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Design and Calibration of a Multi-Channel Aerosol Sampler for Tropopause Region Studies from the CARIBIC Platform

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An aircraft-based, multi-channel aerosol sampler for studies of the upper troposphere and lowermost stratosphere from the CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) platform was designed and calibrated. The sampler operates with an impaction technique at a flow rate of 10.4 lpm and consists of sixteen sampling channels. Samples are collected in a time sequence. Each channel contains two sample types that are used for quantitative measurement of concentrations, using particle-induced X-ray emission (PIXE), and single particle analysis with electron microscopy. The minimum detection limits for PIXE analysis after 1.5 h sampling are, for example, 2.0, 0.14, and 0.02 ng/m^3 STP (standard temperature and pressure) for sulfur, potassium, and nickel. Calibration included penetration studies of a cyclone arrangement used to define the upper size limit in the sampling to 2.0 μm diameter and the collection efficiency of the impactor. Both components of the sampling system showed penetration and collection efficiency close to 100%, respectively, in the particle size range of interest. The impactor cut-off was found to be dependent on the ratio of the impactor upstream-to-downstream pressure for ratios well below the critical pressure drop (i.e., the pressure where the jet reaches sonic velocity) being 0.15 μm and 0.08 μm for ratios 0.41 and 0.2.

INTRODUCTION

Because of its light-reflecting properties as well as their interaction with clouds, atmospheric aerosols induce a radiative forcing thereby affecting the global climate (IPCC 2001). This is one reason for the large number of recent studies about atmospheric aerosol. Systematic studies of upper tropospheric and lower stratospheric aerosol have been based on the use of remote sensing techniques such as LiDAR (Zuev et al. 2001) and satellite measurement of atmospheric aerosol concentration (Bauman et al. 2003) as well as balloon-borne and aircraft-borne aerosol particle counter measurements (Hofmann 1993; Hermann et al. 2003). They have given us a great deal of information about atmospheric aerosol concentration. Systematic studies of chemical characteristics of the aerosol in the tropopause region are scarce (Martinsson et al. 2001a; Papaspiropoulos et al. 2002; Martinsson et al. 2005). Still much remain to be discovered, in terms of both chemistry, composition, and the origin of the aerosol.

The CARIBIC project (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrumentation Container) (Brenninkmeijer et al. 1999) was initiated in response to the need for an improved knowledge of the chemistry and composition in the tropopause region. The project involves the development and operation of a laboratory in an airfreight container onboard a passenger aircraft. From the aircraft, regular observations are made of the upper troposphere and lowermost stratosphere. The CARIBIC measurement programme includes analyses both in situ and laboratory-based of a large number of trace gases as well as physical and chemical characterization of the aerosol.

The previously used CARIBIC aerosol sampler (Papaspiropoulos et al. 1999) was replaced in connection with a change in cooperation partner from LTU International Airways to Lufthansa, which also entailed a change from a Boeing 767-300ER to an Airbus 340-600. The new sampler design allowed a broadened analytical programme and improved time resolution in the measurements. Samples for quantitative atmospheric elemental concentration determinations (Papaspiropoulos et al. 1999) will be analyzed using PIXE (Particle-Induced X-ray Emission), whereas single particle morphology (Sheridan et al. 1994) and elemental composition (Xu et al. 2001) measurements will be based on electron microscopic analysis.

This work presents the design of the aerosol sampler that will be used in the CARIBIC project to collect particles in the upper troposphere and the lowermost stratosphere for subsequent analyses with PIXE and electron microscopy. Calibration results concerning the collection efficiency of the sampler are presented as well as the penetration of a cyclone with tubing installed upstream of the aerosol sampler, which defines the upper particle size limit to be collected.

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AEROSOL SAMPLER

Aerosol samples from the upper troposphere and the lowermost stratosphere are collected during CARIBIC flights. Capability to quantify aerosol elemental concentrations in environments with low aerosol concentration from an aircraft requires optimization of the sampling and analytical protocol. As the analytical method, PIXE was chosen because of its excellent absolute sensitivity (Johansson and Campbell 1988) allowing trace element analysis from samples with small total mass of only a few nanograms (Papaspiropoulos et al. 1999). To make maximum use of this feature, a sampling method that exposes a minimum amount of sampling substrate mass to the analytical beam is needed. Impactors concentrate particles to small spots of deposits. Using impaction in combination with a thin sampling substrate (AP1^{TM}; polyimide film of 0.2 μm thickness, provided by Moxtek inc., Utah, USA), meets the requirement. Therefore, this method was chosen to collect the aerosol particles.

Single particle analysis involves both elemental composition as obtained from scanning electron microscopy with energy dispersive detection of X-rays (SEM/EDX) and particle morphology measured with transmission electron microscopy (TEM). The latter technique requires thin sampling substrates. To meet this need, samples for single particle analysis are collected on thin carbon foils using impaction.

Below is a description of the second generation multi-channel aerosol sampler developed for CARIBIC to improve the previous version (Papaspiropoulos et al. 1999) with respect to time resolution in the sampling and sample capacity and to allow for a broadened analysis of the collected aerosol. After passing through the CARIBIC inlet system (Hermann et al. 2005) placed outside the aircraft, the aerosol passes an arrangement of a cyclone and connectors that together define the upper size limit of particles reaching the multi-channel sampler.

The multi-channel sampler, (see Figure 1) consists of 16 sampling channels. A sampling channel contains one sampling position for quantitative analysis of concentrations (PIXE sample) and two for single particle analysis (EM samples). Solenoid valves are located below the sample plane in all channels and are used to switch sampling on/off in a channel. An electronic circuit is used for communication between the valves and a computer on the aircraft, which is programmed so as to open or close the valves following a pre-defined sampling strategy. The aerosol flow is drawn from the sampler inlet into the lid and through nozzles of the channel with the solenoid valve opened. Aerosol particles exceeding the cut-off size will impact onto the impactor sampling substrates placed in the sample plane of an open channel.

The sampler is loaded with sampling substrates to cover both the outbound and the return flight of the aircraft. Fourteen of the 16 channels are used to collect samples in a time sequence (sequential samples). The remaining two channels are open during the entire outbound or return flight (integral samples). These two channels are used for the purpose of controlling contamination by comparing the collected elemental masses of an integral sample to the sum over the sequential samples. Each of these two channels contains one type of nozzle. The nozzle has one orifice, whose diameter is 0.5 mm. The distance between the orifice and the impaction plate is 6 times the orifice diameter. The critical flow rate of the nozzle is 2.0 $\pm$ 0.03 l/min. Every sequential channel contains two nozzles, one for quantitative measurements of elemental concentrations and one for single particle analysis. The former contains four 0.5 mm orifices and the critical flow rate of the nozzle 8.0 $\pm$ 0.08 l/min, whereas the other contains two 0.15 mm orifices with the critical flow rate of the nozzle 0.37 $\pm$ 0.01 l/min. The two orifices of the latter nozzle are separated by 4.2 mm in distance, thus making room for two EM samples in each channel. The EM samples are collected on 3.05 mm 600 mesh copper grids coated with thin carbon films. The grids are mounted in a specially designed grid holder before they are placed in the sampler. The AP1^{TM} sampling substrate for quantitative concentrations is mounted on a 40 $\times$ 28 mm Plexiglass frame over a 16 mm diameter hole. These frames are placed over the white circular features made of delrin plastic (see Figure 1), which provide support for the film and centering of the aerosol deposit.

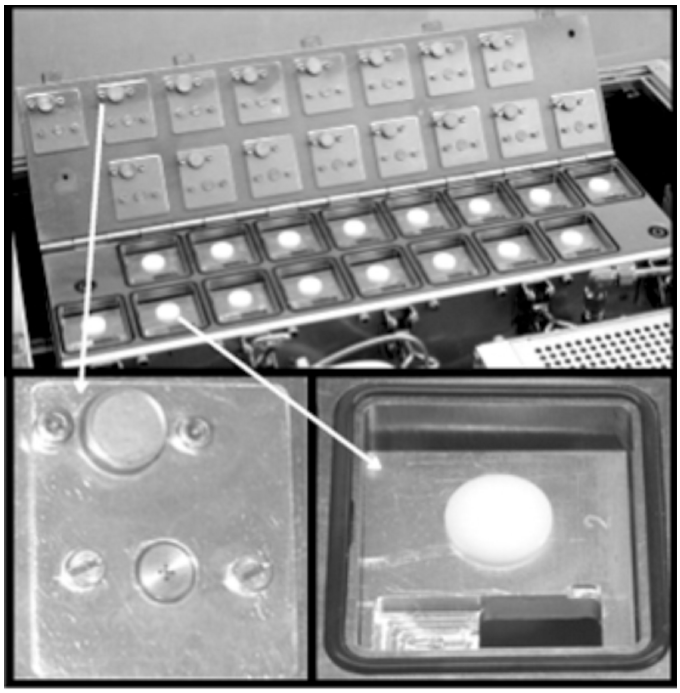

FIG. 1. The aerosol sampler, with the lid in its open position, consisting of 16 channels. Details are showing the nozzles (left) and the position where the samples are placed (right).

EXPERIMENTAL SET-UP

Since it is essential to obtain quantitative information from the PIXE samples, measurements were carried out on the cyclone arrangement and an impactor replica to determine the particle penetration and collection efficiency, respectively. In contrast,

the mass depositing on the EM samples is not important information for the single particle analysis. Consequently, these nozzles were not calibrated. Ideally, the cyclone and the impactor should be calibrated with the operational pressure at the inlet. The CARIBIC sampling normally is undertaken in the 200–300 hPa atmospheric pressure range. Due to the compression appearing during deceleration of the air in the sampling inlet, the pressure upstream of the aerosol sampler typically is 310–400 hPa. The Reynonds number of the impactor jet is between 2000 and 2700 in this pressure range. A laminar boundary layer of the jet does appear at these Reynolds numbers, which could worsen the cut-off characteristics of the impactor. For practical reasons, the impactor was calibrated using sea level pressure at the inlet. This can be justified by the sampling strategy used, where a single size fraction is taken with the 50% cut-off diameter far out in the small-particle tail end of the mass size distribution. The characterization of the cyclone arrangement was undertaken at 360 hPa.

The part of the set up, which was similar in both calibrations of the impactor and the cyclone, consists of a constant output atomizer (TSI model 3076) used to produce aerosol droplets from a solution of Dioctyl Sebacate (DOS) and uranine in isopropyl alcohol. The droplets were thereafter dried by particle-free, pressurised air in mixing chamber 1, producing particles consisting of 94% DOS and 6% uranine by mass. The polydisperse particle flow was bipolarly charged to equilibrium by a bipolar charger containing a radioactive source (10 mCi ^{85}Kr), see Figure 2. The aerosol flow was directed into a copy of a Vienna type Differential Mobility Analyzer (DMA) to obtain monodisperse particles of desired size (Reischl et al. 1997). Before DMA entrance, the particle flow passed a pre-impactor. The pre-impactor minimizes the disturbance from multiply charged particles of the DMA and hence the particle size broadening caused by multiple charges. A differential pressure meter (a Siemens model 7MF4420) connected across an orifice was used as a flow meter at the aerosol inlet of the DMA (Martinsson et al. 2001b). Filtered, pressurized air was supplied via a precision pressure regulator (Norgren model 11-818) and a needle valve to the sheath air inlet of the DMA, which was monitored by a mass flow meter (Bronkhorst F-112C-FA).

The monodisperse aerosol with particles of desired size was mixed with pressurized, particle-free air under vigorous

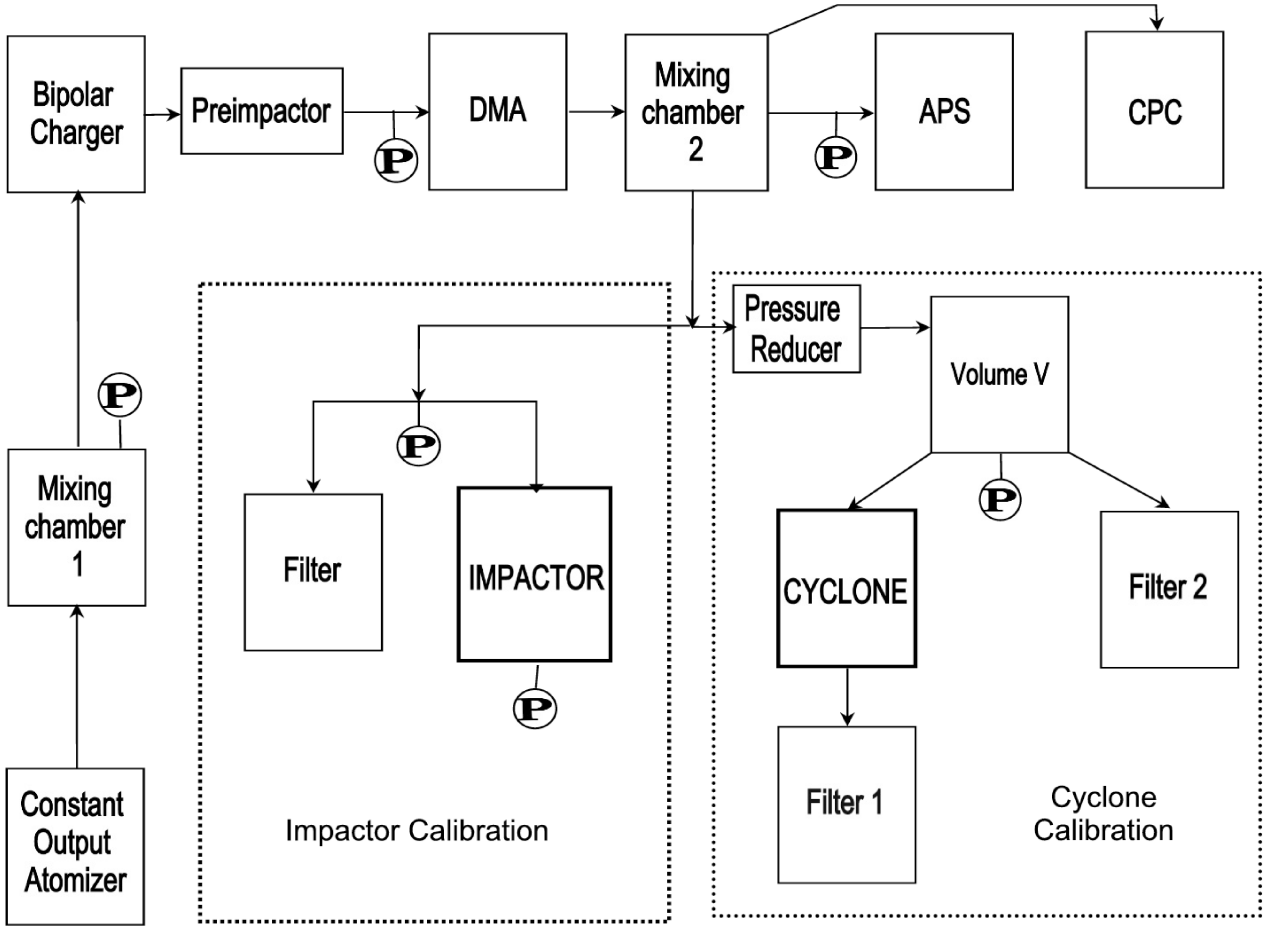

FIG. 2. The setup used in determining of the penetration of the cyclone arrangement and the setup used to determine the collection efficiency of the aerosol sampler.

turbulence in mixing chamber 2. A CPC (Condensation Particle Counter; TSI model 7610) was used to observe its particle number concentration. Furthermore, an APS (Aerodynamic Particle Sizer; TSI model 3300) was connected to mixing chamber 2 to monitor multiply charged particles.

The specific setup for each calibration will be described in the following sections.

Sampler Collection Efficiency

After mixing chamber 2, the aerosol flow was split into two streams (see the left dotted frame in Figure 2). One stream was directed to a replica of one channel of the sampler and the other to a filter (Nuclepore^{TM} filter with the pore diameter of 0.4 μm). The filter was placed inside a filter cassette. A mask placed underneath the filter concentrated the particle deposit into a central area of the filter in order to avoid losses to and contamination of the cassette. A needle valve behind the filter was used to adjust the flow to the same rate as the impactor replica. This flow rate calibration was made before each measurement using filtered air to avoid contamination of the Nuclepore^{TM} filter and impactor replica.

It is noteworthy that the sampler calibration system is closed relative the atmosphere. Therefore, four pressure taps were mounted in the system to monitor the pressures. The pressure drop of the pre-impactor varied with cut-off size. This was taken care of by allowing the pressure at its inlet to change as the pressure drop changed, keeping the pressure at the inlet of the impactor replica constant throughout the experiment at 1002 hPa. The pressure was monitored upstream of the pre-impactor, at the inlet of the DMA and upstream of the impactor replica. A fourth pressure tap was located after the impactor replica to measure its downstream pressure. Needle valves operated at critical conditions were used to set flow rates with the exception of the impactor replica. This flow-rate was defined by the impactor nozzle, which was operated at critical conditions. The downstream pressure of the impactor replica was varied by a downstream needle valve. Flow rates were set using a Bios International DryCal DC-1 flow calibrator (accuracy $\pm$ 1%).

A fluorescence-washing technique that was optimized by Tolocka et al. (2000) was used to wash and analyse the samples. A solution of distilled water and 0.01 N sodium hydroxide was used to extract the uranine mass from the samples with help of an ultra sonic bath. Each sample was extracted twice in 4 ml solution using a Transferpette (BRAND 0.5–5 ml), in order to account for residues after the first extraction. The mass of the second extraction was typically 6.4% of the first. After extraction, a TD-700 laboratory fluorometer (Turner Designs) was used to determine the uranine concentration. The mass collected on the filter or on the impactor film was determined using a calibration curve of uranine fluorescence intensity as a function of different, known concentrations. Measured blanks for filters and impactor films were determined and subtracted from the results of the measurements.

Cyclone Penetration

After mixing chamber 2, the aerosol flow has to pass through a pressure reducer (see the right doted frame in Figure 2). The reducer consisted of an impactor nozzle and an impactor house. Furthermore, from the volume V the aerosol flow was split into three streams. The first stream was directed into a 0.4 μm pore size Nuclepore filter, which was similarly mounted as in the impactor calibration. This filter was used as the reference filter. The second stream was oriented into the cyclone arrangement consisting of a cyclone, tubing and three elbows and union connectors, similar to the design upstream of the aerosol sampler in the CARIBIC container. A filter of the same kind as the reference filter followed downstream of the cyclone. The third stream was connected to a pump through a needle valve. By adjusting this valve, the pressure of the volume V could be maintained at 360 hPa. The pressures inside the setup were monitored by using four pressure taps. The penetration of the arrangement was obtained as the ratio of fluorescence analysed concentration on the filter downstream of the cyclone arrangement and the one on the reference filter.

Data Evaluation

Sampler Collection Efficiency

The impactor calibration data was first corrected for multiple charging affecting the DMA, which was minimized by the use of the pre-impactor. Then correction was undertaken for the collection efficiency of the nuclepore filter, which was determined experimentally as a function of particle size for the flow rate used in this study. The collection efficiency of a given particle size was calculated by dividing the corrected mass collected on the impactor film to the corrected mass on the filter.

The uncertainties in determination of the collection efficiency consisted of a variation of the flow rate through the impactor replica and through the filter cassette that was estimated to be less than 5% and a variation of the volume of the washing solution of 1%. Additional uncertainties resulted from the effect of the multiple charged particles occurred inside the DMA, which was less than 1%. The collection efficiency of the filter calibration was estimated to less than 0.5%. The fraction lost when washing off the particles from the impaction substrate and from the filter was estimated to between 2 and 14%, depending on the amount of the mass on the substrate and on the filter. The total uncertainty was between 3 and 16%, depending on particle size.

The Cyclone Penetration Efficiency

The uncertainties in determination of the penetration efficiency comprised a variation of the flow rate though the reference filter and the downstream filter that was estimated to be less than 2% and the variation of the volume of the washing solution of 1%. In addition, the fraction lost when washing off the particles from the filter was estimated to be between 0.3 and 12%. The total uncertainty was between 2 and 12%, depending on particle size.

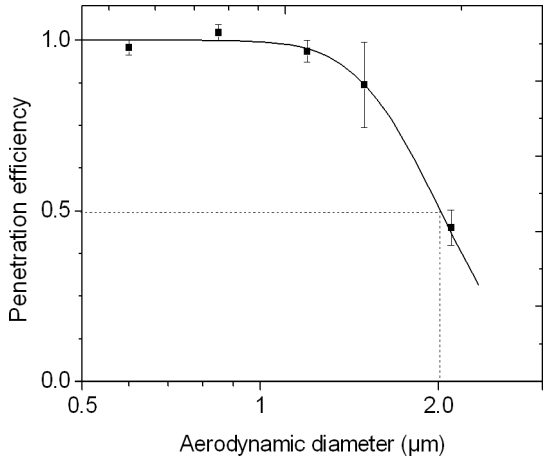

FIG. 3. The penetration of the cyclone as a function of particle size.

RESULTS AND DISCUSSION

Sampler and Cyclone Characteristics

The cyclone arrangement upstream of the aerosol sampler is used to define the upper particle size limit of the aerosol particles reaching the sampler. Coarse and fine particles are emitted from different kind of sources. They are largely externally mixed in the atmosphere and should preferably be sampled as separate size fractions. For keeping the sampler simple, a single particle size fraction of fine particles is collected. The results of the calibration are shown in Figure 3. It can be seen that the cyclone arrangement has a 2.0 μm cut-off diameter and that the penetration for small particle sizes is close to 100%.

The particle size dependence of the sampler collection efficiency was determined by the aid of a replica, as described in section 3. The calibration was undertaken with 1002 hPa pressure at the inlet of the sampler and the impactor nozzle was operated at its critical flow rate. Figure 4 shows the results of the calibration. The filled squares show the results obtained with maximum pumping capacity of the setup applied, resulting in the ratio of the downstream to upstream static pressure r = 0.2. The collection efficiency is high for large particles, on average 96.5%. The 50% cut-off was found to be 0.08 μm.

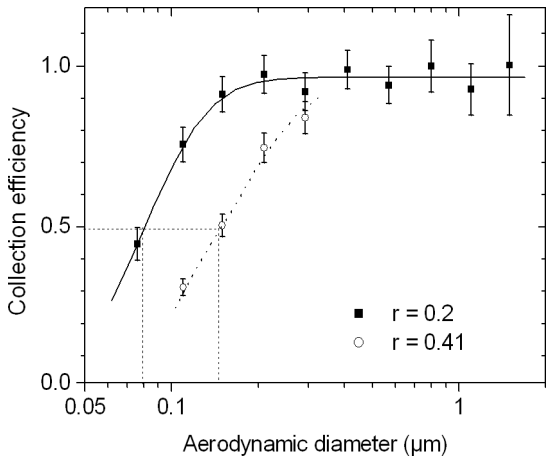

FIG. 4. The collection efficiency of the aerosol sampler as a function of particle size.

When making theoretical calculations on the cut-off characteristics of an impactor it is usually assumed that the pressure in the impaction zone equals the pressure upstream of the impactor nozzle as a result of the dynamic pressure exerted by the impactor jet. Making such a computation with sea level pressure upstream of the nozzle, the resulting cut-off diameter of a choked 0.5 mm orifice is 0.16 μm, thus significantly deviating from the experimental results obtained. Biswas and Flagan (1984) investigated high-velocity impactors and found that the conservation of the nozzle upstream pressure in the impaction zone is valid only for small to moderate pressure drops. As the nozzle approaches critical conditions, the pressure in the impaction zone starts to decrease in an accelerating way as the static pressure downstream of the impactor decreases. As a result, the 50% cut-off moves to smaller particle sizes.

In order to test this behavior, the static downstream pressure was increased to r = 0.41. This change in r did not affect the flow rate of the impactor, because it was still operated at critical conditions. The results are shown by the open circles in Figure 4. The cut-off diameter obtained was 0.15 μm. This clearly demonstrates how the downstream static pressure of the impactor affects the cut-off characteristics also in the pressure region where the flow rate is at the critical level.

The aim of the sampling is to obtain representative collection of fine particles, defined by the cyclone arrangement as particles less than 2.0 μm. Not all particles less than that size are collected by impaction. However, the aerosol mass contributed by particles less than about 0.1 μm generally is very small. The impactor upstream pressure will be in the range 300–400 hPa during CARIBIC flights, which is substantially lower than the pressure during the calibration. Making the same calculation as above for this upstream pressure range, the cut-off diameter of the impactor becomes 0.07 μm (for 300 hPa) and 0.09 μm (400 hPa). The pump used during flights delivers a downstream pressure of less than 100 hPa. The effect demonstrated in Figure 4 of low pressure ratios (r) can thus, combined with the effect of the reduced inlet pressure of the impactor, be expected to further lower the cut-off according to the calibration of 0.08 μm, to well down in the tail-end of the mass distribution.

Minimum Detection Limits

With the properties of the aerosol sampler and analytical parameters known, it is possible to estimate the Minimum Detection Limit (MDL) for PIXE analysis of the samples produced. The MDL of 21 elements were calculated based on these properties and the properties of backing film. The AP1^{TM} sampling substrate was thoroughly investigated already in the first phase of aerosol sampling in CARIBIC. This implies that we have a wealth of information about the properties of that substrate

TABLE 1
Minimum detectable concentration for 1.5 h sampling of a sequential sample

Element	Concentration[a] ng/m^3 STP[b]	Element	Concentration[a] ng/m^3 STP[b]	Element	Concentration[a] ng/m^3 STP[b]
Aluminum	12	Titanium	0.054	Zinc	0.34
Silicon	40	Vanadium	0.015	Gallium	0.011
Phosphorous	1.3	Chromium	0.034	Germanium	0.022
Sulfur	2.0	Manganese	0.016	Arsenic	0.015
Chlorine	0.83	Iron	0.25	Selenium	0.030
Potassium	0.14	Nickel	0.021	Bromine	0.050
Calcium	0.46	Copper	0.10	Lead	0.030

[a] The detection limits are given at the 99% probability level.
[b] The detection limits are given at standard temperature and pressure (STP).

obtained from the analysis of in total 133 blank films. In the calculation of the MDL, the time used to collect a sequential sample was set to 1.5 h, the sampler inlet pressure to 325 hPa, the diameter of the accelerator-produced beam to 4.5 mm and the beam charge to 80 μC. As shown in Table 1, the MDL of the elements varies by more than three orders of magnitude from 0.01 to 40 ng/m^3 STP. Some of the elements are affected by blank concentrations in the AP1^{TM} film (e.g., silicon and zinc), which can be observed as high MDL relative to neighbouring elements with respect to atomic number. These low detection limits were obtained as the result of the use of PIXE, which has high, absolute sensitivity, optimization of the analytical parameters (Papaspiropoulos et al. 1999) and the properties of the aerosol sampler presented here.

CONCLUSIONS

The second generation CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) aerosol sampler was designed and calibrated. It is used for sampling from a passenger aircraft in the upper troposphere and the lowermost stratosphere. The sampler contains 16 sampling channels, from which two integral channels are used for contamination control and 14 sequential channels are used for collection of samples in a time sequence. The collected samples are analyzed with Particle-Induced X-ray Emission (PIXE) to obtain quantitative information about elemental concentrations as well as single particle analysis with electron microscopic methods (EM).

A cyclone arrangement upstream of the aerosol sampler is used to define the upper size limit of the collected particles to 2.0 μm diameter. Calibration in the particle diameter range 0.6–2.1 μm revealed that the penetration of sub-micron particles is close to 100%. The sampler is based on impaction, with separate nozzles for PIXE and EM samples. The former part of the sampler was calibrated with respect to particle collection efficiency using a fluorometric method to analyse the calibration samples. It was found that the collection efficiency was high, 97%, for particles larger than approximately 0.2 μm diameter and 50% cutoff was obtained for 0.08 μm. The calibration was undertaken with sea level pressure at the impactor inlet and the pressure ratio downstream/upstream the nozzle was r = 0.2. The collection efficiency was dependent on downstream pressure. With r of 0.41, that is, the critical flow rate maintained through the nozzle, the 50% cut-off diameter was 0.15 μm, thus demonstrating cut-off dependence on the pump capacity at pressure ratios larger than the critical pressure ratio.

The sampler developed shows excellent sampling characteristics for fine particles. The samples produced for quantitative analysis provide low detection limits after 1.5 h sampling. In addition, samples are collected for single particle analysis. In conclusion, the second generation CARIBIC aerosol sampler is a powerful tool for aerosol characterization from an aircraft.

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