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Status report: The Groningen AMS facility

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Abstract

The Groningen AMS facility has been in operation since 1994. The AMS is based on a 2.5 MV tandemron accelerator. It is an automatic mass spectrometer, dedicated to ^{14}C analysis. Thus far, a grand total of about 16 000 ^{14}C targets have been measured. We report here on the status and performance of the facility, technical improvements and a precision study on atmospheric samples. © 2000 Elsevier Science B.V. All rights reserved.

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Keywords: AMS; Radiocarbon; Standards

1. Introduction

The Centre for Isotope Research (CIO) of Groningen University started in the 1950s as one of the first ^{14}C laboratories, and employs presently a high-precision conventional laboratory with proportional counters for 1–25 l of CO_2 [1]. The CIO obtained funding for an AMS system in 1990. The Groningen system was the first one built by high voltage engineering in Amersfoort, The Netherlands [2] and is a ‘sister’ machine of the ones operational at Woods Hole, USA [3] and Kiel, Germany [4]. The first results obtained at the Groningen AMS facility were presented at the 15th International Radiocarbon Conference in Glasgow [5] and at the Seventh International AMS Conference in Tucson

[6]. For a description of the AMS system we refer to these earlier reports and to Purser [7].

The Groningen AMS is dedicated to ^{14}C only. Routine precision is about 5‰ in $^{14}\text{C}/^{12}\text{C}$, and about 1‰ for $^{13}\text{C}/^{12}\text{C}$; routine backgrounds correspond to ages of 45–50 ka. The AMS is being used in almost all applications of (natural) ^{14}C . A great advantage is the infrastructure of the CIO which includes a high-precision conventional $^{14}\text{C}/^3\text{H}$ laboratory, a stable isotope facility (^2H , ^{13}C , ^{15}N , $^{17,18}\text{O}$) and a newly established laser isotope ratio spectrometer [8]. Apart from service work, the largest research projects for the AMS are ^{14}C calibration-related projects, archaeology, ocean sciences and atmospheric programmes.

For ^{14}C calibration, the highlight is the ^{14}C measurements for a varved sediment in Japan back to 45 ka [9]. Now more than 300 macrofossil samples from this key site have been measured. Also, Holocene wood samples from Japan are being measured to establish a thus far floating

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dendrochronology. There is not enough wood for high-precision conventional measurements [10]. Finally, we have shown that peat deposits can be dated accurately by ‘wiggle matching’ of selected botanical remains [11].

In *archaeology*, a new development is the possibility to date cremated bones. These do not contain any collagen, the datable fraction for normal bone material. It appears that for cremation rests, apatite yields very good results. A large series of cremated bone samples with known age (based on other prehistoric evidence) has been dated with a success rate of practically 100% [12].

For *ocean sciences*, two special projects were funded: ^{14}C in ocean dissolved organic carbon (DOC), and $^{13,14}\text{C}$ in particulate organic carbon (POC) combined with chemical characterisation [13,14]. In *atmospheric sciences*, the CIO participates in ^{14}C projects involving many hundreds of samples annually. Examples are the project COOH (^{14}C in CH_4 and CO), reported by Brenninkmeijer et al. [15], event-trapping experiments [16,17] and ^{14}C in atmospheric CO_2 . For many years, our laboratory measured $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ in atmospheric CO_2 for samples collected in the established atmospheric network of Keeling and coworkers [18] and for the years 1985–1991 selected samples were frozen back after measurement of the stable isotopes on the IRMS and stored in breakseals – waiting for ^{14}C analysis by AMS which became operational since 1994. For this study, a thorough investigation of precision and standards for AMS will be presented in the next chapter. The final results for $^{14}\text{CO}_2$, based on this analysis, for two key monitoring stations, Point Barrow, Alaska (61 samples), and South Pole (59 samples), for a seven-year period are shown in Fig. 1. The $\Delta^{14}\text{C}$ values at both sites show a decreasing trend of 10‰ per year, and the seasonal cycle on both sites could be reconstructed. A full analysis will be published shortly [19].

2. Results

2.1. Measured samples

Over the last five years, around 16000 targets were measured. This includes the ‘overhead’ of test

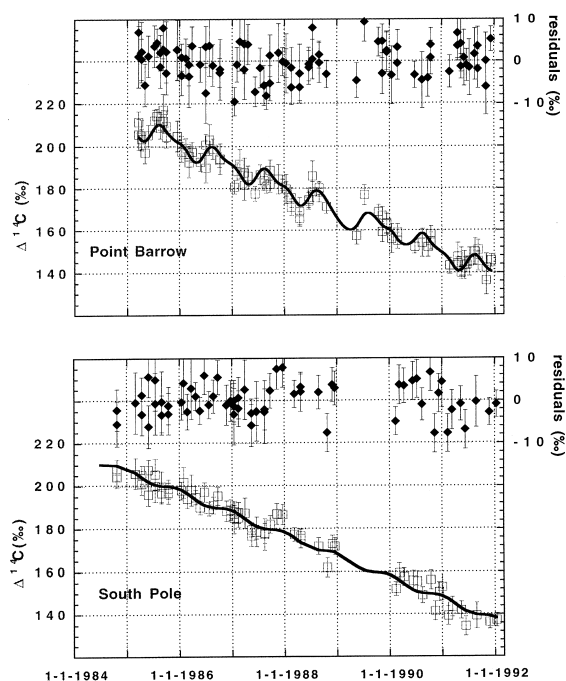


Fig. 1. Results for atmospheric $^{14}\text{CO}_2$ measurements by AMS for the stations Point Barrow (Alaska) and South Pole (PTB respectively SPO).

samples, standards and backgrounds. Automatic batch measurements are run around the clock. A target wheel can contain a maximum of 58 samples. A typical batch consists of 56 targets: 6 backgrounds, 12 standards and 38 unknowns. The remaining two targets are samples to tune the accelerator. Such a batch takes about 44 h of measuring time. Usually, two wheels (batches) per week are measured.

A diagram indicating the various sample materials handled by the Groningen AMS laboratory is shown in Fig. 2. The diagram represents five years (1994–1999) of operation. As can be deduced from Fig. 2, the largest field of science is archaeology. The ‘various’ category includes graphite or CO_2 produced by other laboratories and measured in Groningen. This is possible because the AMS can measure more than our sample preparation laboratory can handle. Moreover, the high-throughput machine is ‘standby’ for about 50% of the time.

This situation will improve this year by using the automatic combustion/trapping system (see

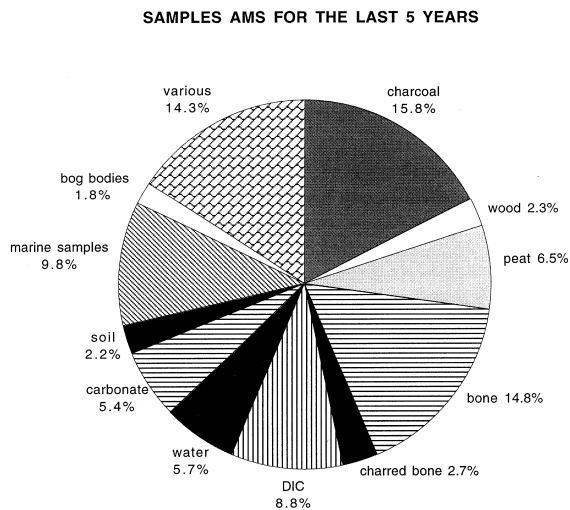


Fig. 2. Diagram indicating the variety of samples measured by the Groningen AMS laboratory.

below). Further automation (for carbonate, water, breakseals) is considered. To guide operation, a database system is set up based on Filemaker Pro. It is a truly relational database. It contains all relevant facts about the AMS samples such as pretreatment, graphitisation, customer administration, storage, and the information on measured ^{14}C plus ^{13}C . The database is present on the server of the CIO, and accessible over the laboratory's computer network (MacIntosh based). Also, the stable isotope laboratory is under the guidance of the database system. In the near future, the conventional ^{14}C database will also be transferred to the Filemaker Pro system.

2.2. Technical improvements

2.2.1. Accelerator

A variety of technical improvements to the accelerator system increased the reliability of operations considerably over the last five years. An overview of selected incidents during this period is shown in Fig. 3. In 1994, the downtime was 35% (of all available time in the year); this decreased to 13% in 1998. In 1998, the accelerator was measuring samples for 35% of the time; the remaining time (52%) was 'idle time' and is in theory also

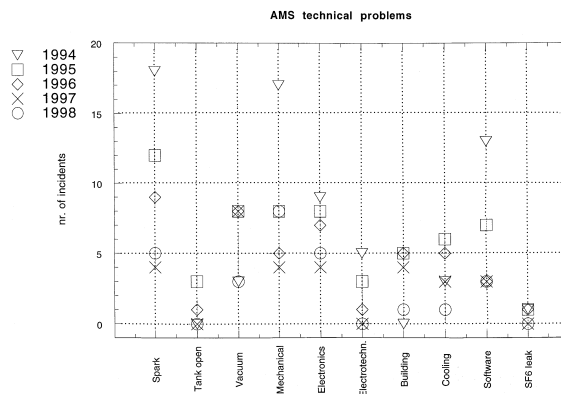


Fig. 3. Overview of problems with the accelerator system during 1994–1998.

available for measurement – the bottleneck being sample preparation.

We present here a list of the most important changes: ferro-fluidics feedthrough for the chopper wheel; a new 'switch mode' power supply for the four recombinator magnets; a target arm without bellows; a fast loading system enabling the exchange of one target without breaking the source vacuum; remote control of the stripper valve; installation of a residual gas analyser (RGA) in the high-energy beamline; a new target carousel with delrin inserts; a softstarter for the HV driver; a new cooling system with a primary and a secondary closed cooling loop; a gradual replacement of most optical interface cards by a more stable design; and finally an unfortunate necessity: an uninterruptible high-power UPS as a backup power supply.

2.2.2. Sample preparation

For sample preparation, the most important improvement of recent years is the automation for organic sample combustion. The combustion system is a CarloErba 1500 elemental analyser (EA), on-line with a Micromass Optima stable isotope mass spectrometer (MS). The EA consists of a Cr_2O_3 flash combustion tube, a silvered cobaltous cobaltic oxide purification furnace, a Cu reduction tube, a water trap and a gas chromatographic (GC) column to separate CO_2 and N_2 . The EA is coupled on-line to a continuous flow MS enabling precise $^{15}\delta$ and $^{13}\delta$ measurements.

Originally, CO₂ was trapped manually after the GC column for graphitisation; this has now been changed as follows. The EA/GC/MS is designed originally for stable isotope measurements. In order to optimise performance for AMS purposes, the EA has been modified; it is originally designed to deliver 2–10 µg C to the MS only. After modification, the gas flow (CO₂ after combustion plus the He carrier gas) is 120 ml/min with 1 mg C, and is split in two pathways: (1) 119.5 ml/min He ('waste') with ≈2 mg C to the cryogenic trapping system, and (2) 0.5 ml/min with ~8 µg C to a new GC column (smaller than original design) followed by a splitter valve. The latter tunes the flow to the MS to 0.1 ml/min (2 µg C), whereas its waste line contains the remaining waste (He and 6 µg C).

The advantages are: better timing between the MS and the cryogenic trapping (the N₂/CO₂ separation is now cryogenic); no GC column on the pathway towards the cryogenic collection system; 99% of all the C combusted is collected for AMS.

The cryogenic system consists of 40 Pyrex cold traps. Each sample combusted by the EA will deliver its CO₂ to one of the traps. Each trap has a pneumatic valve. A small cart/elevator with a liquid air dewar can move from trap to trap. The system is shown schematically in Fig. 4. The system is fully automated using LabView, under communication with the EA/MS commercial software.

In principle, the system can run unattended and automatically combust/measure ¹³δ/trap CO₂. The trapped CO₂ will be collected and transferred to the graphitisation setup. We employ the method of reduction under hydrogen excess with iron powder as catalyst: CO₂ + 2H₂-(Fe)-2H₂O + C. The graphite is then pressed into a target which fits into the ion source carousel, using an automatic press [20].

2.3. Contamination tests: memory effects

A continuous effort is the testing of backgrounds. Various background materials were measured and reported earlier [20]. Results for the build-up of the blank during the various stages of sample preparation are shown in Fig. 5.

The lowest backgrounds have been obtained for a graphite rod, which can be mounted in a target holder without any handling in the sample preparation laboratory. This 'machine blank' measurement is 0.08% (activity reported according to [21]), corresponding to an age of 60 ka. Graphite powder is also available; the only handling for this material is the pressing in a target holder. Romenhöller is a fossil (¹⁴C-free) CO₂ gas which provides background values for the graphitisation process; the background level for this material is a little more than 0.2%. The combustion background is anthracite, for which we employ two

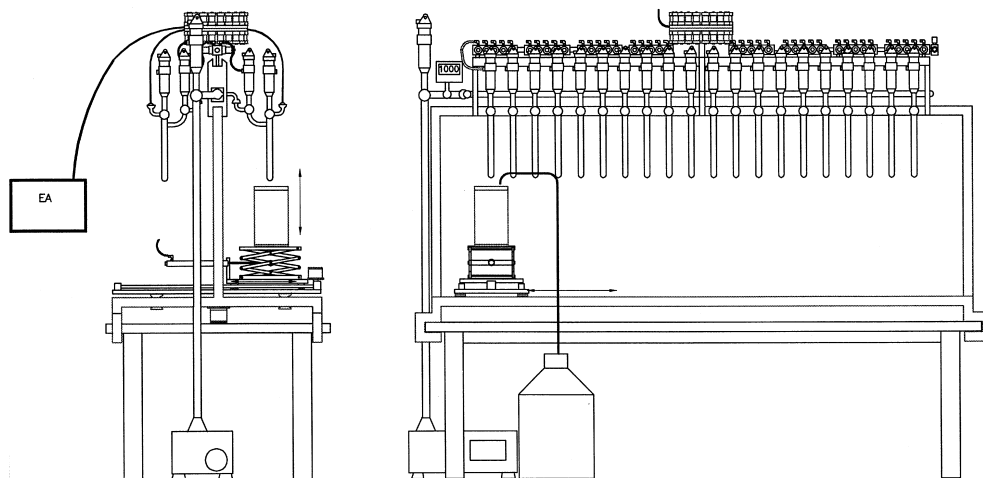


Fig. 4. Schematic view of the automatic cryogenic CO₂ trapping system.

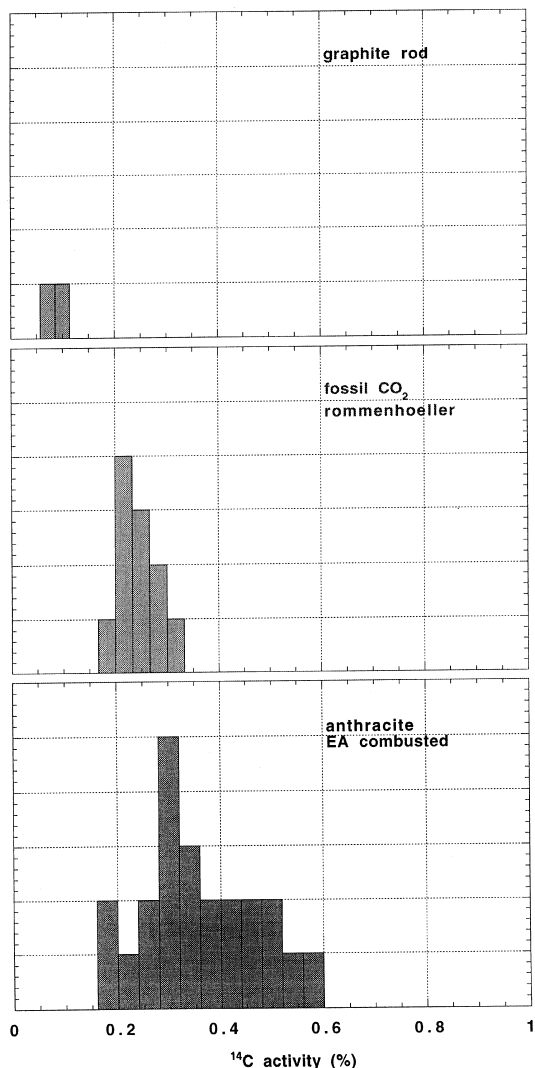


Fig. 5. Background measurements for the AMS proper (top), graphite system (middle) and combustion system (bottom).

batches. One is anthracite combusted by the conventional laboratory and used as a background for the proportional counters; this high-purity CO_2 can be trapped and transferred to the graphitisation setup. The second batch (used most of the time) is anthracite combusted by the EA, as used for all organic AMS samples. This combustion contributes apparently an extra 0.1–0.2% to the background. The background is an average 0.35%, corresponding to an age of 45 ka – the usual value

for combustion samples. It is also obvious (see Fig. 5) that the EA also introduces spreading. The backgrounds improve usually by replacing the combustion tube more often.

2.4. Precision: a case study with atmospheric samples

In general, precision for AMS systems are quoted as $<5\text{‰}$ for $^{14}\text{C}/^{12}\text{C}$, and $<2\text{‰}$ for $^{13}\text{C}/^{12}\text{C}$ isotopic ratios, respectively.

For ^{13}C , the precision for a typical batch of samples is 1‰ in $^{13}\delta$. The $^{13}\delta$ value is in practice a useful tool to indicate machine stability during the run. An example of a set of $^{13}\delta$ measurements where AMS results are compared with those from one of our stable isotope MSs is shown in Fig. 6. There is one outlier in ^{14}C due to fractionation during the graphitisation process.

For ^{14}C , we have investigated the possibility of improving measuring precision, in particular for ^{14}C in atmospheric CO_2 . To use atmospheric $^{14}\text{CO}_2$ for global carbon cycle studies the optimum in precision is preferred. Since such samples are ‘recent’, statistics is not a limiting factor for AMS.

A grand total of 176 atmospheric samples has been measured. A typical batch measurement consists of a run of 56 samples: 40 unknowns, 4 backgrounds and 12 standards. The samples were measured in 6 batches, of which 5 were measured duplo as well. There was one batch with small

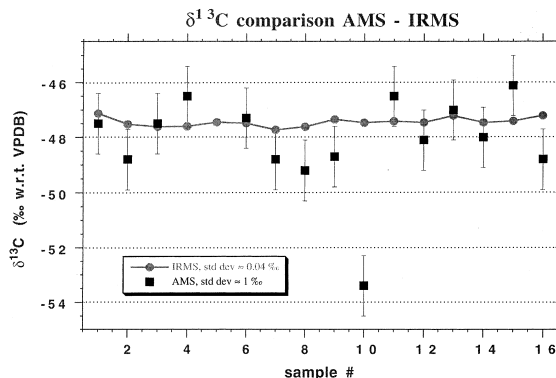


Fig. 6. Comparison between AMS measured $^{13}\delta$ and IRMS results for the same samples. There is one obvious outlier due to fractionation during the graphitisation process.

samples and not enough graphite to measure in duplo. Each batch included 12 standards: 4 ANU-sucrose (IAEA-C6) and 8 oxalic acid (Ox-1). The backgrounds were made from ^{14}C -free Rommenh oller gas. Including the duplicate measurements, the yield per sample is more than 300 000 ^{14}C counts. The contribution of statistics is below 2‰. The results for the standards, with emphasis on precision analysis, are summarised in Table 1.

Since the AMS measures $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratios, the value for the reference material has to be set in the analysis programme. In this case, the first standard is Ox-1 with a ^{14}C activity ratio (reported according to [21]) of 1.0398 and a $^{13}\delta$ value of $-19‰$. The error is the spread of the measured values around the literature value. The samples and second standard (C6) are measured relative to the first standard. The values for C6 are calculated and shown in the fifth column of Table 1.

Because of the way the measurements and primary analysis are performed, the values for the first standard (Ox-1, column 2) are the same of course, while those for the second standard (C6, column 5) show scatter. It is, however, more likely (thus having a higher probability in the statistical sense) that the measurements of both the calibration material (the standard, oxalic) and the reference material (sucrose) spread around their average value on a batch-to-batch basis. It is better to divide the scatter among the two standard ma-

terials, which can be done by introducing a ‘weighing factor’, based on weighted errors, and use a minimisation routine to find the best solution. The outcome of both Ox-1 and C6 measurements are then modified. This was done for each batch or wheel; the results are also shown in Table 1. The results are also plotted in Fig. 7(a).

In addition, we have more information: all but one batch has been measured twice. The minimisation also decreases the difference between both duplo runs. From this analysis we learn that the standard deviation for the 82 Ox-1 measurements is 3.6‰ and the standard deviation for the 41 C6 measurements is 4.9‰ (after removal of three clear outliers 3.9‰). These numbers are upper values. The residuals from the fitting function through the data are smaller (see Fig. 1). The data set also serves as an independent determination of the ^{14}C value for ANU-sucrose; the final value for the ^{14}C activity ratio of C6 (based on Ox-1) is 1.5074 ± 0.0024 . This corresponds well with the adopted value of 1.5061 ± 0.0011 [22].

In addition to the atmospheric data set described above, we analysed 9 other batches (part of the regular CIO measuring programme) which included two different reference materials: C6 and Ox-2. These batches were analysed the same way as described above. The results are shown in Fig. 7(b) and summarised in Table 2. In this case the first standard is C6 (about 8 per batch). The

Table 1
Results for standards (C6 and Ox-1) for 11 batches, including five duplo batches (d)

Batch	Ox-1	Error (1 σ)	Nr. of Ox-1	C6	Error (1 σ)	Nr. of C6	Modif. factor	Modif. Ox-1	Error	Modif. C6	Error
C036	1.0398	0.0021	8	1.5105	0.0030	4	0.9990	1.0387	0.0017	1.5089	0.0018
C036d	1.0398	0.0018	8	1.5033	0.0026	4	1.0014	1.0412	0.0015	1.5053	0.0015
C039	1.0398	0.0015	8	1.5114	0.0019	4	0.9986	1.0383	0.0012	1.5092	0.0011
C039d	1.0398	0.0014	8	1.5084	0.0018	4	0.9996	1.0394	0.0011	1.5079	0.0011
C040	1.0398	0.0030	8	1.5030	0.0043	4	1.0015	1.0414	0.0024	1.5053	0.0025
C040d	1.0398	0.0029	8	1.5041	0.0041	4	1.0011	1.0409	0.0023	1.5058	0.0024
C043	1.0398	0.0021	8	1.5047	0.0028	4	1.0009	1.0408	0.0017	1.5061	0.0016
C043d	1.0398	0.0015	7	1.5072	0.0018	4	1.0001	1.0399	0.0012	1.5074	0.0011
C053	1.0398	0.0020	5	1.5117	0.0024	3	0.9984	1.0381	0.0016	1.5092	0.0015
C053d	1.0398	0.0020	9	1.5066	0.0029	4	1.0002	1.0401	0.0016	1.5070	0.0016
C060	1.0398	0.0032	5	1.5073	0.0052	2	1.0000	1.0399	0.0027	1.5074	0.0028

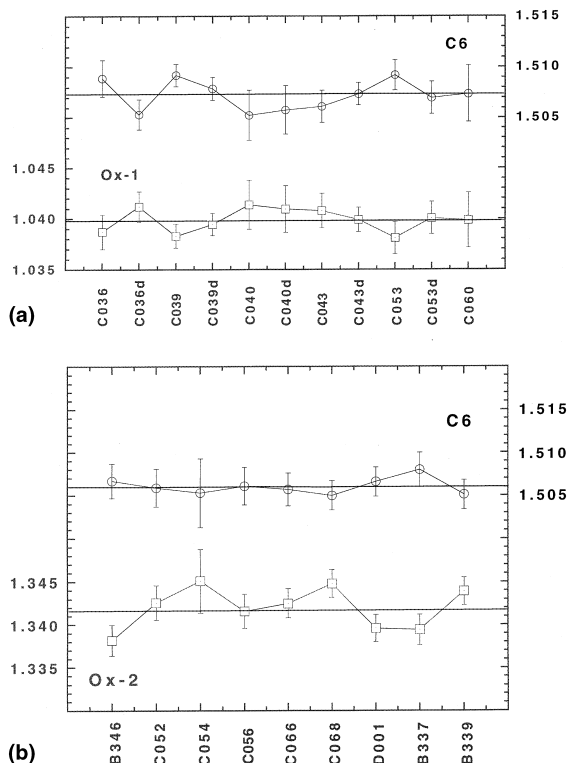


Fig. 7. (a) ^{14}C activity ratios for the standards C6 and Ox-1 used for the atmospheric data set (11 batches); (b) ^{14}C activity ratios for the standards C6 and Ox-2.

end result here is calculated for Ox-2: the ^{14}C activity ratio is $1.3417 \pm 0.0022\%$, based on analysis of 74 C6 and 25 Ox-2 measurements. This is the correct value within error.

Table 2
Nine batches for analysis of standards (C6 and Ox-2)

Batch	C6	Error (1σ)	Nr. of C6	Ox-2	Error (1σ)	Nr. of Ox-2	Factor	Modif. C6	Error	Modif. Ox-2	Error
B339	1.5061	0.0020	7	1.3448	0.0029	3	0.9994	1.5051	0.0017	1.3439	0.0016
B337	1.5061	0.0026	8	1.3377	0.0027	6	1.0013	1.5080	0.0020	1.3394	0.0018
D001	1.5061	0.0019	8	1.3391	0.0036	2	1.0003	1.5066	0.0017	1.3396	0.0016
C068	1.5061	0.0020	9	1.3458	0.0032	3	0.9992	1.5050	0.0017	1.3448	0.0016
C066	1.5061	0.0022	7	1.3428	0.0031	3	0.9998	1.5057	0.0019	1.3425	0.0017
C056	1.5061	0.0024	8	1.3416	0.0044	2	1.0000	1.5061	0.0022	1.3416	0.0020
C054	1.5061	0.0044	8	1.3458	0.0082	2	0.9994	1.5053	0.0040	1.3451	0.0037
C052	1.5061	0.0025	7	1.3428	0.0043	2	0.9998	1.5059	0.0022	1.3426	0.0020
B346	1.5061	0.0021	12	1.3377	0.0049	2	1.0004	1.5067	0.0020	1.3382	0.0018

The results for $^{13}\delta$ as measured by the AMS are as follows. For the atmospheric data set (Table 1), the mean for Ox-1 is set to -19% (by definition); the standard deviation is 1.4% . For C6, the mean value measured is $-10.6 \pm 1.7\%$. For the second data set (Table 2), the mean value for C6 is -10.44% (by definition) with a standard deviation of 1.1% . For Ox-2, the mean of the measured values is $-17.4 \pm 1.6\%$. We could have used the same ‘maximum likelihood’ approach to the ^{13}C values as we did to the ^{14}C values; however, the deviations between the ‘known’ $^{13}\delta$ values from stable isotope mass spectrometry and the AMS-determined ones are less than 0.5% , and the effects on the ^{14}C results would be minimal and insignificant.

We conclude that the final precision for this exercise is about 4% as a standard deviation for a single ^{14}C sample measurement. Our error calculations include scatter in both ^{14}C and ^{13}C measurements for the standards and the backgrounds, and statistics. This ‘precision barrier’ of 4% is apparently hard to break, given the large effort used in our experiment.

Concerning the C6 value determined here, we observe a peculiarity. Our measurement by AMS is 1.5074 ± 0.0024 , well in agreement with the adopted value of 1.5061 ± 0.0011 [22]. There have been complaints (not published) that the latter value is possibly too high; also, the Groningen conventional results (included in the IAEA value 1.5061) were low at the time of reporting. In the

meantime, our conventional measurements have been repeated in several counters; all measurements averaged together yields a value of 1.5022 ± 0.0007 . Actually, this low value was one of the reasons for determining an accurate measurement by AMS as well. We note that our measurements, both conventional and AMS, were done mostly for the same gas prepared in bulk by the large combustion/purification setup. The observed difference of 5‰, also observed by others, appears to be significant, is difficult to explain and will be the subject of further investigation, possibly involving intercomparison as well.

3. Conclusions and discussion

The Groningen AMS, a dedicated ^{14}C system, has been operational now for 5 years. A grand total of about 16 000 targets were measured during this time. Automatic batch measurements (maximum 58 targets per batch) are run around the clock. The simultaneous analysis of the 3 carbon isotopes is an on-line diagnostic tool, which checks the performance of the AMS continuously.

The combustion line has been automated employing a combined EA, MS and automatic cryogenic CO_2 trap.

Concerning precision, in a thorough series of runs with 3 standards (ANU-sucrose, Ox-1 and Ox-2) we have shown that for single standard deviation a limit of 4‰ is achieved at best. This precision barrier for close to modern samples is mainly caused by scatter of standards. This barrier is a hard one to break and includes all possible contributions: scatter in ^{14}C and ^{13}C of standards and backgrounds and statistics.

In the near future, work is planned on further automation of sample handling and on isotopic enrichment of AMS-size samples [23].

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