

CARIBIC—Civil Aircraft for Global Measurement of Trace Gases and Aerosols in the Tropopause Region

C. A. M. BRENNINKMEIJER,* P. J. CRUTZEN,* H. FISCHER,+ H. GÜSTEN,+ W. HANS,# G. HEINRICH,+
J. HEINTZENBERG,@ M. HERMANN,@ T. IMMELMANN,& D. KERSTING,# M. MAISS,* M. NOLLE,+
A. PITSCHIEDER,& H. POHLKAMP,& D. SCHARFFE,* K. SPECHT,& AND A. WIEDENSOHLER@

* *Max-Planck-Institut für Chemie, Abteilung Luftchemie, Mainz, Germany*

+ *Institut für Meteorologie und Klimaforschung, Forschungszentrum Karlsruhe,
Universität Karlsruhe, Karlsruhe, Germany*

Gesellschaft für angewandte Systemtechnik, Friedrichshafen, Germany

@ *Institut für Troposphärenforschung, Leipzig, Germany*

& *LTU International Airways, Dusseldorf, Germany*

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ABSTRACT

The deployment of measurement equipment in passenger aircraft for the observation of atmospheric trace constituents is described. The package of automated instruments that is installed in a one-ton-capacity aircraft freight container positioned in the forward cargo bay of a Boeing 767 ER can register a vast amount of atmospheric data during regular long-distance flights. The air inlet system that is mounted on the fuselage directly below the container comprises an aerosol inlet, a separate inlet for trace-gas sampling, and an air exhaust. All instruments, the central computer, and power supply are mounted in aviation-approved racks that slide into the reinforced container. The current instrument package comprises a fast-response chemiluminescence sensor and a conventional UV absorption detector for O₃; a gas chromatograph for CO; two condensation nuclei counters for particles larger than 5 and 12 nm; and a 12-canister large-capacity whole air sampler for laboratory trace-gas analysis and isotopic analysis of CO₂, CO, CH₄, and N₂O. First measurement results of the operational Civil Aircraft for Remote Sensing and In Situ Measurements in Troposphere and Lower Stratosphere Based on the Instrumentation Container Concept (CARIBIC) container are reported.

1. Introduction

Despite concern about environmental consequences of emissions from the growing fleet of aircraft, reliable impact assessments are not yet possible due to our deficient knowledge of details of physical and chemical processes in the upper troposphere and lower stratosphere and their interaction (Schumann 1994). Clearly, not only aircraft emissions affect the chemistry in the region of the tropopause. Other anthropogenic activities, including the large-scale burning of forest and savanna, also can bring about widespread profound chemical changes in the upper troposphere and lower stratosphere. Also the uplifting of polluted surface air into the upper troposphere affects the region's chemistry. However, compared to the planetary boundary layer where most atmospheric measurements takes place, the upper troposphere is extremely poorly investigated for the following reasons: 1) the tropopause is distant from

our daily living environment, 2) the high cost of atmospheric research aircraft flights and satellites, and 3) the inherent limitations of remote sensing capabilities.

Although the available data in combination with comprehensive three-dimensional chemical-transport models (CTMs) provide a synthetic picture of our current knowledge of the dynamics and chemistry of the tropopause region, considerable uncertainties remain (Mahlman 1997; Prather 1994; Holton et al. 1995). In particular, to completely understand the impact of changing chemistry and aerosols on the climate system (Andreae and Crutzen 1997), it is necessary to obtain more information about the ozone distribution and about the physical and chemical properties of aerosols in the submicrometer range in the tropopause region. Because fine aerosols control the number and surface distribution of cloud droplets, they influence cloud radiative properties, thereby possibly leading to an indirect radiative forcing of climate. Furthermore, the available droplet surface controls heterogeneous chemistry and therefore, among other things, influences the concentration of ozone and the oxidizing capacity.

The use of commercial airliners enables us to better observe the spatial and temporal distribution of relevant

Corresponding author address: Dr. Carl A. Brenninkmeijer, Max-Planck-Institut für Chemie, Atmospheric Chemistry Division, P.O. Box 3060, D-55020 Mainz, Germany.
E-mail: carlb@mpch-mainz.mpg.de

species than irregular flights on research aircraft (Fleming 1996), and importantly, it may be more cost effective. Despite confirmation of the feasibility of this concept after the GASP project (Perkins and Papathakos 1978; Nastrom 1979), the development stagnated. The need to make scientific equipment designed for laboratory use comply to the strict safety regulations of commercial airliners has played a possible role in this delay.

Continuing, if not growing, concern about aircraft emissions and their effect on ozone concentrations as well as their possible climate forcing has renewed the interest in the use of commercial airliner as platform for analytical instrumentation. In this decade new programs such as MOZAIC (Measurement of Ozone and Water Vapor by Airbus In-service Aircraft) (Marengo et al. 1997), JAL (Foundation Japan Airline) (Matsueda and Inoue 1996), and NOXAR (Measurement of Nitrogen Oxides and Ozone along Air Routes) (Brunner et al. 1994) were initiated. MOZAIC uses several aircraft for building up an impressive climatology for O_3 and H_2O . JAL uses one aircraft equipped with an ingenious flask sampling system. For these two projects, the scientific equipment has become to some degree a permanent fixture on the aircraft. NOXAR also involved one aircraft that carries an NO and NO_2 analyzer, demonstrating the feasibility of some rather difficult measurements using passenger aircraft.

Here we present CARIBIC (Civil Aircraft for Remote Sensing and In Situ Measurements in the Troposphere and Lower Stratosphere Based on the Instrumentation Container Concept), that is, an automated instrument container that offers the capability of deploying a broad range of equipment. The development from the concept to the first measurement flights has taken approximately 4 years, involving collaboration by scientific institutes, an aircraft system engineering company, the airline, the manufacturer of the aircraft, and the aviation authorities. This paper is not an exhaustive technical document, but rather intended to communicate the main technical and practical aspects and findings of this exciting use of passenger aircraft in atmospheric chemistry.

2. CARIBIC container system description

The application of a standard aircraft container for holding all scientific measurement and supporting equipment is attractive. Unloading of the equipment for the purpose of servicing and calibration for air sample retrieval, or simply for waiting for the aircraft to be routed to desired destinations, can be performed almost as routinely as the procedure for unloading cargo. It is essential not to disturb the routine operation of the aircraft. In addition, the container approach has the significant advantage of ample space for bulky instruments. Finally, there is increased flexibility as to what equipment package is to be deployed. Obviously, modifications and various equipment configurations all have to

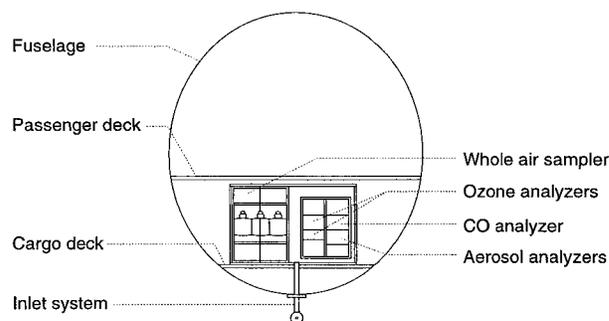


FIG. 1. Cross-sectional view of the LTU Boeing 767 300ER registered as D-AMUN with measurement container and inlet system.

be approved by the national aviation authorities (German LBA).

The 200-ft³ (5.5 m³) luggage/freight container has a payload capacity of about one ton. The aircraft used is a Boeing 767 300ER, which is owned and operated by the German International Airways LTU (Lufttransport Unternehmen, Dusseldorf, Germany), one of the world's larger charter airline companies. Space for the CARIBIC container is in the forward cargo bay directly against the bulkhead (Fig. 1). A custom-built inlet system (Fig. 2) with a heated inlet cone is mounted directly underneath the container outside the fuselage, approximately 10 m from the front of the aircraft. The mounting flange of the inlet assembly is connected via a box construction to the ribs according to specifications from Boeing. The mast of the inlet system is 25 cm tall, sufficient for both gas and aerosol sampling inlet tubes to be outside of the aircraft's boundary layer. Waste air is expelled through slits in the rear of the mast close to the skin. The air intake for CO analysis and air sample collection is a 125-mm-long, 12.5-mm interior diameter tube, from which air is drawn perpendicular via identical tubing. This air inlet tube can be sealed off at the upstream side, by which less particulate matter is entrained; however, this is at the cost of losing some pressure. Alternatively, the tube can be sealed at the downstream end, increasing the inlet pressure at the cost of increased contamination. Currently, the inlet tube is left open at both ends. Air for the ozone analyzer is drawn from the aerosol tube via a Teflon-coated 6.0-mm tube.

The design of the aerosol intake tube for particle counting and collection is critical. This tube is also mounted at a fixed pitch parallel to the aircraft length axis and is provided with an inlet cone with an orifice diameter of 4.0 mm. The air expands into the wider inlet tube section with an internal diameter of 44.5 mm, by which it is slowed down. Sample air is drawn at a rate of several liters per minute (STP) from the rear cylindrical section through a 6.0-mm tube. The configuration is suitable for the collection of small particles, specifically submicron particles. Larger particles are not entrained into the sampled air due to their larger inertia.

The collection of aerosol using a fast-flying aircraft

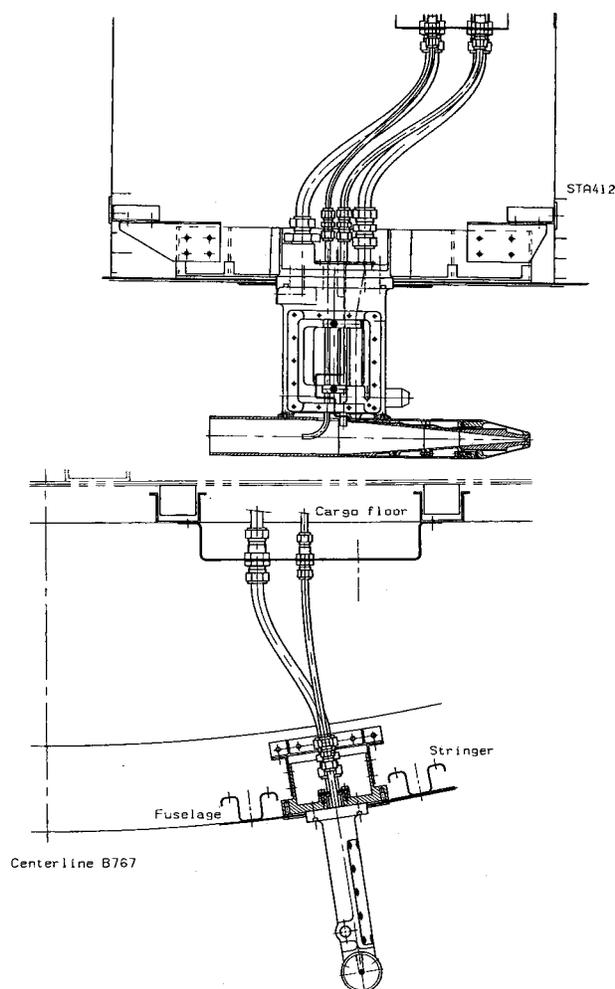


FIG. 2. Cross-sectional views of the inlet system. The system is connected to the aircraft via a box construction, which is attached to the ribs. Stainless steel tubing connects the air inlets and the waste air outlet to the couplings mounted in a depression in the floor of the cargo bay. The larger lower section is the aerosol air intake in which air to be sampled is slowed down by expansion. The tube has a heated front cone to prevent icing. Air for aerosol counting, and the planned aerosol collection and size distribution analysis, is withdrawn via the curved tube. Air for the ozone analyzers is drawn from the same air flow at a rate that is negligible compared to the overall throughput. Above the aerosol inlet is the 12.5-mm inlet for air sample collection and CO analysis. Waste air from the air sampling flow through the equipment is vented at the rear end of the mast close to the skin of the aircraft.

(about 235 m s^{-1}) without modification of the particles or incurring losses is difficult, and certain compromises have to be made. Because the airflow through the analyzer is only a few meters per second, the air has to be slowed down. By using a gradually widening inlet tube, turbulence is minimized. However, because total length of the inlet tube is limited by various requirements of modifications to the aircraft, turbulence cannot be eliminated. Furthermore, the slowing down of air causes heating, and particles may be affected. Therefore,

a dummy of the inlet system has been tested extensively in the laboratory, results of which will be published separately. The inlet system is connected to the inlet ports that are fixed just above the bottom plate inside the container via flexible hoses using quick-connectors.

The measurement and sampling apparatus, the central computer, pumping unit, and the main power supply are mounted in aviation-approved racks. The left half of the container is occupied by the whole air sampler, which can be pulled away from its position for transportation to the laboratory for sample analysis. Aircraft 115 V, 400-Hz power is converted to 28 V DC by the main power supply (Type 6060, Aircraft Electronic Engineering GmbH, Seefeld, Germany). Thus, electronics and pumps all use DC, and frequent use is made of DC-DC converters. These converters of the HF switching type provide galvanic separation, are small and compact, have efficiencies above 85%, and are used as on/off switches for the units.

The central computer communicates with the various units via RS-type serial ports. Equipment switch-on commands are given in sequence when the aircraft is at a safe distance from the frequently highly polluted airport environment. Partly because the analytical equipment has been developed at different institutions, but mainly because the measurement data recorded during the flight are only raw data, most units have their own controller and have separate data storage on solid-state memory cards.

Two issues have proven to be rather troublesome. One issue is fluctuations of power supplied to the container caused by switching from ground to aircraft power when leaving the terminal position or other power interruptions. The power irregularities have caused erratic behavior of some equipment, which was remedied by power buffering of most controllers. The other issue is that testing under flight conditions is not as straightforward as with research aircraft. During flights there is no access to the cargo hold. This has extended the "teething problem" period. To obtain the required flight parameters, an ARINC 429 (Aeronautical Radio Incorporated) interface with two channels is connected to the ARINC buses of the aircraft data system. Parameters recorded by the central computer are date, time, altitude, latitude, longitude, ground speed, temperature and pressure, wind direction, and velocity.

Overriding all issues involved in the container project is the one of safety of passengers and crew, and the aircraft as a whole. In this respect, an intrinsic advantage of the container concept is that the container is placed in a cargo bay and that it is not a permanent fixture to the aircraft. Nevertheless, the requirements for mechanical integrity, for electrical noninterference, and for fire risk safety are still extremely important. For obtaining the required mechanical stability, the use of approved materials, structures, and calculations appears not to form a great difficulty. For excluding electromagnetic interference, equipment is tested separately and in var-

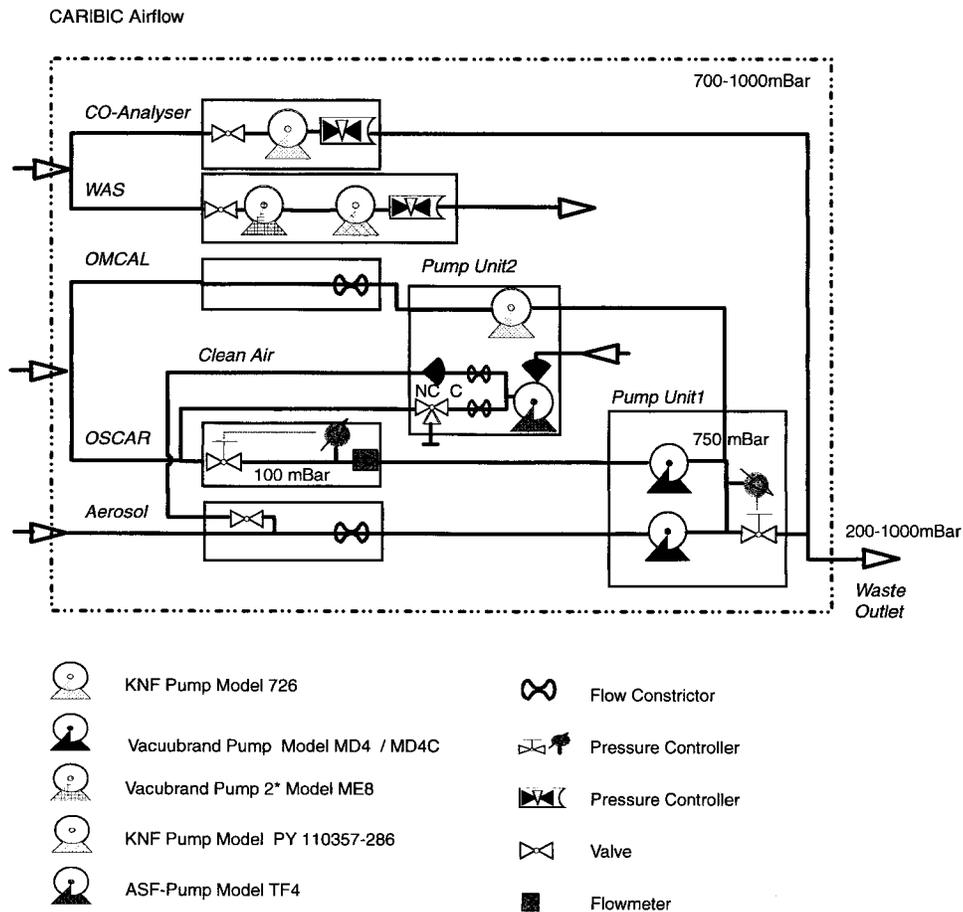


FIG. 3. Schematic of equipment and airflows. The pressure in the cargo bay ranges from about 0.7 to 1 bar and the intake pressure from 0.2 to 1 bar. A major distinction in airflow is that for the CO analyzer and air sampler, compressors and pumps are at the inlet side. For the other analyzers, air is drawn through the equipment by downstream pumps. The ozone analyzer OSCAR and the aerosol unit can be flushed with filtered clean air to prevent contamination when the aircraft is in the vicinity of the often rather polluted airports.

ious configurations. Proper shielding and grounding generally prevented problems of in this respect. All equipment complies with the international aviation requirements RTCA 160C.

To preclude any harm from fire, combustible materials, as well as those that can produce toxic fumes, are banned in the equipment to a high degree. We note that compared to a freight container loaded with a broad mix of passenger's suitcases, scientific equipment as required for research purposes offers little flammable matter. Nonetheless, specifically the measurement container is connected to electrical power, potentially increasing fire risk. As a countermeasure the cooling air of all units is ducted past smoke detectors. The control board for the container in the cockpit incorporates an overheat and smoke indicator/alarm and a main switch.

3. Instrumentation

The current instrument package (Fig. 3) consists of a fast-response chemiluminescence ozone sensor as well

as a conventional ozone monitor, a gas chromatograph for CO analysis, two condensation nuclei counters for submicron particles larger than 5 and 12 nm, and the 12 canister large-capacity whole air sampler for post-flight concentration and isotopic measurement in the laboratory. With the exception of the fast-response ozone sensor and the whole air sampler, all apparatus are commercially available laboratory instruments that have undergone varying degrees of modifications and improvements to operate in the aircraft. A central technical problem is the reduced inlet pressure of around 200 mb. Leakage of air from the cargo space, which is kept in equilibrium pressure with the passenger deck at about 850 mb, into the various apparatuses has to be prevented. Furthermore, all equipment has to be vibration proof and withstand up to 9g acceleration. This requires sturdy rack constructions and additional fixtures for printed circuit boards inside electronic units. Operating temperatures are between 0° and 35°C but are typically between 10° and 15°C. Upon opening the cargo

doors at airports in the Tropics, condensation problems can occur. Even though the container has ample capacity, we have tried to reduce weight as far as possible. A total of near one ton is the result, of which a large part of 300 kg is occupied by the whole air sampler. The original tarpaulin type of front cover of the container has been replaced with aluminum doors, giving mechanical and electrical containment.

The choice of the current equipment is mainly to be able to

- 1) measure the classical duo O_3 and CO (Fishman and Crutzen 1978),
- 2) measure for the first time using passenger aircraft aerosol abundance, and
- 3) collect large air samples for trace-gas isotope as well as concentration analysis.

Only a freight container in a larger aircraft can contain this package. We will not dwell on details about the specific scientific questions that can be addressed by data obtained with this equipment. With each instrument described below we briefly mention the most salient scientific aspects of the data only.

a. Ozone analyzers

Aircraft emissions, in particular NO_x , in the upper troposphere play a major role in the photochemical production of ozone. As shown by Hidalgo and Crutzen (1977) and Johnston et al. (1989) in model calculations, excess ozone can be produced in that region from aircraft NO_x (Brasseur et al. 1996). It is of special interest to understand the ozone production because the upper troposphere is the region in which ozone is most effective as a greenhouse gas (Lacis et al. 1990). Furthermore, ozone is a frequently used tracer for stratospheric air masses.

With the fast-response ozone sensor in the CARIBIC container, we can obtain detailed horizontal ozone profiles at the cruising altitude in the tropopause region with a high spatial resolution. At a cruising speed of close to 250 m s^{-1} of a passenger jet, the ozone sensor has to be fast. On the basis of a previously developed fast-response ozone sensor (Güsten et al. 1992; Güsten and Heinrich 1996) for eddy covariance measurements, the current ozone sensor OSCAR (Ozone Sensor CARIBIC) was developed. The time resolution of OSCAR reaches up to 10 Hz. For the first test flights it was set to 1 Hz. (Details of OSCAR will be published separately.) Since the detection principle of OSCAR is chemiluminescence of an organic dye adsorbed on silica gel in the reaction with ozone, which has a variable response, this sensor is continuously calibrated in flight against the UV absorption ozone analyzer.

For this purpose a conventional ozone analyzer (Environnement 03 41M, Paris, France) has been modified. The 28 V DC from the power supply is fed into the instrument as 24 V DC via a DC-DC converter. Also

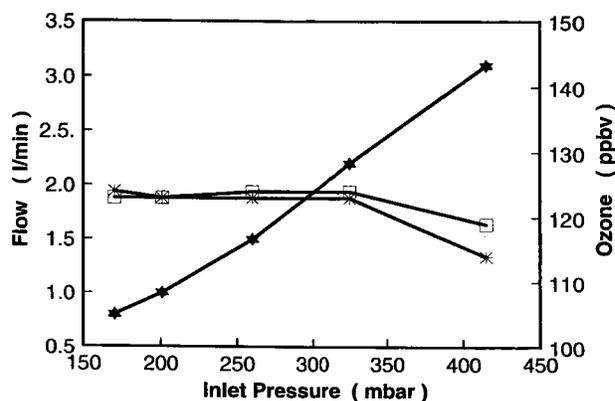


FIG. 4. Calibration results for the ozone analyzer OMCAL showing a near linear drop in flow rate from about 3 to 1 L min^{-1} (at 200 mb) with decreasing intake pressure. The ozone mixing ratio at the inlet, as measured at near 1000 mb with an ozone transfer standard, is given as ppbv by the open squares. The response of OMCAL is given by the asterisks.

other electrical parts were changed from AC to DC, including the pump. The air to be analyzed is drawn through the optical cells to avoid having to compress the air at the inlet with the risk of affecting its ozone content. The constriction originally present in the unit for limiting the air flow has been removed. Since ozone analyzers of this type are UV absorption photometers designed to operate as surface instruments, the internal correction for pressure and temperature, normally operative down to pressures of 500 mb only, had to be extended. Hence, the software was changed by reprogramming the EPROM.

The remodeled instrument called OMCAL (Ozone Monitor Calibrator) operates automatically down to pressures below 200 mb with a time resolution of 10 s. OMCAL was thoroughly tested in the laboratory using low inlet pressures and a range of ozone concentrations, which were calibrated against an ozone transfer standard. A calibration was also performed using an 80 m^3 chamber with various constant ozone volume fractions. Furthermore, OMCAL was tested using a dynamic setup in combination with another ozone analyzer recording the inlet air ozone concentration at ambient pressure. Figure 4 shows that the dropoff in the air flow through the instrument due to decreasing pressure has little effect on the recorded ozone values.

b. The carbon monoxide analyzer

Carbon monoxide is one of the most widely distributed air pollutants and a known precursor for tropospheric ozone (Fishman and Crutzen 1978). Except for CO_2 , more CO is released into the atmosphere from anthropogenic and natural sources than any other pollutant. Most of our knowledge about the distribution of CO in the atmosphere is based on ground-based observations, for which NOAA/CMDL in Boulder, Colorado,

has an extensive global network. No systematic detailed information is available for the free troposphere, which, for a trace gas like CO with a lifetime that allows vertical gradients and with many surface sources, is a serious deficiency. Some remote sensing data for CO from the ground are available, and currently satellites are being prepared with CO monitoring capabilities. Comparison with the CARIBIC data most certainly will be useful.

For convected polluted surface air we expect to measure enhanced CO. For transects of the aircraft intercepting the stratosphere, we expect distinctly increased ozone values, coupled with low CO of near 30 nmol mol^{-1} . However, at times CO is transported at high concentrations into the stratosphere as was reported by Bregman et al. (1997). This was a demonstration not only of local cross-tropopause transport but also of the vast amounts of CO produced by biomass burning. On tropical destinations, the widespread pollution from biomass burning, as, for instance, observed from a space shuttle (Reichle and Connors 1999), should be detectable.

The CO analyzer is a modified commercial gas chromatograph (RGA3, Trace Analytical, Menlo Park, California). The actual CO detection is based on the reaction of CO with hot mercuric oxide to produce CO_2 and mercury vapor, $\text{CO} + \text{HgO (solid)} \rightarrow \text{CO}_2 + \text{Hg(gas)}$, which is sensed by its strong light absorption from a small low-pressure mercury vapor lamp at 254 nm (Seiler and Junge 1967). Minute amounts of mercury are produced and trapped by a scrubber. Prior to detection, the CO is separated on a gas chromatography column (3 m, 1/8", 13X molecular sieve). The detection limit is well below 3 nmol mol^{-1} , which is sufficient to gauge CO in clean remote air masses. The accuracy and precision are about 3%. Comparisons with the results obtained for the laboratory analyses of the whole air samples allow for an additional quality control.

The commercial CO analyzer virtually had to be rebuilt. Electrical parts such as pumps, heating elements for detector and column oven, fan, etc., were replaced by the appropriate 24-V units to be powered by a 28/24 V DC-DC converter. The UV discharge lamp of the analyzer operates on 115 V AC, generated by its own DC/AC 24/115 V AC converter. A more powerful inlet pump was mounted on shock absorbers inside the unit. Small compressed air cylinders with the different calibration mixtures for in-flight calibration are mounted in a separate box. The CO analyzer, which operates at a pressure of 1 bar, was tested in the laboratory down to an inlet pressure of about 100 mb. The measurement cycle comprises four sample injections, sandwiched by single injections of two CO standards of nominally 90 and $150 \text{ nmol mol}^{-1}$. The obtainable time resolution is, in principle, determined by the volume of the air sampling loop and is of the order of a second or less. However, the elution of the CO peak from the chromatography column takes about 3 min. Therefore, the effective time resolution is about 3 min. Care is taken to cross-

check CO calibration with NOAA/CMDL standards. The analyzer requires a warmup period, which makes it necessary to activate part of it well before the 500-mb level is reached, at which point the measurements start. Corrections for nonlinearity of the detector response are also based on occasional laboratory tests.

c. Aerosol analyzers

Global chemical transport models (CTMs) in the atmospheric compartment reaching from the lower troposphere to the lower stratosphere are beginning to carry aerosol information, primarily as mass concentration of sulfate and biomass burning aerosols. The long-range transport of these mostly anthropogenic aerosols chiefly takes place in the free troposphere. On the scale of current CTMs there is practically no information on the chemical composition and mass concentration of aerosols in the tropopause region that could be used to validate these models. It is further desirable to develop chemical fingerprints of aircraft emissions from multi-element aerosol analyses. This information is needed to obtain information both on the distribution of natural background aerosols and the particulate matter produced by aircraft. In tropical regions, smoke from biomass burning has been observed at altitudes up to the cruising level of commercial aircraft. On few research flights both gaseous tracers (Kley et al. 1996) and particle samples (Ikegami et al. 1994) have given clear indications of strong vertical transport from the surface to the tropopause region in the Tropics.

Two standard condensation particle counters (CPCs) based on the detection of small particles after being increased in size by condensation (TSI Model 7610, TSI, St. Paul, Minnesota) were modified for low-pressure operation. About 10 L min^{-1} of air at ambient pressure is drawn from the inlet to minimize aerosol losses. The particles are counted in a smaller flow of air of approximately 1.5 L min^{-1} by a light scattering sensor after condensation in a saturated butanol vapor. The counters were made vacuum-tight and combustible plastic components were replaced by Teflon or metal. The airflow is entirely in a closed system, and waste air is vented to the outside. Various DC voltages are generated using the DC-DC converters. An industrial 486 computer controls the rack unit temperatures, the saturation and condensation temperatures, the inlet pressure, and data collection.

The modified CPCs were thoroughly tested and calibrated at pressures ranging from 1000 to 160 mb. Down to 300 mb, the results obtained in the laboratory show a better counting performance at reduced pressures compared to standard pressure. In Fig. 5 the pressure dependence of the counting efficiency is shown for a temperature differential of 17° between saturator and condenser. For detecting the 5-nm particles a temperature differential of 27° is used. A preliminary report on the calibration procedure is given by Hermann and Wie-

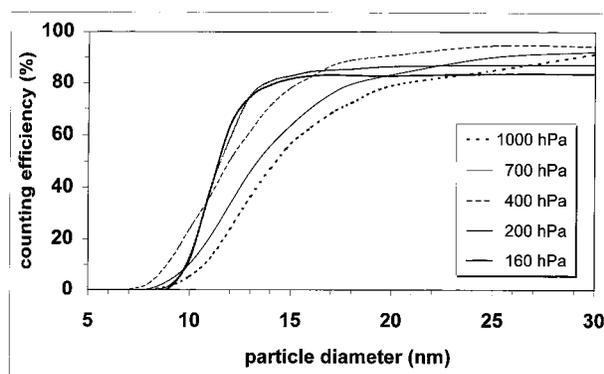


FIG. 5. Particle detection efficiency of the modified CPC TSI model 3760/7610 for five pressure regimes as a function of particle diameter, using a temperature differential of 17° . The upper cutoff for particle size is determined by the inlet system at approximately $2 \mu\text{m}$. The detection efficiency over the range up to $2 \mu\text{m}$ is constant.

densohler (1996) and Hermann and Wiedensohler (1998). The modified CPC has a detection range of 1–20 000 particles per cubic centimeter with a (software determined) time resolution of 4 s.

d. The whole air sampler

For trace gases that are sufficiently stable, air sampling offers the possibility to measure a wide spectrum of trace-gas concentrations, thus providing detailed information about the origin, mixing, and concomitant chemical modification of air masses. Using regular flights, for the first time seasonal cycles for gases like CO_2 , CH_4 , N_2O , and others can be established for the free troposphere. Chemically active species, like various nonmethane hydrocarbons with a relative short lifetime against OH, can be used to indicate rapid convection of surface air; SF_6 also can show this when its specific industrial sources are active, but otherwise this very long-lived gas serves as a tracer of long-range transport (Maiss et al. 1996; Denning et al. 1997, manuscript submitted to *Tellus*).

Given the space and capacity of the container, sampling of large quantities of air sufficient for isotopic analysis has become feasible, and, consequently, the current whole air sampler comprises tanks and compressors capable of sampling 350 L air per event. Because details of the whole air sampler will not be published elsewhere, and isotopic analysis is a rather specialized activity in atmospheric chemistry, we give some more information here.

The isotopic analyses of atmospheric trace gases, predominantly CH_4 , CO , CO_2 , and N_2O , are used for better understanding the origin and cycles of these main trace gases. In particular, isotopic analysis of CO , in which ^{14}C is an excellent tracer for stratospheric air (Brenninkmeijer et al. 1995) and ^{17}O and ^{18}O are useful indicators of the exposure to OH (Röckmann et al. 1998a), gives new insights. Isotopic assay is performed in the

laboratory after cryogenic trace-gas separation, followed by stable isotope mass spectrometry or accelerator mass spectrometry in case of ^{14}C (Brenninkmeijer 1993). Aircraft-based air sampling at high altitude and low pressures represents a modest technical problem. It is important to minimize the collection time of several hundred liters of air (Brenninkmeijer and Roberts 1994) to obtain a reasonable spatial resolution. The minimum sample size, on the other hand, is prescribed by the counting statistics of ^{14}C ions and the overall system efficiency in ^{14}C accelerator mass spectrometry; ^{14}CO has an exceedingly low abundance of only 5–150 molecules per cubic centimeter.

Combined with the trace-gas concentration measurements, for each sample over 50 concentration and isotope ratio numbers are obtainable. The added benefit of the large sample size is an increased stability of many of the trace gases collected due to an improved sample size-to-surface area ratio. The pumping system contains a trap at the high pressure side that can be filled with a compound to scavenge O_3 . Some problems have been experienced by elevated O_3 in sample air collected in the stratosphere. Because O_3 is isotopically strongly enriched, even the production of a few ppbv of CO in the air sample cylinders or pumps due to the reaction of O_3 with organic compounds (Röckmann et al. 1998b), can corrupt the ^{18}O content of CO samples collected in the stratosphere. The problem is currently being addressed with by using the chemical scrubbing of O_3 . No drying of the air prior to storage in the cylinders is applied.

The whole air sampler (Fig. 6) designed for collecting the large air samples of 350 L incorporates two diaphragm pumps (Type ME8, Vacuubrand, Wertheim, Germany) to initially boost the pressure from about 200 mb to just over 1 bar. For this, the total of eight cylinder heads are configured in parallel, using a manifold of 0.25-in. thin-walled stainless steel tubing. Subsequently, the sample air is fed into a two-stage diaphragm compressor with interstage cooling (Type PJ11035-286, KNF Neuberger, Freiburg, Germany) to obtain a final pressure of 17 bar within 20 min. The compressor has a bypass valve to short circuit inlet and outlet for minimizing resistance during the start up. It appears that this particular combination of pumps and compressor leads to a nearly flat pumping rate, independent of the instantaneous pressure in the recipient. The advantage of this arrangement is that each sample represents the 250 km that are covered during the roughly 20 min the filling requires without spatial bias.

Twelve stainless steel cylinders (Crown Sheet Metal, Invercargill, New Zealand) are used. These cylinders, which are originally intended as lightweight LPG storage tanks, are custom made for scientific air sampling using bud welding to provide a smooth inner surface (Lowe et al. 1991). Cleaning and conditioning of the cylinders for minimizing the growth or destruction of trace gases was achieved by steam cleaning them inside an oven using some distilled water. In flight, the cyl-

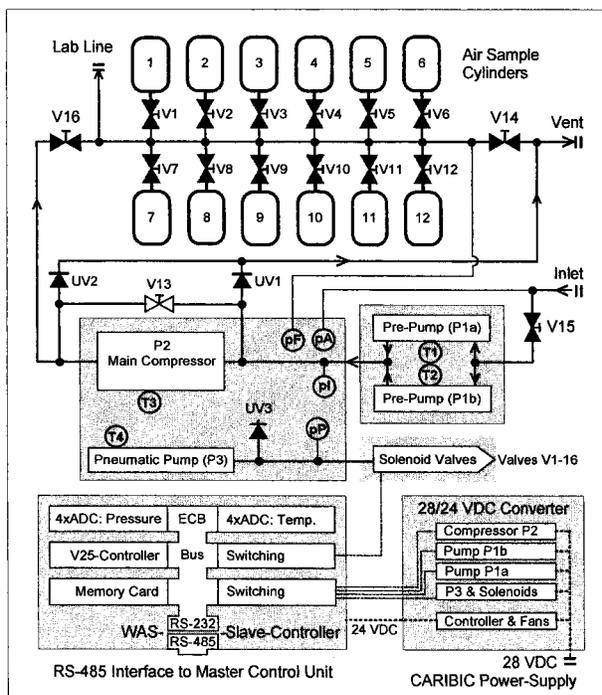


FIG. 6. Schematic of the whole air sampler. The 12 stainless steel air sample cylinders of 21-L water volume are connected via compressed air actuated valves to the manifold. The diaphragm vacuum prepumps boost the pressure of the air sampled via the $\frac{1}{2}$ -in. valve V15 to 1 bar. The main compressor can be shunted by V13 for eliminating a pressure gradient during its startup. The two mechanical pressure relief valves (UV1 and UV2) prevent overpressure at the outlet of the prepumps and main compressor, respectively. A small diaphragm pump P3 supplies compressed air at 6 bar for opening the air actuated valves. For supplying air to the air actuated cylinder inlet valves, miniature 0.65-W solenoids (Clippard, Ohio, type EM) valves are operated by the electronic controller.

inders are filled sequentially after flushing by repeated pressurization to typically 3 bar and purging to 1 bar. It is foreseen that filling can be triggered to capture air masses that are considered of special importance for analysis. Passive and active components protect the system against overheating and overpressure. Temperature of the cylinder heads is gauged as potentially critical for corruption of the trace-gas composition due to chemical reactions. These data and filling details are stored on a PCMCIA card.

The collection of samples with the whole air sampler takes place as a rule during the return voyage. This praxis reduces the time for which sample air is stored prior to analysis. Moreover, for flights to the Tropics, the return leg reaches into the stratosphere, which is not the case for the southbound leg. For air sampling, the entire whole air sampler is transported to the laboratory. In this way contamination and the risk of introducing leaks by decoupling and coupling air cylinders is excluded.

4. First measurement results

Because cargo space is often available for tourist flights from Munich to the Republic of the Maldives in the Indian Ocean, Male (4°N , 74°E) has become one of CARIBIC's current frequent-flyer destinations. This route is of special interest because the Tropics are reached and on the return flight often the lowermost stratosphere is intercepted. Considering the very dense air traffic across the northern Atlantic, the flights to Male give relatively little information about the direct pollution from aircraft. However, the complex interaction of polluted air masses from the Tropics, and from the densely populated rapidly industrializing Indian sub-continent, via chemical changes and cloud formation in the chemically very active area of the Tropics undoubtedly needs closer investigation.

A demonstration of CARIBIC's potential is given in Fig. 7. Results are shown for O_3 , CO , aerosol, ^{14}CO , CH_4 , CO_2 , N_2O , and SF_6 against longitude for the return flight from Male to Munich on 26 November 1997. Flight altitude increased from 9.5 to 10.7 km. Very high CO levels of near $200 \text{ nmol mol}^{-1}$ were encountered in the Tropics over the Indian Ocean, which are caused by biomass burning. Over most of the Middle East, CO drops to about $100 \text{ nmol mol}^{-1}$, which is considered normal. Over the Black Sea, CO levels decline further, and an increase in O_3 is observed. Meteorological data confirm that at that stage, the aircraft reached the lowermost stratosphere. The increase in the number of ^{14}CO molecules is very clear, rising from a tropospheric level of around 10 to over 30 molecules per cubic centimeter STP. This is indicative of a substantial amount of stratospheric air in the air sampled. The data also show a good agreement between the CO as determined for the individual samples compared to the in situ monitored values. Very sudden increases in particle number concentration were observed, in particular around 30°N , demonstrating the high temporal resolution of the aerosol counting process. Concurrent with the increased aerosol, there was an increase in CO , as well as SF_6 and CH_4 , all of which are clear indicators for the convection of polluted boundary layer air. A detailed analysis will be published separately.

5. Conclusions and outlook

CARIBIC is a good demonstration that the automated measurement container for intercontinental passenger flights is feasible. Regular measurement flights at a rate of about two per month are planned, giving time for air sample analysis. In actual practice, one does not fly the container incessantly like the aircraft itself (well over 12 h day^{-1}) because freight cost would become prohibitive compared to the current economic countervalue of the additional scientific information obtained. Also, the retrospective analysis of the air samples requires time. Rather, flight routes that are of

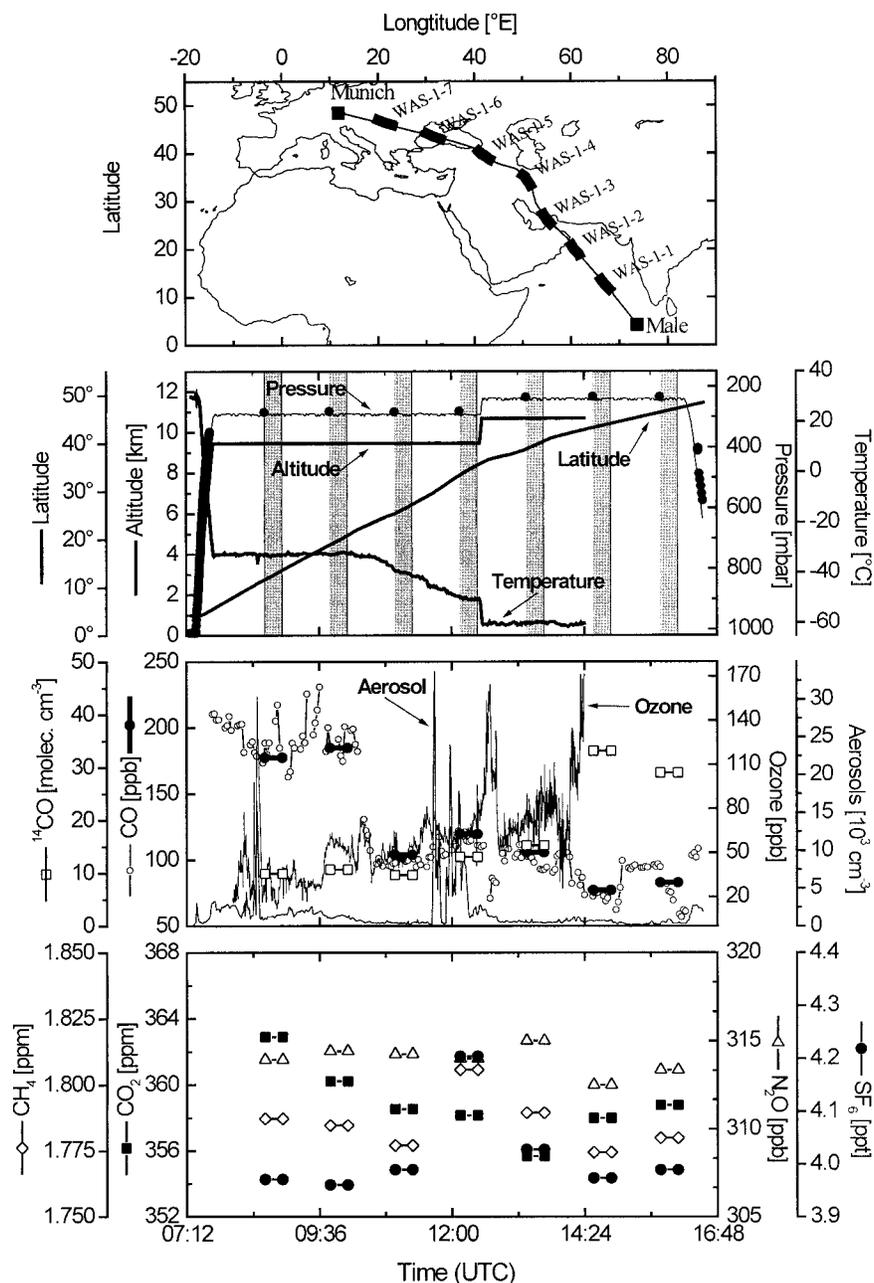


FIG. 7. Measurements obtained during the first return flight from Male to Munich 26 Nov 1997. The air sampling rate was determined by the filling times of the cylinders and the repeated flushing of the cylinders prior to collection. For subsequent flights, the flushing times have been reduced to be able to collect 12 samples instead of only 7. The ozone data from OMCAL show the considerable fine structure in ozone abundance. Large excursions up to near 150 ppbv are due to the interception of air mass of stratospheric origin. Also very sudden sharp features of increased aerosol have been detected, probably indicative of uplifted contaminated surface air masses. Only data for particles larger than 12 nm are shown.

greatest interest are selected. Transplanting the container into another aircraft, however, will require identical wiring and an inlet system to be available on the alternative plane. Compared to MOZAIC, CARIBIC will produce a less extensive, but very detailed data-

base, with a scope somewhere between that of routine monitoring and research flights.

The cost of the use of a measurement container in passenger or freight aircraft is very dependent on specific conditions. The lower limit of the running cost are

the actual freight charges, which, of course, are much lower than that for hiring a smaller aircraft for carrying out research. The cost of the inlet system and obtaining the approval may be higher for larger aircraft than for smaller ones.

Flexibility is also provided by the large amount of space available in the container and by a modular organization of the equipment. Furthermore, the very fact that the container is removed regularly from the aircraft provides time for attending to calibrations and possible problems without any interference with the aircraft operations. However, if different configurations of equipment are to be flown, then electromagnetic interference tests have to be passed for each configuration.

Limitations to research are posed by the inlet system and by requirement of safety and unattended operation. Also, certain substances such as liquid nitrogen cannot be used. Undoubtedly, analytical equipment will be developed further to increase the range of trace gases that can be detected and to reduce weight. It is foreseen that the immediate extension of the CARIBIC payload comprises a submicron particle collector and a multichannel aerosol spectrometer. Extensions with a water vapor sensor, an NO and NO₂ analyzer, a radon analyzer, and mass spectrometer are well feasible, thanks to the generous payload capacity.

The essential element for the success of a project such as CARIBIC is the committed cooperation of the airline, which has as its primary interest the transportation of passengers. Awareness of environmental impact of aviation, and the necessity of a proper assessment thereof in the light of the many pollutants released into the atmosphere by the wide range of anthropogenic activities, is a sufficient incentive for further innovative projects with the current one setting the trend.

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