft can be easily identified.

Using civil aircraft as a measurement platform is not a new idea. W. Bischof in his 1970 Tellus paper already refers to the collection of air samples in flight for CO_2 measurements in 1962, and the practice of using commercial airliners gained some
10 momentum in the late sixties and seventies when CO and O_3 distributions were determined using instruments strapped onto passenger seats analyzing air from the cabin ventilation system of commercial airliners (Seiler and Junge, 1969, 1970; Tiefenau et al., 1973; Fabian and Pruchniewicz 1977). Note that in those days ozone was not removed from the cabin air supply. In the seventies, a dedicated inlet and measure-
15 ment system for CO, condensation nuclei, and O_3 were permanently installed on five Boeing 737 aircraft within the GASP (Global Atmospheric Sampling Project) project of NASA (Falconer and Holdeman, 1976; Nastrom, 1979; Pratt and Falconer, 1979). After that, commercial airliners were not used, with the exception of an instrumented airfreight container operated by two persons onboard a Boeing 707 of Lufthansa Cargo
20 Services (Dickerson, 1984; Connors et al., 1991).

Several projects involving civil aircraft, but using different approaches, were initiated in the 1990s. The MOZAIC project (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) utilizes instruments permanently installed in nominally five Airbus A340-300 aircraft to provide measurements of O_3 and H_2O (since
25 1991) and of CO and NO_y (since 2001) (e.g., Marenco et al., 1998). MOZAIC continues, but eventually will be superseded by a new approach using smaller instrument packages being developed and certified for Airbus A340 aircraft (IAGOS project, http://www.fz-juelich.de/icg/icg-ii/iagos). In the NOXAR project (Measurements of Nitrogen Oxides and Ozone along Air Routes) a Boeing 747 of Swissair was equipped

with an NO, NO_2, and O_3 analyzer (Brunner et al., 2001). The JAL project (Japan Airlines) was initially based on the collection of air samples in flasks using the air compressed by one of the jet engines. The samples were analyzed for greenhouse gases and in several instances even for their isotopic composition (Matsueda and In-
5 oue, 1996). The JAL project is being extended, incorporating five aircraft for continuous CO_2 monitoring, whereas two of these have air sample collection systems (deployed monthly). Finally, the CARIBIC project started regular flights in November 1997 (Bren-ninkmeijer et al., 1999, http://www.caribic-atmospheric.com). CARIBIC relies on an au-tomatically operating airfreight container equipped with in-situ measuring instruments
10 and sampling devices for more than 60 different trace gases and aerosol properties. The strategy of CARIBIC is to optimize the number of trace compounds that are de-tected and collected on the basis of regular flights. Thus once a month the container is installed in the aircraft and de-installed after presently 4 sequential flights.

One may ask what advantages a container system offers and how CARIBIC can fill
15 gaps in the current observational capacity. Foremost is that the container concept al-lows a wide range of equipment to be deployed. The extensive array of in situ analyzers combined with air and aerosol samplers renders CARIBIC a "flying observatory". The comprehensiveness of the payload enables studies of chemical and dynamical interac-tions between trace species, and the combined information facilitates the analyses of
20 source and sink processes. The time elapsed between flights is used to analyze the air and aerosol samples while the container equipment is maintained and calibrated. The turnaround time achieved has been one month, whereas the fastest turnaround time can be one week. Frequent, highly consistent, very detailed datasets become available for several regions of the globe, which are ideally suitable for process and climatolog-
25 ical studies. An example is the reconstruction of the annual cycles of trace gases in the tropical troposphere, the mid-latitude upper troposphere and the mid-latitude low-ermost stratosphere. Moreover, CARIBIC can help verify retrievals from satellite based remote sensing instruments (Peylin et al., 2007).

In this paper we present the new CARIBIC system comprising the inlet system, the

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modifications of the aircraft, and in particular the container and instrumentation. In Sect. 2 we briefly summarize the previous CARIBIC system. General considerations pertaining to the combining of the container concept with the given aircraft type and its operation, as well as the inlet system, the container and modifications to the aircraft are discussed in the following section. In Sect. 4 the current instrumentation is described in detail giving examples of measurement results for equipment new to this system.

2   CARIBIC development

The first CARIBIC container was deployed on a Boeing 767-300 ER of LTU International Airways that had been retrofitted with an aerosol and trace gas inlet system (Hermann et al., 2001). The aircraft modifications and the instrumentation were described by Brenninkmeijer et al. (1999). Until the decommissioning of the aircraft in the course of the LTU fleet renewal in May 2002, 75 flights had been carried out mostly on the routes from Germany to the Indian Ocean and to the Caribbean. Because of the limited range of the aircraft only a limited number of flights into the Southern Hemisphere (in this case South Africa) could be made. The data set has so far provided a basis for studies of aerosol particle elemental composition, formation and climatology of aerosols in the tropopause region (Heintzenberg et al., 2002, 2003; Hermann et al., 2003; Martinsson et al., 2001; Papaspiropoulos et al., 2002), the interpretation of tropospheric O_3 and CO correlations, as well as their correlation in respect to transport across the tropopause and chemical processing (Zahn et al., 2000, 2002a, 2004a and b; Zahn and Brenninkmeijer, 2003), and for process studies (Mühle et al., 2002; Zahn et al., 2002b). The data have also contributed to studies of trace gas cycles in the atmosphere (Krol et al., 2003; Sturges et al., 2000) and enabled studies of the isotopic composition of CO including ^{14}CO (Joeckel, 2000) as a global tracer for OH, and of H_2 and its D/H ratio for constraining budget calculations for the atmospheric H_2 budget (Rhee et al., 2006). Work on the application of CO_2 concentrations and concomitant highly accurate ^{13}C and ^{18}O isotope data is in progress. Furthermore, for 360

air samples extracts of condensable gases (CO_2, N_2O, hydrocarbons, etc.) have been archived for D/H, $^{13}C/^{12}C$ and $^{37}Cl/^{35}Cl$ measurements of hydrocarbons and CH_3Cl (Pupek et al., 2005). All in all a considerable amount of systematic and hitherto unavailable data have been obtained. Data from the CARIBIC-Boeing 767 flights are accessible on request (consult http://www.caribic-atmospheric.com).

3 The new CARIBIC system

3.1 General considerations

The aircraft of choice was a new Airbus A340-600 (certified in 2004), which is a long range (14 000 km) stretched member (up to 380 passengers) of the A340 family (www.airbus.com). Lufthansa acquired 10 of these aircraft over the period May 2003–October 2004, and this fleet is expanding. The integration of the CARIBIC system in the aircraft required several modifications. Most important were the structural changes to the hull around the mounting position of the inlet system, followed by the installation of the air tubing and cables. Also needed were a container connector interface, a control panel in the cockpit, and modifications to electrical hardware and electronics and software. At all stages tests were to be conducted concluded with a test flight. The estimated two weeks of work on the aircraft needed would have inflicted an undesirable downtime and it was decided to use the grounding period of the aircraft used for retrofitting it with a broadband internet system (Boeing Conexxion) by Lufthansa Technik (FLYNET). This system comprises a wireless LAN system with a satellite tracking antenna system mounted inside a special dome on the roof of the aircraft. The joint modifications took place November 2004.

The A340-600 differs from the previously used Boeing 767 in a for CARIBIC important aspect, namely the cargo loading door of the forward cargo bay is located towards the front of the aircraft, and not near the wings (Fig. 2). This means that the measurement container's position is towards the wing box, in the rear of the forward cargo bay

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above the leading edge of the belly fairing. The reason for this is that the CARIBIC container must be loaded first into the aircraft followed by the standard contingent of cargo containers and pallets. At the airport of destination the regular cargo can be unloaded, leaving the CARIBIC container in position. Upon return at the airport of origin, first the cargo is unloaded, followed by the careful de-installation and removal of the CARIBIC container. Thus, the aircraft structure implies that the container position is in the aft of the forward cargo bay.

Ideally, the air/aerosol inlet system should be positioned directly beneath the container. Short tubing minimizes contamination/loss for gases and aerosol particles. However, below the actual container position the belly fairing is present. Its shell is too thin to support an inlet system. Therefore, the actual position of the inlet system was shifted forward by about 2 m. As a result of these aircraft structural considerations, the container is about 27 m away from the nose of the aircraft and the inlet system about 25 m. From a mechanical/structural point of view the height of the inlet system should be minimal. From an analytical point of view one should sample air totally outside the aircraft's boundary layer. Data from Airbus Hamburg about the exact airflow field and pressure distribution around the aircraft were used to find a good compromise. According to their data, the boundary layer thickness at the position of the inlet is ~25 cm. The CARIBIC inlet system is, despite its more complex geometry, not an over-dimensioned probe, being slightly larger than the drain masts and VHF antennas.

The scientific and analytical requirements have to be met within the constraints of the aircraft's structure, safety and operation, and hence compromises concerning the analytical capabilities have to be made. The safety requirements of civil aircraft are extensive and strict. Compounds classified as dangerous (e.g. liquid nitrogen) cannot be used in a passenger aircraft. Concerning the container and the instruments there are mechanical requirements (loads on equipment, racks and container as a whole during acceleration/crash conditions), electrical requirements (bonding, fuses etc), and requirements concerning combustible materials. Furthermore, specifications with respect to the control of equipment (computer controlled switching on and off of the many

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analyzers) and concerning the absence of conducted or radiated electromagnetic energy have to be met. For the inlet system there are strict regulations concerning its impact on the aerodynamics, on the integrity of the aircraft hull, absence of vibrations, its behaviour at icing conditions, stability, etc. The responsibility for the design and construction of all modifications to the aircraft, the integration of the container, and for the certification process with the German and European Aviation Authorities were entirely with Lufthansa Technik (http://www.LHT.de).

The technical process of realizing CARIBIC over a period of about two years can be briefly summarized. After the scientific/analytical design considerations were defined concerning container, inlet, and tubing systems, the technical design was started by LHT in cooperation with Airbus, third companies, and scientists. The construction of the container, racks, and all equipment was an ongoing effort over two years. The new inlet system was designed, and its possible impact on the structure (including stress analyses) of the aircraft was calculated extensively. Thereafter the inlet parts were manufactured, and the inlet assembled in three weeks at Lufthansa Technik in Hamburg. After all other certified materials had been acquired, the construction/installation of the system was executed within two weeks of aircraft modification at Frankfurt airport. This period was completed by a test flight during which an extensive protocol of tests, including vibration measurements, was absolved.

The intention is that the new CARIBIC system will be operational over at least ten years. Airline companies operate in a most competitive commercial environment, and scientific projects (against a background of global change) can in principle be tolerated if these do not disturb routine operation and do not cause significant additional cost. Years of positive experience with CARIBIC involving 2 airlines, 2 aircraft types, and 2 containers especially concerning their installation, flying and de-installation give great confidence in the sustainability in this respect. The environment division of Lufthansa supports CARIBIC as one of several activities to better understand the atmosphere and to reduce pollution and noise. The scientific consortium of CARIBIC pays Lufthansa Cargo standard rates for the airfreight cost of the measurement container.

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3.2 The CARIBIC inlet system

Details of the inlet system, in particular the aerosol sampling characteristics, will be published elsewhere. We summarize the main aspects. The inlet has been designed around the analytical requirements a) to optimize aerosol particle sampling at cruising speeds of $\sim$250 m/s, b) to provide dedicated air intakes for different trace gases with heating systems for inlet tips and tubing, c) to provide ram pressure thus obviating the use of pumps for some instruments, and d) to host a set of three miniature DOAS telescopes for remote sensing and a video camera for observing clouds. The inlet structure is that of a hollow spar machined out of a block of aluminum, to which the three flanges of the three separate probes are fixed. A fore- and aft-shell form the enclosure and contribute to the rigidity of the assembly, using almost 100 fasteners. Figure 3 shows the inlet system as designed for LHT by Garner CAD Technics GmbH (Oberpfaffenhofen, Germany) in close cooperation with the scientists and manufactured by Heggeman Aerospace (Büren, Germany). It consists of 25 structural aluminum elements and is 55 cm long, 34 cm high, 18 cm wide. Its mass is about 3.5 kg. All essential parts exposed into the flight direction are nickel plated to improve hardness. The total number of components is about 60, comprising the aluminum parts, tubing, heater elements, temperature sensors, insulation, plugs, DOAS telescope, brackets, and the video camera.

Using detailed information about the airflow along the fuselage as supplied by Airbus in Hamburg, a computational fluid dynamics (CFD) code (FLUENT) was used to model the air flow distribution around the inlet system. An example of the computation of the total pressure and velocity distribution around the inlet system is shown in Fig. 4. The design is mainly such to prevent supersonic speeds and turbulence.

One of the main scientific design considerations was to obtain representative aerosol sampling characteristics at the cruising speed of nominally 250 m s^{-1}. This means that the aerosol tube (diffuser tube) should sufficiently slow down the air flow from which aerosol particles are to be sampled. It also means that a shroud is applied.

This shroud is a 12 cm long, 4 cm OD aerodynamic cylinder upstream of the aerosol diffuser tube itself and supported by four prongs. Most air entering the shroud passes through the 4 circular slits between the prongs. Because the shroud is heated, one of these $7\times7\,mm^2$ square-cross-section prongs is hollow to supply a channel for feeding through temperature sensor and heater wires. The aerodynamic function of the shroud is to make the aerosol sampling characteristics as little dependent on flight conditions (angle of attack) as possible (cf. e.g., Murphy and Schein, 1998). A fraction of the air flowing through the shroud enters the inlet tip of the diffuser tube with its 6.4 mm inlet orifice which is located centrally 8 cm downstream inside the shroud. The diffuser itself is a four degree half-angle diffuser while the outlet orifice is 4 mm. The actual sample air for the particles is taken downstream inside the diffuser via a centrally positioned, unheated forward pointing electro-polished 3/8″ stainless steel pick-up tube bent with a radius of 100 mm.

The water probe is mounted on the right (looking in flight direction), 1 cm above the aerosol probe. A forward facing (heated) inlet tip of 4 mm inner diameter collects air for total water measurement (gas phase plus droplets/crystals) using ram pressure to force the air through the H_2O analyzers. Calculations by Airbus Hamburg indicate that the air displacement by the aircraft itself causes an enhancement of cloud particles at the position of the total water inlet of 12–19%, depending on the angle of attack. Based on the theory by Liu et al. (1989), the non-isokinetic inlet configuration causes a further cloud particle enrichment of the total water inlet of 210–380%, depending on the actual inlet sample flow of 30–50 vol-l/min. Both cloud particle enrichments are considered for calculating the amount cloud water/ice. A second, also heated sideward facing inlet at near neutral pressure, like a static port, provides air for the measurement of gaseous water. The air from both intakes is transported to the instruments through electro-polished stainless steel tubing heated to 60°C. Just before the H_2O instrument, 0.6–3.1 vol-l/min of the two sample flows of 30–50 vol-l/min are extracted for passing through the three H_2O analysers.

The trace gas probe is located on the left, 7 cm above the aerosol probe. It consists

of a 3 cm inner diameter diffuser tube with an inlet orifice of 14 mm diameter and outlet orifice of 12 mm diameter providing an effective ram pressure of about 90–170 hPa depending on cruising altitude and speed. The inlet tip of the trace gas probe/intake is also heated to prevent icing. A small fraction of the air passing through the diffuser
5 is withdrawn from the center flow by a 12 mm ID PFA lined tube perpendicular to the flow direction. This trace gas tube is heated to 40°C. The design of the trace gas probe ensures separation of droplets as well as ice particles from the sampled air. The exhaust air from the container is vented at the rear end of the flange of the pylon close to the skin of the aircraft.

10 The DOAS telescopes (apertures of 1.5 degree) are mounted in a single aluminum block. Two 6 mm ports allow light to enter the sidewards (limb) looking telescopes (10 degrees below and above the horizon), whereas a similar hole in the inlet flange feeds light into the nadir telescope. The light from the telescopes is conducted into the container to the spectrograph/detector units of the DOAS instrument by three optical
15 cables each containing 4 individual quartz fibers. Details are given in Sect. 4.15.

A video camera (Model AVS470, Aerial View Systems, Newport Beach, CA, USA) with heated window is mounted at the leading edge of the main pylon, and allows inspection of the aerosol shroud to verify the absence of ice/rhyme and the observation of clouds during daylight flights. Between measurement flights, the inlet openings, and
20 the DOAS ports and the camera window are sealed.

The presence of an aperture in the aircraft's skin, which is integral part of the pressurized hull system, is of special concern. To assure structural integrity and to distribute loads a doubling flange at the outer fuselage (Fig. 3) and additional frames have been used. The structural concept of the inlet itself and the concept of the inlet to airframe
25 interface was developed by KOLT Engineering GmbH (Oberpfaffenhofen, Germany) in close cooperation with all partners followed by extensive calculations over a one year period for impacts of acceleration, vibration, fatigue and abuse loads (like bird strikes). Figure 5 shows a finite element based calculation of effect of loads on the combination aircraft hull and inlet system. The incorporation of supporting structural components

and a proper choice of materials and treatment ensures safe long term operation.

3.3 The CARIBIC container

The measurement container (Fig. 6) is a modified LD-11 airfreight container $(3.1\times1.5\times1.6=7.44\,\text{m}^3)$ with a maximum allowable gross weight of 3175 kg. As the ac-
tual gross weight is less than half of it, more equipment can possibly be integrated, although another limiting factor is given by the available electrical power and the amount of heat that can be dissipated in the cargo bay during the flight. The cost of air freight is determined by the volume of the container, not its actual gross weight. The container is based at MPI in Mainz and is placed on a hydraulic platform. This allows the container
to be lifted upwards and rolled into the institute's truck equipped with a roller system. At the airport a high-loader receives the container and lifts it into the aircraft.

The original LD-11 container, manufactured by Alcan Singen GmbH, was modified to meet the requirements given by the accommodation of the instruments and the safety considerations. The modifications included: 1) Using 9 cm high stringers covered by
an aluminum alloy plate, the single sheeted floor plate was turned into a stiff sandwich structure. 2) A rectangular opening was cut in the double floor to provide space for connecting the instruments to the container connector bracket incorporated in the cargo floor of the aircraft. 3) To enable access to the instruments from both sides for maintenance and calibration work, the rear wall of the container housing was replaced
by three removable panels and its front opening was equipped by two large doors. The front doors and rear panels increase the mechanical stability of the container and were designed with special emphasis to reduce emission of electromagnetic radiation. 4) Altogether six aluminum racks of four different types were manufactured to carry the instruments and the auxiliary equipment. All racks are 1.36 m high. The three double
racks and a single one behind the floor opening on the right of the Fig. 6 are 19″ wide. The two racks on the left are 650 mm wide. To facilitate mounting and dismounting, all racks are fastened to shock mounted frames using only 4 screws. For all racks the centers of gravity were calculated and this information was used in the calculations of

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the compatibility with acceleration requirements of up to 9g (depending on the x, y, z direction). 5) A ventilation system was incorporated. Four radial fans force air from the cargo bay into the sandwich flooring space from where it is distributed evenly around all equipment racks. Two additional radial fans withdraw air from the top right hand side (viewed in flight direction) and blow this waste air in the direction of the aircraft's cargo bay air intakes. 6) Two smoke and over-heat detectors as used in Airbus aircraft (Model SD9472-00, Apparatebau Gauting GmbH, Gauting, Germany) are mounted and are linked to a CARIBIC control panel in the cockpit.

The calculations, design and reinforcement of the container, and the construction of the racks were carried out by Enviscope GmbH in Frankfurt, Germany. The total weight of the container with the instruments is currently 1470 kg.

3.4 Sample air, electric and data systems

Figure 7 shows the layout of the tubing system in the bilge of the airccraft, starting at the inlet system via the inlet connector bracket in the bilge of the aircraft to the container connector bracket which is countersunk into the cargo floor. This container connector bracket contains the detachable power, signal, and air connectors. The air tubing connectors are Swagelok company's Quick-fit couplings, except for the aerosol tube which is a standard bulkhead union in order to prevent contamination of the aerosol samples by wear off from the o-rings. There are three 3/8″ O.D. electropolished stainless steel tubes. One is for aerosol particles and two supply the air for measurement of total and gaseous water. The latter ones are heated to 60°C. The air flow from the trace gas intake is split directly above the inlet connector bracket. The air for the whole air sampler is drawn through an unheated 1/2″ O.D. electro-polished stainless steel tube, whereas the air for most of the other trace gas instruments is led through a 3/4″ O.D. PFA lined aluminum tube kept at 40°C. This lining consists of a separate thin walled PFA tube. Each tube is connected on either side (inlet side, and container side) using flexible tubing. This measure is entirely because of aircraft structural considerations and means a complication for the analytical application. For the aerosol tube we used electrically

conductive PTFE in order to eliminate the possibility of electrostatic charging with the risk of particle loss. For the remaining of the tubing flexible PTFE was used as well. Furthermore, all flexible tubing is of the armoured type. The tube heaters are aviation quality Adel Wiggins's (Los Angeles, CA, USA) silicone rubber based ribbon heaters
5 and their temperatures are controlled using temperature controllers and 1000 Ω Pt temperature sensors (Minco, Minneapolis, MN, USA). The gases (i.e. air) exhausted by the container equipment are expelled through a 3/4″ stainless steel exhaust tube. The three quartz fiber cables (3 mm O.D.) from the DOAS telescopes to the analyzer are basically uninterrupted (fragile) connections from inlet to spectrographs, bundled
10 together and protected by a flexible sheath. The control unit (power and data conversion) of the camera is permanently mounted in the aircraft near the inlet connector bracket.

Figure 8 shows a photograph of the fairly crowded container connector bracket. Three electrical connectors provide power and data from the aircraft, and connect the
15 heaters and temperature sensors in the inlet system (a 60 pin connector). One BNC connector feeds the video signal from the video camera control unit. When the container is not onboard, the quartz fiber cables are carefully coiled inside a drum inserted in the tub (not shown in Fig. 8), the tube openings are sealed with blind connector plugs and the container connector bracket opening is covered by a floor panel cover.
20 The operation of the container can only be described briefly. After calibration, thorough testing, and inspection the container is installed by avionic personnel using extensive check lists well within 1/2 h during which also the inlet system, is inspected. In the cockpit on the CARIBIC control panel the system is activated for a measurement flight, while in addition the cargo temperature air conditioning is activated. After for
25 instance a sequence of 11, 2, 2, 11 h of flights the container is de-installed again. The great challenge is to have all 15 apparatus with a total of 20 analyzers work properly, and indeed resume so after each stop-over.

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4   Instrumentation

The scientific and related instruments are powered by two power supplies that convert 3 phase 115 VAC (400 Hz) into 24 and 28 VDC. The smaller of these supplies (Model EL 4kW MegaPAC, VICOR, Sunnyvale, Ca., U.S.A), called base power supply (BPS) (19″, 4 HU, 22.5 kg), provides 2 kW power for the instrument and master (control) computers and other essential functions during the take-off and landing when the power consumption has to be limited. The essential functions include the heat-up and evacuation of certain instrument parts which have a relatively long spin-up time. After take-off, the second, main power supply with a capacity of 8 kW (manufactured by Aircraft Electronics Engineering GmbH, Seefeld, Germany) (19″, 4 HU, 68 kg) is switched on. The actual total power consumption of the fully instrumented container is 5.5 kW and the peak load is 7.0 kW. For comparison, the total in-flight electrical power of an A340-600 is about 4 times 80 kW, yet electrical power usage by the container has to be kept low to minimize heat generation, to save energy, and to not affect any aircraft functions. Extensive use is made of exclusively aviation quality circuit breakers at the supply and at the equipment side.

The power delivered by each of the power supplies is recorded by the master computer. In case of smoke and/or overheat detection, an optical and acoustical warning signal is given by the CARIBIC panel in the aircraft cockpit, and the supply of power to the container system can be switched-off by the pilot. The position and meteorological data recorded by the aircraft are provided via an ARINC bus system and stored by the master computer in the container.

Besides the two power supplies, a data distribution box (19″, 4HU, 20 kg) forms a third interface between the container and the aircraft. It transfers signals from the smoke detectors to the aircraft, creates a relay signal indicating the presence of the container to the aircraft, transfers the ARINC aircraft data to the master computer, controls the temperature of altogether 18 heaters in the aircraft and the container (tip heaters of the inlet, tubing heaters in the pylon, the aircraft and the container), and

provides the power for the camera in the pylon.

All instruments in the CARIBIC container are controlled by the master computer. Its main function is to control the equipment status and to store different types of data (AR-INC, temperatures, pressures, communication with the instruments), but not measure-
⁵ ment data. The master computer communicates with the instruments via an Ethernet network consisting of two buses. After take-off (weight-on-wheels signal disappears), when the power consumption is no more limited, the main power supply is switched on, and from then on the master computer activates sequentially the individual instruments in an orderly manner to avoid power surges. In flight, the computer checks the
¹⁰ status of the individual instruments through simple algorithms and initiates their shut down if the communication is lost. In addition to controlling the equipment status, the computer monitors the conditions inside the container, the inlet system and the sample tubing system using 4 internal pressure sensors, 2 external temperature sensors, and 18 signals from the temperature controllers in the data distribution box. The third major
¹⁵ function of the master computer is the storing of the ARINC data. ARINC (in this case ARINC 429) is the most widespread commercial avionic data bus system. This extensive set of flight data is standard available in civil aircraft, and provide among others the pressure altitude, static temperature, longitude, latitude, time, and flight angles.

Another central provision in the CARIBIC container is the gas supply unit. It was
²⁰ decided to have all calibration gases and supply gases stored in aluminum cylinders inside one rack that also contains the pressure regulators. The only exception is a cylinder with calibration gas used for the PTRMS which for analytical reasons has to be close to the PTRMS instrument. The supply of oxygen for the production of ozone used in the NO and NO_y analyses is in standard aviation steel cylinders with breathing quality
²⁵ oxygen. These cylinders are additionally protected by concentric steel cylinders. The reason for this is to comply with the safety requirement that if in case of an engine burst, fan blades that are ejected at high speed penetrate the fuselage and container, these cannot possibly damage the cylinders to that degree that a rapid release of oxygen would result.

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Table 1 gives the overview of the instruments in the new CARIBIC container with the main specifications and lists the institutions responsible for their development and operation. Figure 9 shows schematically the tubing in the container providing air sample and collecting the exhaust gases of the instruments. Most instruments are equipped with internal pumps (not shown). The relevant aspects of the individual instruments are described below.

4.1 Total and gaseous water

The budget of atmospheric water vapor is of great relevance for climate. It has recently been shown that a significant fraction of upper tropospheric air masses between 40-60 ^{0}N is supersaturated (up to 175%) over ice, a feature until very recently neither captured by the ECMWF meteorological forecast model, nor by climate models. An objective of CARIBIC in this respect is to collect additional information about distributions of over-saturated regions and of the amount of water trapped in cirrus clouds

The water vapor instrument (19$''$, 6 HU, 29 kg) comprises two totally different detectors. A commercial chilled-mirror frost point hygrometer (CR2, Buck Research, Boulder, USA) measures humidity using the well established (absolute) method. Next to this, a two-channel photo-acoustic (PA) laser-spectrometer developed at the Department of Optics and Quantum Electronics at the University Szeged in Hungary (Bozóki et al., 1999 and 2003; Szakáll et al., 2001) measures the water vapor content in a relative mode using the laser light to excite water molecules and measuring their thermal relaxation. The technique is much faster than the frost point hygrometer.

The Buck CR2 hygrometer and one of the PA channels are supplied with air from the forward facing (total water) inlet. The air flow through the 3/8$''$ OD (7.55 mm ID) SS electro-polished inlet tube (heated to 60°C, overall length: 2.6 m) is forced by the ram pressure of 60–100 hPa. An orifice upstream of the water vapor instrument restricts the air flow to ~50 vol-l/min, by which ~75% of the ram pressure is maintained. The air is discharged through the outlet tubing connected to the exhaust in the pylon. The pressure at this point is close to the static outside pressure. Only a small sampling

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flow of 3.1 vol-l/min (driven by the remaining 45–75 hPa pressure difference across the flow restrictor) is branched-off from the main airflow. The split that feeds the air to be analyzed is a 1/4″ OD (4.37 mm ID) SS electro-polished sampling line (heated to 60°C), connected directly upstream of the flow restrictor. The Buck CR2 hygrometer receives a sample flow of 2.5 vol-l/min with the balance of 0.6 vol-l/min being passed through the respective total water PA channel.

The second PA channel is connected to the sideward facing inlet via an identical 3/8″ OD SS inlet tube kept at 60°C. As this inlet opening is slightly below ambient pressure, a 24V DC pump is used to assist the same air flow of 40-60 vol-l/min by suction. Also in this case a sampling flow of 0.6 vol-l/min is branched-off and fed into the second PA channel.

Laboratory based comparisons with a standard chilled mirror frost point hygrometer (MBW 373, MBW Calibration Ltd., Wettingen, Switzerland) give a precision and total uncertainty of the Buck CR2 hygrometer of ~0.5 K. The precision of the two PA spectrometers currently is ~0.5 μmol mol^{-1} at ambient pressures and ~1 μmol mol^{-1} at 150 hPa. The time resolution is 30–180 s for the Buck instrument and 5–15 s for PA sensors, depending on absolute humidity and pressure. The performance in flight is determined by the instrument characteristics, airflows, and by the inlet system, including the heated tubing. An example of the possibilities offered by the H_2O instruments is given in Fig. 10, highlighting the detection of cirrus clouds. The quantification of the water content of the clouds requires an improved assessment of the water droplet enhancement which is in progress.

4.2 Ozone

Ozone is essential and forms a protective layer in the stratosphere against UV radiation, but in particular in the UT/LS it also plays a role in the IR radiation budget. In the UT/LS ozone is an important transport tracer and was for instance used to define a chemical tropopause (Zahn and Brenninkmeijer, 2003). In the troposphere its chemical importance is also crucial since it is the major precursor of the hydroxyl radical.

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For ozone we also apply 2 different detection techniques, namely a very fast solid-state chemiluminescence detector and an accurate, precise dual-beam uv-photometer based on the most frequently used detection principle. The ozone instrument (19″, 6 HU, 29 kg) takes its sample air from the trace gas probe of the inlet system, using the "reactive trace gases" (PFA-coated) tubing to the container (Fig. 7).

The custom made dual-beam uv-photometer measures the light absorption of O_3 in the UV (Hartley band). Light from a low-pressure mercury discharge lamp emitting mainly at 253.7 nm (>90%) is focused through two 38 cm long optical cells. One cell contains the ambient air, the other one ambient air scrubbed of O_3. Major parts of the optics are adapted from the commercial instrument M49 of Thermo Fisher Scientific. The sample flow of $\sim$3 l min^{-1} (volume) is maintained by the ram pressure of the trace gas inlet, i.e., a pump is not needed. The electronics for amplifying and processing the signal as measured by a CsTe photocathode (Hamamatsu, R1228) was newly developed. Using extremely low-noise amplifiers and an integrating 24 bit A/D converter, it measures the light intensity with a frequency of 122 Hz, in contrast to the common sampling rate of typically 0.1 Hz. This high-frequency measurement allows for a partial elimination of photon flux fluctuations that are characteristic for mercury discharge lamps. Comparisons with an extremely stable (but slow) laboratory standard (UMEG-NIST which agrees with the NIST standard SRP #15 within <0.5%) shows differences below 0.5 nmol mol^{-1} over the studied measurement range of 2–400 nmol mol^{-1}. The 1-σ precision is currently 0.3 nmol mol^{-1} at a measurement response time of 4 s and an accuracy of $\sim$1%. This is a factor of 2–3 faster and more accurate compared to similar instruments. Like other UV photometers, it constitutes in principle an absolute measurement device, i.e., the O_3 mixing ratios is calculated solely by using known physical properties (O_3 cross-section, optical path length etc.) and a ratio of measured voltages. Therefore, a calibration is not required and the instrument can be viewed as a (flying) O_3 standard for the fast instrument described below. Nevertheless, the instrument is cross-checked regularly with other O_3 standards. Interferences with water vapor are extremely small mainly due to the newly designed O_3 scrubber that has a

very small surface for water adsorption.

The fast O_3 sensor is based on the chemiluminescence of O_3 on the surface of an organic dye adsorbed on silica gel powder deposited on a 1 cm diameter aluminum disk (Güsten et al., 1992). The measurement frequency is 10 Hz, which, at the cruising speed of ∼250 m/s, result in a high spatial resolution of merely ∼25 m. However, turbulence within the inlet tube causes mixing of air which degrades the spatial resolution to effectively 30–40 m distance, still ample for our purposes. The fast O_3 sensor is linear but needs calibration because of a variable sensitivity. The UV photometer measurements are used during the post flight data processing for defining the fast sensor's scale. An example of the ozone data from UV photometer is shown in the flight-overview, Fig. 11.

4.3 Carbon monoxide

Carbon monoxide (CO), together with methane, constitutes the major global atmospheric burden of reduced gases, and as such affects the oxidative self-cleansing process of the troposphere. It is a major pollutant (energy generation, transport, biomass burning) and is produced in situ in the oxidative chains of methane and other hydrocarbons. CO levels decline significantly crossing the LS, whereas in the UT the frequent large scale pollution plumes carry large amounts. CO and O_3 are often studied together, with CO being an indicator for pollution and a precursor molecule for O_3 formation. The challenge of carbon monoxide measurements in CARIBIC is to have fast, precise data with an internationally accepted calibration.

Gas chromatography with a mercury oxide based reducing gas detector, as used in CARIBIC #1 has been the standard measurement technique for decades. The serious disadvantages are the limited spatial resolution (GC technique) and the non-linearity of the detector. Therefore, a vacuum ultraviolet (VUV) fluorescence instrument (Volz and Kley, 1985; Gerbig et al., 1996, 1999) has been incorporated instead. Here, CO molecules are excited by VUV radiation in an optical cell at a pressure of 10 hPa. The VUV radiation is generated by a RF generated discharge in a cell flushed with a CO_2-Ar

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mixture, while the fluorescence is measured using a photomultiplier. Because oxygen strongly absorbs in the VUV range used, the region between the light source, detector and photomultiplier is flushed with nitrogen gas.

The CO analyzer (19″, 4 HU, 21 kg) is based on a commercial instrument (Model AL 5002, Aero-Laser, Garmisch-Partenkirchen, Germany) which was retro-fitted with valves for automatic calibration and communication board for remote control by the master computer. A flow controller has been added to stabilize the flow of nitrogen used to purge the fluorimeter in order to achieve a more stable detector signal. The flow rate of air is 80 ml min^{-1} at standard conditions (STP, i.e. 273.2 K, 1013.2 hPa). The housing had to be modified to ensure that electromagnetic interference through conduction and radiation (e.g., for the discharge lamp) is absent.

The advantages of the VUV technique are excellent linearity, reproducibility, and fast response. Tests on air cylinders show agreement with other laboratories within fractions of a nmol mol^{-1} over a range of 50 to 300 nmol mol^{-1}. Results are calculated relative to the NOAA/ESRL standards based on as comparisons at the GAW quality assurance center at EMPA, Dübendorf, Switzerland. The time resolution obtained is ~2 s. An example of the CO measurements is presented in the flight-overview, Fig. 11.

4.4 Nitrogen oxides

NO is one of the main catalysts in tropospheric photochemistry. Produced by soils, lightning and combustion processes it reacts readily with ozone, resulting in NO_2 and O_2. At daytime, NO_2 is photolysed, releasing an oxygen atom that reforms ozone. The important catalytic role is clear in reactions such as $HO_2 + NO$, which yields OH and NO_2. NO recycles radicals and maintains the oxidative cycle. In CARIBIC we measure NO and NO_y (the sum of all reactive nitrogen species: NO, NO_2, HNO_3, PAN, 2 N_2O_5, HNO_2, HNO_4, and nitrates), with the future option of also detecting NO_2. In the stratosphere total reactive nitrogen (NO_y) consists mainly of HNO_3. In the troposphere the NO, NO_2, HNO_3 and PAN are the main components of total reactive nitrogen. Systematic data for nitrogen oxides in the UT/LS region are rare which underscores

the importance of this payload in CARIBIC.

The technique applied for the measurement of NO and NO_y is similar to the one used on the DLR *Falcon 20* research aircraft for several years (Ziereis et al., 2000). It is based on detecting the light emission from the reaction of NO with O_3 using a photomultiplier. The sum of reactive nitrogen compounds (NO_y) is measured by a catalytic conversion technique in which all NO_y species are reduced to NO in a gold tube maintained at 300°C using a reducing gas added to the sample air flow. Two NO detectors allow the concomitant measurement of NO and NO_y. For measurements in research aircraft CO is routinely used as reducing gas. However, for its toxicity it is not allowed on civil aircraft. Therefore, hydrogen from a small metal hydride storage system (storage capacity 2 g H_2) is used here. A series of laboratory measurements was performed to study the conversion efficiency for typical NO_y compounds using H_2 instead of CO. This showed that there is no change in the efficiency of the NO_y converter when using H_2.

The NO+NO_y system constitutes one complete rack (19$''$, 24HU, 171 kg) stretching from the front to the back of the container. The chemiluminescence detectors are operated at a sample flow of 1.5 l min^{-1} (STP) extracted from the PFA lined trace gas sampling line. To avoid the loss of nitric acid, a major compound of NO_y, PFA sampling lines heated to 40°C are used also inside the analyzer systems. For obtaining an optimal signal to noise ratio, the photomultipliers are cooled, which is achieved by Peltier elements.

Sensitivity checks of the two NO detectors are carried out before and after each flight using a standard concentration of NO in N_2. The NO standard is diluted into the NO free zero air by means of a mass flow controller. The sensitivities of the two NO channels are about 15 counts mol s^{-1} pmol^{-1}. The background signal is determined every 10 min for a period of about 1 min. The conversion efficiency of the gold converter is checked before and after each flight by adding known amounts of NO_2 produced by gas phase reaction of NO with O_3. Zero air artifacts are also determined before and after each flight and also during the flights itself. The detection limit of the NO and NO_y

system for 10 s data is about 1 to 2 pmol mol^{-1} (2σ). The estimated accuracy for the NO and NO$_y$ measurements is 8 % for an ambient NO level of 50 pmol mol^{-1} and 7 % for an ambient NO$_y$ level of 450 pmol mol^{-1}. During the POLINAT-2 (Pollution from Aircraft Emissions in the North Atlantic Flight Corridor) and EXPORT 2000 (European Export of Particulates and Ozone by Long-Range Transport) campaigns NO and NO$_y$ measurements on board the Falcon 20 made by DLR were compared to similar measurements on board other aircraft during flight. The instruments on the different aircraft showed good agreement (Ziereis et al., 1999; Brough et al., 2003). An example of the NO and NO$_y$ data is given in the flight overview, Fig. 11.

4.5   Aerosol particles – condensation particle counters

Aerosol particles are emitted in the course of a range of natural and man induced processes. As they are also formed in situ by gas-to-particle conversion, their physical and chemical properties vary strongly. Also their number distribution is highly variable mainly due to their short atmospheric residence time in the order of days. Aerosol particles affect the Earth's radiation budget directly through light scattering and absorption and indirectly (cloud processes), clearly having a significant - yet ill quantified - impact on climate (IPCC, 2007). In view of the relatively modest information we have about UT/LS particles, this aspect of CARIBIC has some priority.

   The CARIBIC container houses three units with aerosol apparatus, which are supplied with air via electro-polished stainless steel tubing from the aerosol probe. These units are located as close to the container connector bracket as possible to minimize loss of particles in tubing. The low pressure needed to maintain the flow through the systems using critical orifices is provided by a diaphragm pump in the pump unit of the container. This unit (19″, 8HU, 55 kg) contains two two-stage membrane pumps (MD4, Vacuubrand). In the units named AERO 1 (19″, 6 HU, 30 kg) and AERO 2 (19″, 5 HU, 42 kg) altogether three modified condensation particle counters (CPCs, TSI Model 7610, TSI Inc., St. Paul, MN, USA) are installed. AERO 2 also contains the particle

sampler unit described in Sect. 4.7. CPCs measure the integral particle number concentration of all particles larger than a certain threshold diameter. The sample air is first saturated with the vapor of a working fluid, here butanol, and cooled subsequently in order to grow the particles by condensation of the working fluid. The enlarged particles are finally detected by scattering of light from a diode laser. Extensive laboratory tests at operating pressures of 160–1000 hPa were made to ensure the proper function of the instruments at cruise altitudes, and to obtain pressure-dependent counting efficiency curves (Hermann and Wiedensohler, 2001).

To measure different particle size ranges, the lower threshold diameters of two CPCs were shifted to a smaller and a larger diameter respectively by changing the temperature difference between the saturator and the condenser block. The new temperature differences are 11°C and 26°C (compared to 17°C in the original version) and additional temperature sensors were used to monitor the temperatures of the saturator and the condenser block in each CPC. Consequently, the three CPCs now have lower threshold diameters (50% maximum counting efficiency) of 4, 12, and 18 nm, respectively, at 200 hPa operating pressure (Hermann and Wiedensohler, 2001). The upper threshold diameter of particles detected by the CPCs ($\sim 2\,\mu$m) is determined rather by particle losses inside the CPCs than by the particle sampling efficiency of the CARIBIC inlet system.

The time resolution of the CPCs is set by the control software to 2 s, corresponding to a spatial resolution of about 0.5 km. During data processing, measured number concentrations are corrected for particle losses in the inlet and in the sampling line, for CPC counting efficiencies, for coincidence in the CPC optics, and for pressure-dependent CPC flow rates. These volume flow rates, which are determined by critical orifices in the CPCs, range from 1.1 to 1.6 liter/min (volume) and have been calibrated in the laboratory for operating pressures between 60 and 1000 hPa.

Results of the CARIBIC CPC measurements onboard the Boeing 767-300 ER of LTU International Airways were presented in several publications. The results using the new inlet system and longer sampling lines onboard the CARIBIC Airbus A340-600 are

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similar to the previous data with the simpler inlet system without shroud and shorter transfer tubing. Figure 12 shows an example how particle counts can be combined with the video recordings (Sect. 4.14) to reveal the exact location of particle formation events.

4.6 Aerosol particles - optical particle counter

The third aerosol rack unit (19″, 7 HU, 26 kg) houses an Optical Particle Counter (OPC, PCASP-X, Passive Cavity Aerosol Spectrometer Probe, PMS, Boulder, CO, USA). OPC operates on the principle that the light scattered by individual particles within a laser cavity is a direct function of their size, assuming spherically shaped particles. Each particle produces a pulse of radiant energy during its transit through the laser beam, which is detected by two photo diodes. As the OPC is intended to measure the aerosol particle size distribution in the size range of 0.15–10.0 μm (accumulation and coarse particle mode), about six orders of magnitude in signal intensity have to be covered. Hence, the two diodes have different sensitivities. The loss of particles in the CARIBIC inlet system limits the maximum diameter of detected particles to approximately 5 μm.

For CARIBIC, the original instrument was considerably modified. New photodiodes and new signal processing electronics were installed. The new analog-to-digital converter allows the binning of particles into 256 size channels instead of 32 in the original instrument. The sheath (0.5–1.0 liter/min) and sampling flow rates (0.05–0.2 liter/min) through the OPC are controlled by two mass flow controllers. In order to maintain a constant volume flow rate, the sampling line pressure and temperature are controlled. As for the first two aerosol units, this flow is maintained by the pump unit in the container. The time resolution of the OPC is software controlled and was set to 20 s. However, because of the low concentration of accumulation and coarse mode particles normally encountered in the UT/LS and hence statistical reasons, the time resolution is only on the order of minutes in practice.

The OPC has been operating less regular than foreseen because of different teething

problems. Nevertheless, some excellent results haven been obtained. Shown in Fig, 13 is a comparison of aerosol sulfur mass as detected by PIXE elemental analyses of collected aerosol samples (see following section) and as calculated from the particle size distribution derived with the OPC. Differences were observed in the equatorial region. It is quite possible that these mass differences are actually organic carbon (e.g. Murphy et al. 2006), which is not detected with PIXE.

4.7 Aerosol particle sampling

Aerosol particle collection proved very valuable during the first phase of CARIBIC. Presently a new multi-channel impactor sampler (Nguyen et al., 2006) collects particle samples for quantitative elemental analyses by Particle-Induced X-ray Emission (PIXE) and Particle Elastic Scattering Analyses (PESA) (Papaspiropoulos et al., 1999). In addition, material is collected separately for analyses of single particles with transmission and scanning electron microscopy (TEM, EELS and EFTEM). Particles are collected by impaction on ultra thin, clean plastic foils housed in a sampler block ($16 \times 16 \times 46 \, \text{cm}^3$) inside the AERO 2 unit. It contains 14 channels operated by solenoid valves for sequentially collecting samples ($\sim$1.5 h time resolution) and two channels sampling the whole flight ($\sim$8–10 h). Each sequential channel collects simultaneously one sample for PIXE/PESA analyses and two smaller samples for single particle analyses. A cyclone separator up-stream of the sampler restricts the upper particle size entering the sampler to 2 μm diameter. Calibration with 1013 hPa inlet pressure at the sampler revealed a lower cut-off diameter (50% level) of 0.08 μm. During operation in the UT/LS the reduced pressure will result in an even smaller cut-off diameter.

Figure 13 shows the comparison between elemental analyses and the OPC results, indicate a deficit in mass as detected by PIXE, assuming a certain sulfate content of the particles (100%). PESA analyses to be conducted may help to reconcile the mass budget insofar the assumptions about mass based on light scattering are correct.

4.8 Carbon dioxide

Understanding and quantifying the fluxes of the main greenhouse gas CO_2 on the global scale is a gargantuan task. CARIBIC can contribute accurate data for the UT/LS region for which the present global dataset is very sparse. In flight, the gas also is an important tracer for understanding the composition air masses intercepted as to their origin, like biomass burning, contact with the biosphere during day and night, etc.

The CO_2 analyzer (CONDOR-C, 19″, 6 HU, 37 kg) uses the common non-dispersive infrared absorption (a LICOR 6252, LI-COR Biosciences, Lincoln, NE 68504, USA). The detection is performed at 4.26 μm in a differential mode. The accuracy and the reproducibility are enhanced by using in flight calibration with a set of 3 gases, and by controlling the pressure and flow rate in the optical cells. The spectrometer temperature is kept at 25°C through the combination of a heating element and a fan that produces rapid circulation. The pressure of the sample and the reference cells are regulated at 1080 hPa with an accuracy <0.1 hPa by using two absolute quartz pressure sensors (Druck RPT 200). The flow rate at standard pressure and temperature (STP) of the sample air and the reference gas are also controlled during flight respectively at 600 cm^3 min^{-1} and 70 cm^3 min^{-1} (volume). All process controls are performed under Real Time Linux on a PC104 CPU.

To exclude interference of water with the CO_2 measurement, the sampled air is dried using a bundle of Nafion membrane tubes. The instrument is calibrated using two calibration gases every 30 min. The set of calibration gases in high pressure cylinders is calibrated before and after the flight at LSCE in Paris following the calibration protocol of the CARBOEUROPE project. The precision of CONDOR was determined aboard the DLR's Falcon, during the CAATER (Co-ordinated Access to Aircraft for Transnational Environmental Research) missions (LSCE, unpublished data), to be ~0.15 μmol mol^{-1}. Operation in CARIBIC has proven to be less routine as projected with the performance gradually improving.

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4.9 Oxygen with ultra high precision

Variations in the O_2/N_2 ratio in air not only mirror those in CO_2, but provide additional essential information about the complex interaction between the atmosphere, oceans, and biosphere. To obtain useful information, variations in the O_2 content of air have
5   to be measured with about the same absolute precision as that for CO_2. This means that the relative precision has to be 1000 times better than that for CO_2 measurements. Bearing in mind that it took the "CO_2 community" decades to achieve traceable accurate precise measurements, this obviously is a considerable analytical challenge. As the O_2/N_2 ratio measured in an air flow is prone to variability and biases introduced by
10   fractionation in the inlet systems, the incorporation of equipment to measure the O_2/N_2 ratio in CARIBIC is of experimental nature.

In our opinion the optimal choice of analytical principle for an aircraft is the electrochemical reaction technique. Electrochemical cells basically provide a small electric potential (of the order of 20 mV for 20% of O_2) proportional to the amount of O_2 present,
15   i.e., the partial pressure. Compared to detectors used in the several other methods that have been developed over the last decade or so, these cells offer the advantages of being robust, small and cheap. Their commercial usage is in the measurement of O_2 in automobile exhaust gases. Several cells are used simultaneously. The essence of the O_2 system is to obtain an extremely high degree of control over temperature, pressure
20   and flux of the air in the electrochemical cells. Moreover, the electronic processing of the weak electrical signal (only 1 mV per % O_2) demands a range of measures to enable a sufficient signal to noise ratio at ambient O_2 concentration of approximately 21% with a resolution of 0.0001% O_2 to match the CO_2 resolution.

A unit (19″, 6 HU, 39 kg) contains the small electrochemical cells, temperature/flow/
25   pressure control, computer and also an additional CO_2 analyser intended to help troubleshooting for the difficult O_2 measurements (e.g. fractionation effects). The amplifier unit contains state of the art electronics minimizing noise, giving a noise/signal ratio in the order of 1 part per million only. The short term instability of the O_2 cell requires

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frequent switching between sample and standard gases (as it is done in isotope ratio mass spectrometers) which cancels much of this variability. Exactly this process is not yet operating to the level required in our instrument since (i) the control of flow is not yet adequate and (ii) the sensitivity strongly depends on the absolute flow level. High flow results in a significantly reduced output signal. Hence the noise to signal ratio drops below values that are interpretable. Therefore, oxygen measurement precision is still slightly too low and the four fuel cells arranged in sequence are behaving differently. Most probably this is due to very small temperature fluctuations due to gas flow variations (ca. 0.1°C).

4.10 Mercury

Measuring mercury is of scientific interest because its biogeochemical cycle is not well understood. Moreover, its toxicity, its enrichment in the aquatic food chain and its accumulation in the cryosphere are a cause of concern. There exist relatively few mercury measurements, in particular in the UT/LS, and the global dataset to study transport pathways can be significantly increased by CARIBIC measurements.

Because of the exceedingly low concentrations at which Hg species exist in the background atmosphere, measurement techniques incorporate a pre-concentration step (amalgamation with precious metals, preferably gold). In the remote atmosphere the total gaseous mercury (TGM) fraction consists of gaseous elemental mercury (GEM), the dominant compound by far, and the operationally defined reactive gaseous mercury (RGM) present in much lower concentrations. Beside GEM and RGM, other volatile organic mercury species, such as dimethyl mercury and monomethyl mercurychloride also interact with gold absorbers. Altogether RGM and organomercury compounds represent only a few percent of TGM under most background conditions.

The CARIBIC instrument is based on automated dual channel, single amalgamation, cold vapor atomic fluorescence analyzer (Tekran-Analyzer Model 2537 A, Tekran Inc., Toronto, Canada) which measures TGM at background concentrations. The instrument consists of one rack unit (19″, 7 HU, 36 kg). The sampling interval, initially set to

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15 min (~225 km flying distance) with an air sample flow rate at STP of 0.5 l min^{-1}, has been reduced to 10 min since April 2006. The instrument features two gold cartridges. While one is adsorbing mercury during a sampling period, the other is being thermally desorbed using argon carrier gas stream and Hg is detected using cold vapor atomic fluorescence spectroscopy (CVAFS). The functions of the cartridges are then reversed, allowing continuous sampling of the incoming air stream. A 45 mm diameter Teflon pre-filter (pore size 0.2 μm) protects the sampling cartridges against contamination by particulate matter.

The compression of sample air from the trace gas inlet using the PFA coated tubing into the dual gold cartridge assembly (500 hPa) was achieved by an additional pump inside the unit. This membrane pump has been extensively tested for not contaminating the system with Hg. Since the response factor of the Tekran instrument is pressure dependent (Ebinghaus and Slemr, 2000), a pressure monitoring module is placed at the sample vent. This allows for an empirical pressure correction algorithm normally applied to airborne operations with large altitude changes. A detection limit ~0.1 ng Hg m^{-3} is achieved. The instrument is calibrated after every other flight by repeated injection of known amounts of mercury. An example of the TGM data is shown in the flight overview in Fig. 11. Results of mercury measurements in biomass burning plumes have been published recently and we refer to this work for additional information (Ebinghaus et al., 2007).

4.11 Oxygenated volatile organics by proton-transfer-reaction mass-spectrometry (PTRMS)

Oxygenated volatile organic compounds (OVOCs) are omnipresent be it at low to minute concentrations. These mostly short-lived substances are produced in the chain processes of hydrocarbon oxidation and emitted from combustion and burning. In the upper troposphere they may substantially contribute to the formation of radicals (Singh et al., 1995, 2004). Up to now only a rather limited number of measurements of OVOCs

are available in the UT/LS, exclusively gathered during certain research aircraft campaigns.

The problem of measuring OVOCs has been partly solved by the development of PTRMS by Lindinger et al. (1998) and Hansel et al. (1995)and their colleagues. Its applicability for airborne deployment was demonstrated by Hansel et al. (1999) and de Gouw et al. (2004). The adaptation and automated operation of a PTRMS system has formed a major challenge for CARIBIC. PTRMS applies a soft ionization method (chemical ionization) for trace gases that have a greater proton affinity than water (166.5 kJ/mol). This allows specific ionization and thus the separation of these trace gases from the air matrix molecules. H_3O^+ ions are generated in a hollow cathode discharge in water vapor. The proton transfer reaction takes place in a drift tube (ca. 2.8 hPa). The applied drift tube voltage of $-600 - +700$ V determines the reaction time and thus (together with the pressure) the conversion efficiency and sensitivity (De Gouw et al., 2003). The sensitivity for acetone, for example, is ~ 100 counts mol s^{-1} nmol^{-1} corresponding to a detection limit of ~ 28 pmol mol^{-1} for an integration time of 5 s.

A PTRMS system (Ionicon, Innsbruck, Austria) was modified for automated on-line measurement of selected OVOCs, specifically acetone, methanol, acetaldehyde, and VOCs, specifically acetonitrile. The unit is integrated in a wide rack (26$''$, 22 HU, 123 kg). Improvements were necessary to detect the low concentrations generally occurring in the UT/LS, to automate its operation, to make it compact, and to fulfill the aviation safety requirements. The major modifications are: a) the power supply changed to 24 and 28 V DC, b) the integration of effective filters to suppress electromagnetic interference, c) a reduction of power consumption, instrument size and weight, d) the installation of a third turbo-molecular pump enabling to increase the number of primary ions and to operate the PTRMS at higher drift tube pressure, e) the integration of a more powerful (and much smaller) high-voltage supply for the drift tube, f) the implementation of shock mounts for the most sensitive parts, g) the incorporation of a cylinder with calibration gas (Apel/Riemer, USA) for online calibration during flight and of a zero air filter (Pt-catalyst) for the determination of the background signal, h) the

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addition of temperature control (to ~35°C) of all tubes (made of PFA) that are in contact with the analysed air, i) the implementation of a 3/8″OD PFA bypass line that flushes the inlet line with ~20 l min^{-1} (controlled by a flow controller), and j) a design and an implementation of new measurement and control software (based on LabView).

An example of the usefulness of the PTRMS instrument is shown in the flight overview of Fig. 11, giving results for acetonitrile (CH_3CN), acetone $CH_3C(O)CH_3$, and methanol (CH_3OH). Levels of CH_3CN are generally fairly constant (lifetime ~1year), yet it is a good indicator for biomass burning. Acetone - an important carbonyl component – shows many interesting features related to its direct emissions and omnipresence in the troposphere, and chemical reactivity. Methanol is one of the most abundant OVOC in the atmosphere.

4.12   Air sampling

Collecting air samples is a simple as powerful. Long-lived gases, particularly CO_2, CH_4, N_2O, SF_6, can be measured in the laboratory with required high precision and accuracy, as well as a host of other gases that cannot be analysed rapidly during the flight, such as halocarbons and hydrocarbons. When one includes isotope measurements, ^2H in H_2, ^{13}C and ^{18}O in CO_2, ^{13}C, ^{17}O, and ^{18}O in CO, ^2H and ^{13}C in CH_4, ^2H and ^{13}C in several hydrocarbons, ^{37}Cl in CH_3Cl, most extensive information about a sampled air parcel is obtained. A limitation of air sampling is that the trace gases of interest must be sufficiently stable in the air container. A drawback is that discontinuous information is obtained.

The whole air sampler TRAC (triggered retrospective air collector) consists of a pumping unit and two sample canister units (together 19″, 24 HU, 137 kg) all under control of a separate computer unit, which also controls the absorber unit (19″, 3 HU, 7 kg, see Sect. 4.13). The TRAC is designed to collect sufficient amount of air (about 10 liter STP) within a short time (minutes) in order to resolve smaller scale features, e.g. those associated with convective uplift of boundary layer air, or tropopause folds. In the simplest sampling mode, now in use, samples are taken at regular intervals. The

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pumps are switched off between two samplings to reduce their wear out and to lessen the heat production. In the triggered mode, foreseen for the future, the pumps will be run continuously and the sampling will be triggered by signals from the ozone, CO or aerosol instruments. As the sample air is flowing continuously through a canister, there is some degree of retrospective sampling. The amount of collected air should be sufficient for analyses of hydrocarbons, halocarbons, greenhouse gases and, in the future, of isotopic composition of CO_2, CO, CH_4, and H_2. As the analyses are made using up to 5 different analytical instruments in 3 countries, altogether 3 sets of sample canister units were built to secure the availability of one set for a monthly flight or at an even higher flight frequency. Compared to the previous CARIBIC air sampler, the only trace compound that cannot be measured is the ultra low concentration tracer ^{14}CO which has an abundance of typically only 10 molecules per cm^3 of air STP.

Glass is used for the construction of the canisters. Although this is technically somewhat more elaborate, its inertness has overriding advantages. Also due to its semi-liquid nature, the surface area is smallest. Another advantage of glass containers is that CO_2 and its isotopic composition remain unchanged.

The entire system consists of three units: a three stage pump unit and two canister units with 14 glass canisters each. Two metal bellow pumps (Model PWSC 28823-7, Senior Aerospace Inc., Sharon, Mass., USA), the first one with its two bellows in parallel as a first stage, and the second one with its bellows in series as the second and the third stage, are used to pressurize the air from the 200–250 hPa inlet pressure to about 4 bar. These pumps as supplied by the manufacturers as potable water pumps for aircraft use actually have 0.5 mm holes drilled in the bellows' flanges presumably to automatically release pressure when not running. We sealed these small vent holes using washers with Viton o-rings. Mechanical pressure release valves (Nupro) are used to prevent over-pressurization. The 2.74 liter glass vessels are manufactured by Louwers Hapert Ltd. (Hapert, the Netherlands) from borosilicate glass tubing (5 mm wall thickness), and rated for a pressure of 7 bar. The vessels are sheathed with a polymer liner and housed inside a structure with 14 pigeon holes. They are connected to

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the closing valves via stainless steel tubing which is cemented (Araldite 2020, Vantico AG, Basel, Switzerland) in two glass capillaries protruding into the cylindrical vessel at each end. Two 16 position Valco valves (VICI Valco Instruments Co. Inc., Schenkon, Switzerland) are used to switch between canisters. Usually all canisters are filled during the forward flight. During the return flight, half of them are vented and refilled.

Sample analyses of over 250 samples collected until December 2006 show that some trace gases that are difficult to preserve in stainless steel canisters, such as alkyl nitrates and DMS, are preserved. Some results are illustrated in the flight overview of Fig. 11.

4.13 Trace gas sampling using adsorption tubes

Measurement of low level trace gases that cannot be collected with flasks because they are lost at surfaces is a challenge and the deployment of a PTRMS (Sect. 4.11) is one strategy. Another way to measure VOC and OVOCs is to pre-concentrate them reversibly on substrates that do not absorb the abundant atmospheric gases and to perform post flight analyses. In the UT/LS the dryness of the air is conducive to adsorptive sampling, yet ozone may pose problems for sampling of some compounds.

A system based on 16 adsorption tubes for the determination of selected OVOCs was constructed (19″, 5 HU, 21 kg). The sampler uses standard size Silcosteel tubes (3.5″ long, 1/4″ O.D., Restek, Bellefonte, PA, USA) that contain the adsorbent, presently Carbopack X. The tubes are located in channels in an aluminum block that is cooled by Peltier elements to a temperature of down to –15°C. The reason for cooling is to be able to select and maintain stable temperatures below the container temperature which may reach 25°C.

To prevent contamination the sample air is drawn through the tubes by a pump downstream of the tubes. The air flow is controlled using a low resistance flow controller. The adsorption tubes are switched using two 16 position Valco valves (VICI Valco Instruments Co. Inc., Schenkon, Switzerland). The switching of the valves, the pump and the flow controller are controlled by the same V25 computer which also controls

the whole air sampler. The unit functions well with research on recovery and interferences in progress.

4.14 Video camera

During CARIBIC phase #1 particle events have been observed, often in association
with strong convection. However, there was no information about the presence of
clouds, because the old system did not have respective water sensors. In CARIBIC
phase #2 information on clouds can be gained not only by the fast water sensors (cf.
4.1), but also by a video camera, which observes the surrounding of the aircraft in
forward direction. Another important function of the video camera is to check whether
ice builds up on the leading edge of the shroud, which would influence the particle
collection efficiency. Color images of 768×494 pixel resolution are recorded at a 2 Hz
repetition rate and stored on a video recorder in the OPC unit in the container. An
electronic shutter with 10^{-4} s time response prevents damaging of the CCD sensor in
case of direct view into the sun. The images obtained show the presence of contrails,
other aircraft, clouds, and the occasional occurrence of lightning (at night).

4.15 DOAS based remote sensing

The DOAS (Differential Optical Absorption Spectroscopy) instrument adds an important remote sensing capability to CARIBIC. The system is small, has no moving parts,
and analyses certain trace gases (e.g. HONO) that are otherwise not detected. This
all renders the DOAS experiment in CARIBIC interesting, if not highly relevant. The
DOAS instrument consists of two parts: a telescope block integrated in the pylon of the
inlet system, and three spectrograph-detector units located in the container instrument
(19″, 3 HU, 16 kg). Quartz fiber cables transmit the light from the telescopes to the
detectors in the container.

Each spectrograph-detector unit records spectra of scattered sun light in the UV-
visible range, from one of the respective three different viewing directions (nadir, 10°

above, and below the horizon). After each flight the recorded spectra are analyzed using the DOAS technique by which various trace gases (e.g., halogen oxides, O_3, NO_2) can be assayed by means of their individual absorption structures. The different viewing directions of the telescopes (Multi-Axis DOAS) yield additional information on the distribution of the trace gases above and below the aircraft (see also Heue, K.-P. et al. (2005)). The continuation of measurements during ascent and descent of the aircraft allow the reconstruction of vertical profiles of the respective trace gases. The sun being the source of scattered light, the DOAS technique gives data only during local daytime (can be as low as 50% of the flight time).

The three Ocean Optics USB 2000 spectrometers are custom made miniature spectrographs (with a wavelength range of 280–420 nm) which include a one-dimensional CCD-detector with 2048 pixel. They are connected to a computer and supplied with power via USB. The spectrographs are temperature stabilized using Peltier elements, an electronic controller and a Pt 100 temperature sensor. The simultaneous measurements of the three spectrograph devices are controlled by a single computer (only 10 W power consumption). Spectra are typically recorded every 30 s and stored on a flash memory card (1 Gbyte).

The telescope block ($13.5 \times 6.7 \times 1.5$ cm^3) is mounted inside the pylon of the inlet system. Incoming light (viewing angle 1.5°) is turned by 90° by quartz prisms (5×5 mm) and then focused by a quartz lens (f=15 mm) on 4 quartz fibers (200 μm) inside a cable. To reduce instrumental stray light a band pass filter (UG 5) is placed on the lens. The glass fiber cables are connected to the telescopes by 1/16″ Swagelok Ultra Torr adapters. At the telescope side the 4 quartz fibers are arranged circularly to adjust to the focus of the lens whereas at the monochromator end the fibers are set up in a line to adapt to the entrance slit of the spectrograph. During flights, the telescope block is heated with three heating pads (15 W unregulated) to avoid freezing of water on the outside surface of the prisms. Furthermore, each telescope unit is vacuum sealed to prevent condensation of water vapor. The DOAS system functions, although the light yield from the two limb channels is low. An example of NO_2 data obtained from DOAS

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measurements is shown in Fig. 14.

5 Conclusions

The first generation of CARIBIC demonstrated that valuable measurements of more than 50 atmospheric trace species including aerosol particles can be made using a freight container equipped with automated instruments onboard a commercial airliner. Regular measurement flights at a rate of one flight per month were achieved over period from the November 1997 to April 2002. The project was terminated because of the change of aircraft fleet.

The second generation of CARIBIC in ongoing operation since 2005, significantly extends the measurement capability by using improved instruments, by adding instruments for CO_2, O_2, Hg, H_2O, selected organic compounds (PTR-MS), accumulation mode particles, as well as NO_2, BrO and other atmospheric trace gases (MAX-DOAS), and of a video camera. In addition, the sampling capabilities were enhanced: the number of air and aerosol samples per flight was substantially increased and sampling of volatile organic compounds on adsorption tubes was added. The post-flight analyses of impactor samples is under improvement using PESA.

The two years of operation of CARIBIC with a new airline company, using a new Airbus A340-600, with a new container and a new sophisticated inlet system convincingly show in more than 80 individual flights that the approach of detailed monitoring of atmospheric composition by passenger aircraft, thus forming an "automated flying analytical observatory", is well feasible.

Concerning the operation of the CARIBIC system there is a need for further improvements. Foremost is the improvement in the reliability of the equipment. While the air sampler, the CO and Hg analyzers have attained a figure of merit of over 90%, some systems have not reached this level yet. We emphasize that unlike research aircraft, CARIBIC with currently 15 experiments on board has to perform unattended for days at a stretch. This requires a high degree of reliability of the equipment which means that

in the course of time improvements are necessary in equipment design, construction and electronics. We consider this as a high priority. While gaining more experience, all equipment is increasingly performing failure free, but investments in new, improved equipment is needed. Another or additional strategy could be the implementation of
a communication system, for instance real time control of equipment from the ground. However, experience shows that the most effective measure is improving equipment. By doing this, CARIBIC will become a fully operational tool.

The container concept also allows for modifications and extensions of the scientific payload. One issue is the detection of NO_2. Here the development of UV light emitting
diodes offers the possibility of adding a photolysis reactor without needing substantial amounts of added electrical power. High on the list of priorities is the measurement of SO_2. The scope of saving weight and space and minimizing power consumption, in particular by the pumping systems, together with current developments of sufficiently sensitive SO_2 systems based on chemical ionization mass spectrometry render this
tangible. Beyond this, also developments in particle mass spectrometry are promising, thus offering another important extension of analytical capabilities of the flying laboratory.

CARIBIC can contribute substantially to the research on atmospheric chemistry and climate change now and in the future by filling the gaps between satellite based re-
mote sensing, land based observatories, and research aircraft based campaigns. With the vastly improved atmospheric chemistry models available to date, systematic and detailed information will be essential for their evaluation and further development.

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Table 1. Instrumentation of the CARIBIC container onboard of the long-range Airbus A340-600 of Lufthansa.

Species	In-situ or Laboratory	Principle	Time Resolution	Institute
O_3	In-situ	UV absorption	8 s	IMK
		Chemiluminescence	0.2 s	
CO	In-situ	VUV fluorescence	2 s	MPI
Aerosol concentration	In-situ	CPC>4 nm	2 s	IFT
		CPC>12 nm	2 s	
		CPC>18 nm	2 s	
H_2O total	In-situ	Laser photoacoustic	10–90 s	IMK
H_2O gaseous	In-situ	Dew point	10–90 s	IMK
		Laser photoacoustic	10–90 s	
NO	In-situ	Chemiluminescence	10 s	DLR
NO_y	In-situ	Chemiluminescence with Au converter	10 s	DLR
CO_2	In-situ	NDIR	15 s	LSCE
O_2 with high resolution	In-situ	Electrochemical cells	15 s	University of Bern
OVOCs	In-situ	PTR-MS	20–60 s	IMK
Halogen oxides, NO_2, O_3	Remote	DOAS	30 s	University of Heidelberg
Hg	In-situ	Enrichment and atomic fluorescence	15 min	GKSS
Aerosol size distribution 150–5000 nm	In-situ	Optical particle counter	20 s	IFT
Clouds	In-situ	Video camera	0.5 s	IFT
Aerosol elemental analyses	Laboratory	Impactor, PIXE analyses	16 samples	University of Lund
Particle morphology	Laboratory	Impactor, electron microscope	16 samples	University of Lund
CO_2, CH_4, N_2O, SF_6	Laboratory	Glass canisters, GC-FID-ECD	28 samples	MPI
NMHCs	Laboratory	Glass canisters, GC-FID	28 samples	MPI
VOCs	Laboratory	Adsorption, GC-MS	16 samples	MPI
Halocarbons	Laboratory	Glass canisters, GC-MS	28 samples	University of East Anglia

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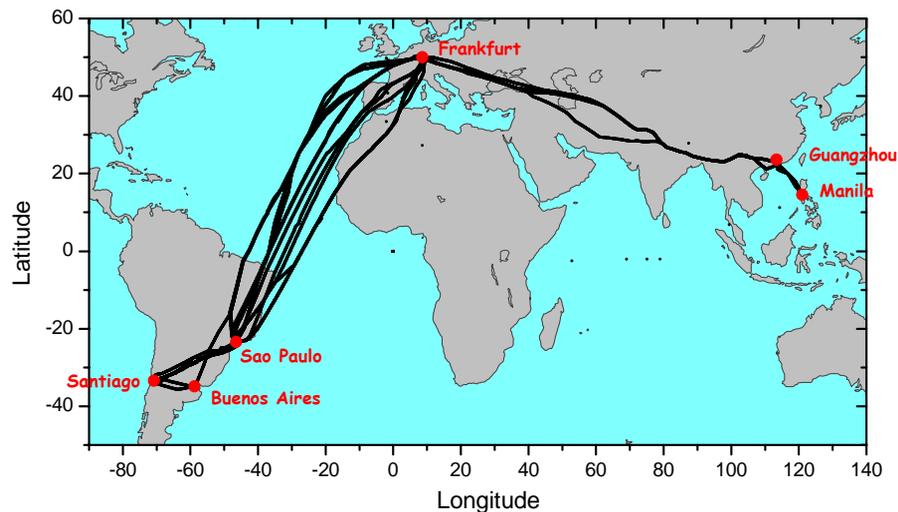
Fig. 1. Examples of flight routes to Santiago de Chile and Manila involving 4 flight legs each (brief stopovers in Rio de Janeiro or São Paulo and Guangzhou). The variations in the exact route and altitude mean that generally other air traffic has no influence on the intercepted air. For the flight levels of long range aircraft and typical routes, about 40% of the intercepted air is stratospheric.

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Fig. 2. Position of the CARIBIC container and the inlet system on the Airbus 340-600.

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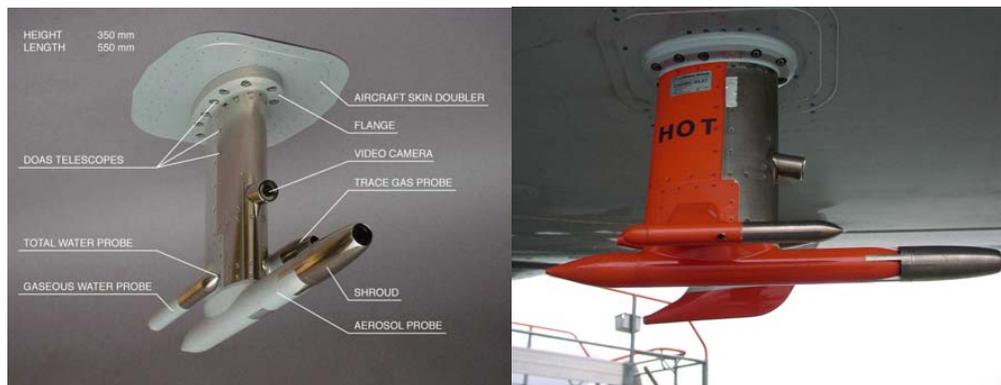
Fig. 3. Photographs of the CARIBIC inlet system.

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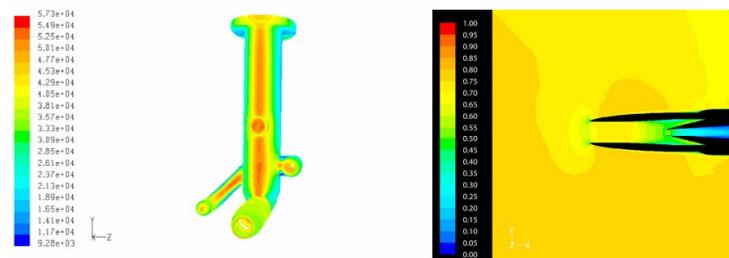
Fig. 4. Model simulations. Typical pressure distribution in hPa (left). The air speed distribution as Mach number contour plot (right)in the x-y plane cross-section of a 3-D model run for 39 000 ft altitude at a cruise speed of 0.83 M.
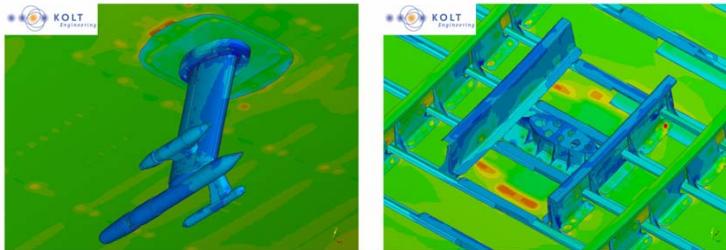
Fig. 5. A finite element based von Mise stress calculation of the combination aircraft hull and inlet system.

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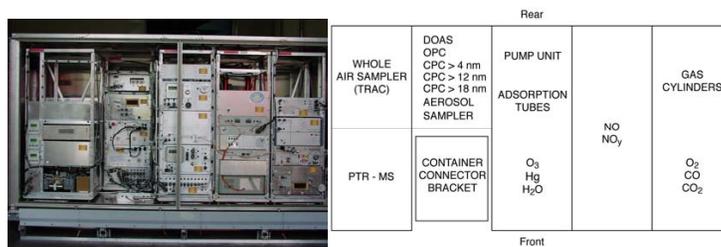
Fig. 6. Frontal photo of the container and a schematic drawing of the position of individual instruments viewed from above.
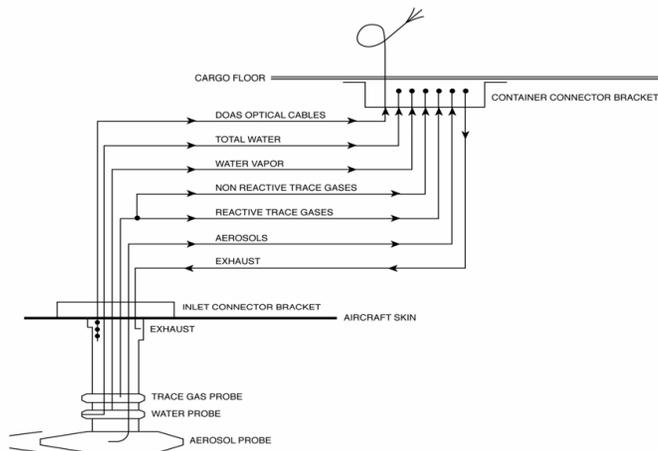
Fig. 7. Tubing connecting the inlet with the container connector bracket. The 3 optical fiber cables are continuous form inlet telescopes to the spectrographs in the container.

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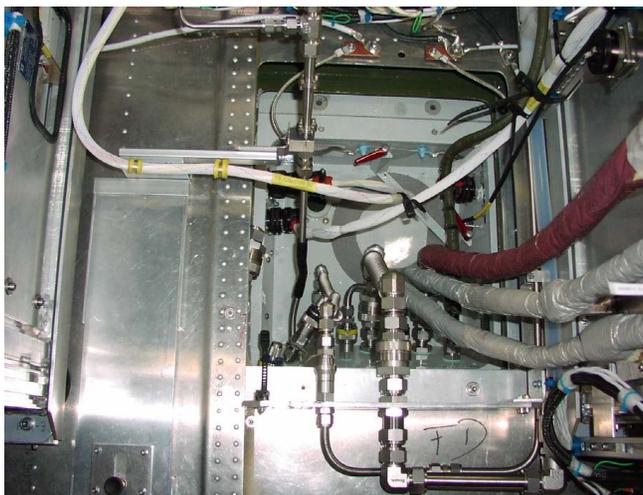
Fig. 8. Photo of the container connector bracket viewed through the cutout in the container floor.

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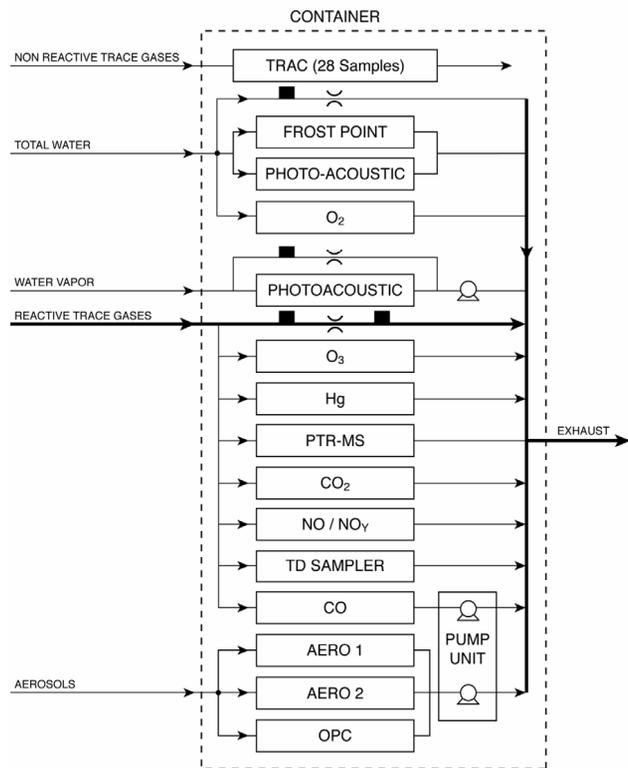
Fig. 9. Air distribution system inside the CARIBIC container. The black squares indicate pressure sensing locations. The water vapor measurements do require a suction system as there is no ram pressure on this port.
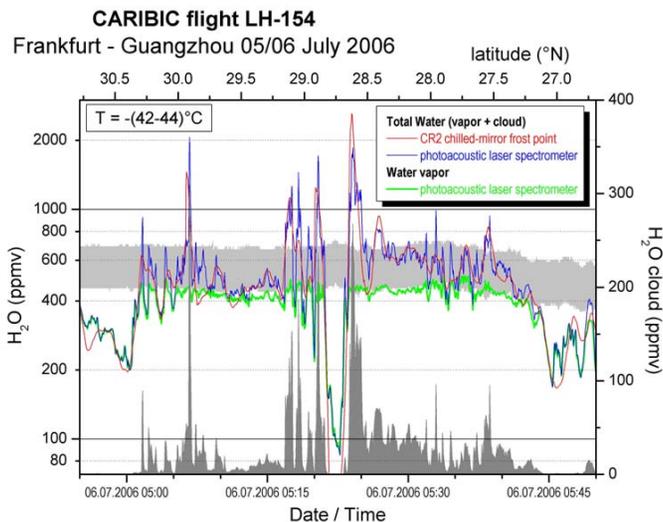
Fig. 10. Example showing the potential of both measuring gaseous water and total water using fast detectors. Detection of cirrus clouds over China at ~11.4 km pressure altitude. The grey band is the range between 100 % saturation relative to ice and water. The photo-acoustic signal (green) for water vapor reaches saturation versus ice at 05:02–05:20 and 05:23–05:42. The photo-acoustic signal for total water (blue, vapor plus ice crystals) exceeds the vapor signal during these two periods, although otherwise agreement is close. This indicates that 2 fields of cirrus clouds were intercepted. The photo-acoustic measurements are fast (<5 s response time).

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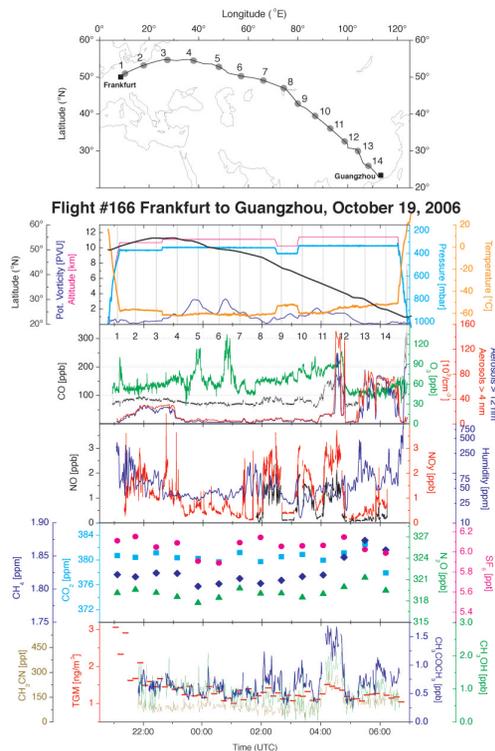
Fig. 11. Overview of the data from flight #166 from Frankfurt to Guangzhou. The route and the sites of whole air samples are shown in the uppermost panel. Flight altitude, latitude, and some meteorological parameters are given in the uppermost field of the diagram. The two fields below show O_3, CO, particle densities, NO, NOy, and humidity data. The last but one field contains the results of analyses of whole air samples for CH_4, CO_2, N_2O and SF_6. Data on acetone, acetonitrile, methanol, and total gaseous mercury (TGM) are shown in the lowermost field. We refer to the KNMI website (http://www.knmi.nl/samenw/campaign\support/CARIBIC/#LH) for detailed meteorological information.

Flight #116 Santiago de Chile to Sao Paulo, 12. Juni, 2005
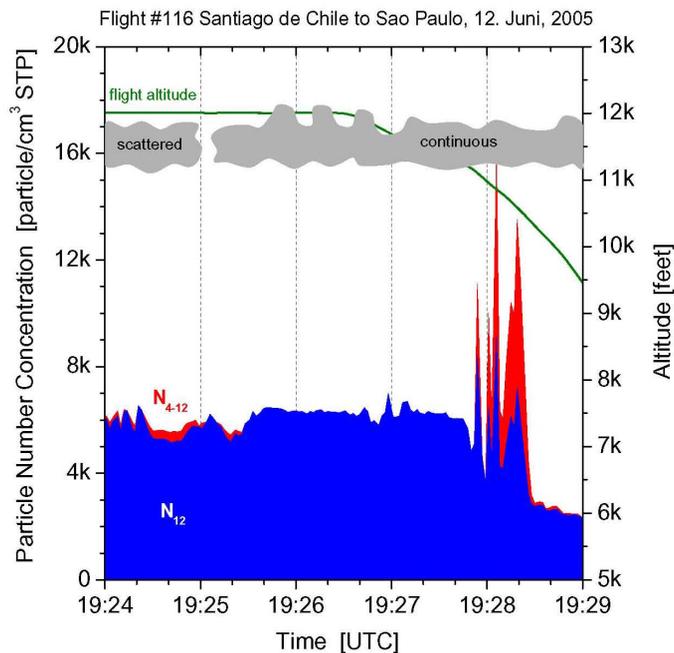
Fig. 12. The occurrence of ultra fine particles (between 4 and 12 nm) was recorded just after descending through clouds (but not inside the clouds) on approaching São Paulo Airport. The use of the camera allows the important conclusion that fragments of cloud droplets or ice particles impinging on the leading edge of the shroud and aerosol diffuser tube do not enhance particle counts. The origin of the ultrafine particles below the cloud cover is a question to be answered using all combined data.

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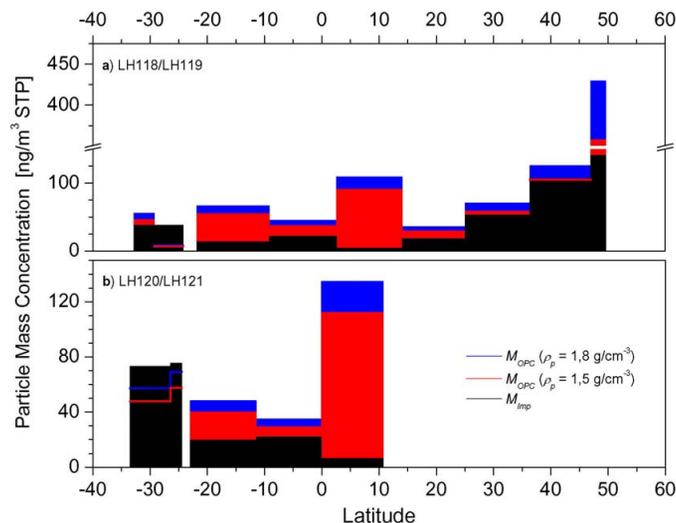
Fig. 13. Aerosol mass as calculated for the OPC measurements assuming spheric particles compared with aerosol mass inferred from the PIXE analyses assuming the aerosol mass is sulfate. These measurements were made during cross Atlantic flights (http://www.knmi.nl/ samenw/campaign\support/CARIBIC/#LH). Considering the vastly different techniques used, the agreement at mid latitudes is encouraging. In the equatorial region, the OPC data indicate substantially higher aerosol masses. A plausible explanation is that this missing mass is carbon, which is not detected by PIXE but can be detected in the future using PESA in Lund.

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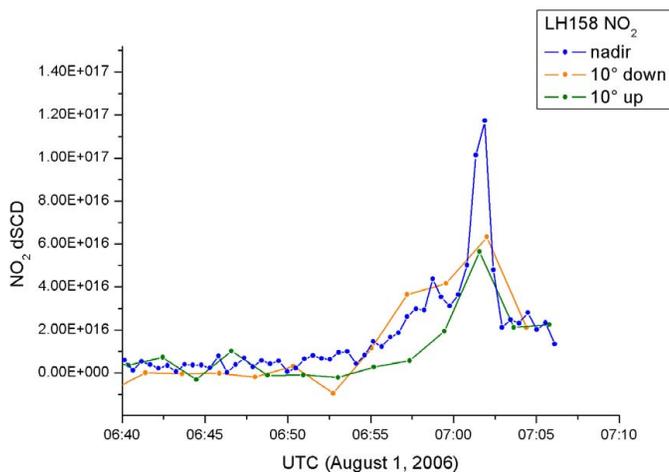
Fig. 14. A typical DOAS result upon landing in Guangzhou, China. All viewing directions show a rising slant column of NO_2 at 06:55 UTC, when the airplane is just inside the boundary layer at a height of about 2.3 km. The different sensitivities of the viewing directions can be used to model the light paths and the aerosol load inside the boundary layer in order to derive the local NO_2 concentration profile.