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Atmospheric Chemistry and Physics Discussions

ACPD

7, S3205–S3215, 2007

Interactive Comment

Interactive comment on "Civil aircraft for the regular investigation of the atmosphere based on an instrumented container: the new CARIBIC system" *by* C. A. M. Brenninkmeijer et al.

C. A. M. Brenninkmeijer et al.

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Referee # 1

We thank the referee for the comments. In general the reviews have lead to a substantial amount of improvements and refinements thus better specifying the instrumentation and its performance. Concerning the question "Should CARIBIC be managed as a resource to the community similar to a satellite, or should itĚ." we note that this is a complex discussion. (if the status "as a resource" would mean funding, it would be a pleasant discussion). Our colleagues indeed can well judge the value of the detailed datasets over long time periods.

(5284) Concerning the issue of flight routes we state that the present situation is that all routes are very valuable. Using the CARIBIC aircraft to attend AGU Fall meetings certainly brings us to higher latitudes and stratospheric altitudes. The most effective way to increase the value of observations is to fly longer distances, for instance we presently discuss Frankfurt-Seoul-Frankfurt-Denver-Frankfurt.

(5288) Concerning the important point about the aerosol inlets we state that the inlet tips and shroud are heated to prevent icing. The amount of heat supplied is insufficient to impact on the aerosol. It is the aerodynamic heating inside the diffuser tube and inside the aircraft (temperature below the cargo floor) and in the container that determines to what degree water is shed by the aerosol. This is now addressed in the paper.

(5289) Concerning cloud water: The ratio U/Uo (aircraft air speed/ inlet sample speed) which mainly determines the inlet enhancement factor Einlet is measured accurately (error $<$ 5%). Einlet of our configuration is small compared to most systems installed on research aircraft, see e.g. Davis et al., (Measurement of total water with tunable diode laser hygrometer: Inlet analysis, calibration procedure, and ice water content determination, J. Atmos. Ocean. Technol. 24, 463-475, 2007) who has a factor of ˜ 35. Davis et al. (2007) write: "IWC accuracy ranges from 20% at the largest IWC to 50% at small IWC (˜5 mg m-3)". We have not yet conducted a similarly detailed error analysis. This will be reported in a separate technical paper.

(5297) Concerning ambient pressure, we noted this point and modified the text.

(5300-5301) Concerning NO and NOy, we note that in contrast to research aircraft (e.g. Falcon) we could not use CO gas, and use H2 instead. The conversion efficiency has been tested and the results are now mentioned. The gold converter should be ideally at the inlet but this was not possible. We have not tested our system for losses and memory effects but its behavior is assumed on the basis of experience the NOy community has with PFA and heated tubing (Neuman et al., 1999, now cited in the text).

ACPD
7, S3205–S3215, 2007

(5304) Concerning the high resolution of 256 channels, the point made is accepted.

(5307) Concerning the O2/N2 ratio we remark that this expression may be not optimal, it is the determination of the O2 content of dry air. The measurement using fuel cells is not absolute and is not a ratio measurement as such. Also frequent comparisons with standards is required. The principle is that the amount of air in the cell is very accurately controlled, and the resulting emf from O2 measured. In this way a ratio is established. Concerning the minor points, these have been gratefully included in the improved version.

(5312) Concerning the canisters, we have clarified the point that the analyses in 4 laboratories in 4 countries require a sufficient supply of units to obtain a turnover rate of 2 per month.

(5313) Concerning the filling this point has been clarified. Minor points (wording): these have been gratefully accepted.

(5289) The ratio U/U0 (aircraft airspeed / inlet sample speed) which mainly determines the inlet enhancement factor Einlet is precisely measured (error $<$ 5%). Einlet of our configuration is small compared to most system installed on research aircraft, see e.g. Davis et al. (Measurement of Total Water with a Tunable Diode Laser Hygrometer: Inlet Analysis, Calibration Procedure, and Ice Water Content Determination, J. Atmos. Ocean. Techn. 24, 463-475, 2007) who has a factor of ˜35. Davis et al. (2007) write: “IWC accuracy ranges from 20% at the largest IWC to 50% at small IWC (˜5 mg m-3)”. We have not yet conducted a similarly detailed error analysis. This will be done in a relevant future technical paper.

Referee # 2

We thank the referee for the extensive comments and many changes have been made. Concerning the statement “.., this paper is weighted far too heavily towards the engineering side..” we do not entirely agree. We do however agree that this paper is weighed towards the engineering side, and we make sections indicated by the referee more concise in this respect. When we consider the aircraft provisions with container, the inlet etc. as one apparatus for making atmospheric observations, specific changes to the aircraft are essential part of the technical/scientific description. We considered submission to Atmospheric and Oceanic Technology, but reflected that a technical note/paper with some engineering information would suit ACP, certainly given the fact that the container system is unusual. In the end we believe the revised version is weighted towards the engineering side to an acceptable degree. When or if other groups would like to set up a system like CARIBIC, the current communication should contain the necessary information, also highlighting the most salient engineering aspects that are typical for this type of observatory. The intended publication forms the sole possibility to present the entire system, be in a concise form. Nevertheless in the revised version we have taken the comments seriously. A fully annotated version of all changes can be made available on request.

2) Concerning figures 4, 5 and 8 we insist to conserve figure 8 (Now numbered figure 6 as 4 and 5 have been deleted).

3) We shortened the description of instruments under development, the TD device and the O2 instrument (now 3 short paragraphs).

4) Uncertainties for the 15 experiments are now included.

5) We cannot give references for the air sampler, other techniques are well documented in the literature.

6) This is very important but for the many compounds and analysers a long discussion, beyond the scope of this paper. For a great deal of the compounds measured there is a low degree of uncertainty about the measurement errors stated. For instance CO, or O3. For NOy and acetone for instance, or particles, it is much more complex. The estimation in the uncertainty of such data relies for a great deal on the experience of the PI operating the instrument. Comparison flights with research aircraft is the most

ACPD

7, S3205–S3215, 2007

Interactive Comment

objective way of comparing. Also checks of the internal consistency of the datasets helps us to detect possible deviations.

A) We removed the sentence line 20-21. The remaining brief statement is the most essential one for any other group wanting to construct a system like this.

B) We have removed 80% of the requested amount.

C) We removed lines 20-24 and figure 4.

D) We removed lines 27-29 only and figure 3. The statement about mounting a sizeable inlet system on a passenger aircraft is kept however.

E) Section 3.3. We would like to keep this section as it describes the container in 40 lines of text only. Cooling fans and temperature are essential to the operation of the system, and by no means trivial points, on the contrary. We would like to communicate somewhere what is involved, and this technical paper is the best place for the general features. Engineering details we have fill a bookshelf.

F) We wish to keep these 7 lines describing the essential interface container/aircraft with figure 8. The coupling is more completely described in the caption.

G) and H) = Instrumentation section general part. We shortened this part of the overall instrumentation, it is however essential information on the CARIBIC system described in this paper. For instance how the calibration and working gases are supplied is essential to the entire set up (5295 lines 19-29) and part of the innovation.

I) Done.

J) We presume the statement about DOAS being suited is meant here. We have reworded this statement completely. K) Done.

L) We feel this goes a bit far. When we for instance specify the amount of power used for the system this is certainly useful to know, but does not pertain the evaluating the data quality. The paper also is a description of the entire system.

ACPD

7, S3205–S3215, 2007

Interactive Comment

2) A) Figure 4 has been removed

B) Figure 5 has been removed.

C) Figure 8 (now Fig. 6) as stated we like to retain.

3) A) We have shortened the description of this instrument considerably.

B) Also the description of the TD collector is shortened.

4) These omissions have been addressed in the text and the Table 1.

4) A) The cloud particle enhancement at the location of the total water inlet was calculated by Airbus Industries (based on fluid dynamic calculations) to be 12-19% for different flight conditions (differing in pressure, aircraft speed, angle of attack etc.), see section 3.2. As given in the text, the time response and time resolution of the chilled mirror hygrometer is much worse than the ones of the photoacoustic device, particularly at very low humidity, as can be seen in Fig. 10. The chilled mirror hygrometer cannot follow the rapid fluctuations in humidity and shows the overshoots typical for this sensor type, e.g. at 5:22. In a first step of data post-processing the chilled mirror hygrometer data are corrected for the sensors' time lag (measured in the laboratory and ranging from 10 sec at 1000 ppmv to 250 sec at 10 ppmv). Thereafter, by correlating the signal of the photoacoustic sensors (microphone voltage) with the humidity of the chilled mirror hygrometer an actual photoacoustic sensitivity (in nV/ppmv) is derived. For this continuous calibration of the photoacoustic device a running mean of the data over about one hour is applied. Over a flight sequence of four flights the sensitivity of the photoacoustic device typically changes by ˜25% due to slow (mostly temperature driven) drifts of different parts (laser, electronics,..).

B) The description of the CO instrument has been extended. The modifications of the instrument, its operation and the treatment of the data are described. Typically achieved performance is specified. The sample air is not being dried because of the very low water content of the air at flight level. No measurements are being made at

ACPD

7, S3205–S3215, 2007

pressures above 650 mbar.

C) The text has been extended to add information about the conversion efficiency determined in our laboratory including that of HCN. The operation of the instrument (zeroing, determination of the zero air artifact) and its achieved performance are also specified.

D) The uncertainties for Aitken mode and ultrafine particle number concentrations are now given.

E) The section 4.7 Aerosol particle sampling and analysis has been modified considerably to address the points raised.

F) Use is made of 3 calibration gases (was wrongly reported). Two gases sufficiently close to the atmospheric values would in principle be sufficient.

G) Uncertainty estimate is given. The RGM transmission of the inlet tubing was not investigated but the sampling conditions are similar to those encountered during the measurements in the Antractic made by Temme et al [2003] which is mentioned in the text now. At low humidity of the sampled air RGM was found to pass heated PFA tubing and FTFE filter.

H) The information was indeed missing. The typically achieved performance of the PTRMS is now specified in appropriate detail.

I) The flushing time is now specified.

J) The DOAS section has been upgraded, and the degree to which quantitative information can be retrieved is specified.

5) We have given the relevant references in the text.

6) We have added a small section “Operations” and address the issues raised by the referee, namely data quality/plausibility and calibrations in the conclusions.

Technical corrections. All with the exception of point 5) have been incorporated.

Referee # 3

We thank the referee for the comments. We have removed several technical details. The acquisition of vertical profiles is indeed desirable. We note however that most of our systems refrain from taking in air below ˜ 5 km in order to avoid high humidity and contamination. To measure with one system the lowest concentrations as repeatedly encountered in the lowermost stratosphere as well as more strongly “polluted” air is a challenge that we cannot meet. We also note that the long distance flights imply that well over 90% of the time we are at cruising level. Nonetheless the DOAS system operates down to lower levels and can capture pollution plumes at low altitudes.

(5301) Concerning the length of tubing with respect to NOy measurements we note that the flow rate is 1.5 l/min (STP) and that the tubing is lined with PFA and heated to 40oC. The absence of loss of NOy is solely based on experience with other systems and tests using PFA and other materials (Neumann et al., 1999).

(5304) The upper threshold diameter of particles detected by the CPCs is estimated to be 2 μm based on knowledge about the old CARIBIC inlet and will have to be determined more accurately by either wind tunnel or CFD modeling in the future. But its exact value is not important since the Aitken mode particles usually dominate the total particle number concentration in the upper troposphere.

(5304) Concerning “teething” problems, this colloquial expression has been removed.

(5312) Concerning air sampling, the flushing of the glass cylinders, and flow rate and timing have been addressed in the text

(5313) Concerning ozone we agree that this important and difficult. It is known that alkenes tend to form in dry metal canisters, and we assume this is not due to ozone chemistry. Tests of our pumping system for ozone destruction are planned. With results all more reactive species collected in the flasks we are extremely cautious. Tests we

ACPD

7, S3205–S3215, 2007

Interactive Comment

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have made by measuring CO (Dr. Ingeborg Levin, Umweltphysik, Heidleberg) in the flasks show elevated CO values compared to the in situ data for stratospheric samples. In the previous CARIBIC system (using ss flasks at 17 bar) we had isotopic evidence of the formation of CO with oxygen from ozone, unpublished results). We are continuing tests to characterize pumps and flasks better.

(5335) Concerning Figure 10 (now Fig. 8), this is now addressed in the caption.

(5336) Concerning the O3 and NO data reproduced in Fig. 11 we are grateful for the sharp eyes of the referee: Preliminary raw O3 data were shown. The final and accurate O3 data are displayed now. The reason for the enhanced NO and NOy concentrations in the second half of the flight shown in Fig. 11 are plumes which are also visible in the CO data.

Referee # 4

We thank the referee for the comments. We have removed several technical details, and some non-scientific assessments about the system. Concerning the redundant figures we would like to keep the photo in Figure 8 being about the most essential interface. We have added text in the caption to clarify the connections. The other 2 figures (4 and 5) have been removed. Concerning the performance parameters, here we have made improvements and better quantified these wherever possible.

O3 scrubber: The text was unclear indeed. We actually conducted a lot of laboratory tests to determine the efficiency of the scrubber in dependence on scrubber surface and temperature. The final and actual scrubber design is on the one hand able to destroy quantitatively O3 as high as 800 ppbv and on the other hand has a small scrubber surface to minimize artifacts due to high and variable water vapor concentrations. This effect is well-known and recently reviewed by Wilson and Birks (2006). The new CARIBIC O3 instrument currently in development will be furnished with a Nafion drier to eliminate this effect.

ACPD
7, S3205–S3215, 2007

Chemiluminescence O_3 detector: The sensor disc is typically used for ˜5 flight sequences (20 flights) during which the sensitivity gradually decreases to ˜1/3 of the initial value due to aging process. In addition, high water vapor concentrations increase the sensitivity of the detectors for O_3. This interference occurs only at H_2O mixing ratios $>$ 3000 - 5000 ppm, i.e. essentially only during the ascent and descent. The H_2O interference has not been considered so far, but can be corrected using the simultaneously measured H_2O vapor concentration.

Concerning the CO analysis using VUV: The modifications made to improve the performance of the commercial CO instrument are now described, and typically achieved performance is specified. Using the suggestion by the referee we could use the event of cruising in a large uniform air mass (uniform with respect to CO) for over 500 km, to establish that the 10 second variability of the CO data was about 1.5 nmol/mol only. Compared to laboratory conditions, a main concern is the temperature stability of the instruments sensitivity.

Concerning the NO/NOy instrument: Its description was expanded and typical performance is given. O_3/O_2 flow is being humidified. With humidification no sensitivity changes have been observed during numerous flights in the last 10 years. This justifies the determination of the sensitivity before and after each flight. The changing zero-air artifact is determined every 2 h during the flight as described now in the text. Because of the restriction imposed on the passenger aircraft it was not feasible to determine the transmission of HNO_3 of the inlet and the tubings (all lined or made of PFA and heated to 40oC) at flight conditions. Tests in laboratory simulating the conditions in the flight, however, have not shown significant losses of HNO_3. This is in agreement with measurements reported by Neumann et al. (1999). Results of our measurements of the conversion efficiency are now mentioned in the text. The PMT are not cooled by Peltier but by closed cycle coolers.

Concerning air collection in flasks we have added Fig. 12. The amount of stainless steel in the flask system is small (2.5 mm OD tubing to the Valco valves on each side

ACPD

7, S3205–S3215, 2007

Interactive Comment

of the flasks) and also the stainless steel body of the valve exposed is very small. The rotor material is suitable for CG application of trace gases and should not interfere. We also note that we have 10 l air in the flasks. The bonding cement used is 2 component epoxy glue compatible with HV applications. Outgassing is inherently low and the exposed epoxy surface is only about 0.3 mm3. No outgassing for the measured VOCs was observed. Fast losses in the pumping system for certain compounds cannot be excluded. Work related to problematic species is in progress. Like all other air sampling systems some trace gases may not be reliably measurable with this system, yet the generous amount samples, and the use of glass is the best option we believe. The pumps are switched off between sampling to safe power and to keep the pump temperatures as low as possible. Sampling of 28 samples on the first outward bound leg is practiced to be sure that when further sampling would fail, we still would have the full contingent of 28 samples.

(5299) Concerning the fluorescence measurement of O3: The sensor disk is typically used for ˜5 flight sequences (20 flights) during which its sensitivity decreases to ˜1/3 of the start value. This is a gradual and slow aging process. Beyond that a cross-interference with H2O exists. High H2O concentrations increase the detector sensitivity for O3. This cross-interference is visible only during the descent and at H2O mixing ratios $>$3000-5000ppmv. Currently these data are not considered, but can in case be corrected by using the simultaneously measured water vapor.

Other minor comments incorporated with thanks.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 5277, 2007.