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The next chapter in radiocarbon dating at the Australian National University: Status report on the single stage AMS

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ABSTRACT

In February 2007, a single stage AMS from the National Electrostatics Corporation (NEC) was installed at the Research School of Earth Sciences at the Australian National University. This instrument is equipped with a gas/graphite hybrid ion source allowing direct analysis of CO_2 as well as traditional graphite targets. The instrument was funded by an Australian Research Council grant and by a consortium of eight universities plus the CSIRO Division of Land and Water. Its purpose is to support integrated environmental and archaeological research and access is open to the wider research community. In the first year ~1500 samples have been analysed. A precision of ~3‰ is routinely observed. The background for processed graphite (coal, calcite, Carrera marble) is better than 45,000 years. This paper summarizes the first year of measurements.

1. Introduction

A Single Stage Accelerator Mass Spectrometer (SSAMS) from National Electrostatics Corporation [1] has recently been installed at the Australian National University to complement the AMS activities that use the much larger 14UD accelerator. The instrument was delivered on January 17th, 2007 and accepted on March 7, 2007. Continuity with its predecessor, the ANU Radiocarbon Laboratory, which relied on beta counting and commenced in the early 1970s, is ensured by the SSAMS being housed in the former benzene-synthesis laboratory.

Our SSAMS system was the sixth built and is closely similar to the other SSAMS reported by Lund University and SUERC [2,3]. The stripper, analysing magnet, 90° electrostatic analyser (ESA) and detector are all mounted on a 250 kV high-voltage platform in air. Features that are specific to our instrument are:

(1) The single 40 sample ion source is followed by a 15° ESA based on the manufacturer's experience that backgrounds were higher in its absence.
(2) The ion source is modified from the traditional NEC design to incorporate several of the modifications described in Southon and Santos [4]. These include a new Cs oven and delivery system, a spherical ionizer, a cathode immersion lens, and a new extractor that is more open than the previous "snout" version [4] (Table 1).
(3) In addition, a gas handling manifold allows on-line analysis of CO_2 as well as graphite samples.

2. Results

The percent modern carbon (pMC) values for unprocessed and processed "blank" samples are shown in Fig. 1. The top panel shows both USGS coal that was combusted in sealed tubes and calcite that was hydrolysed by phosphoric acid in glass blood vials. These are closely similar and have an average value of 0.34 ± 0.05 pMC or 45,200 ± 950 years. The center panel shows "dead" CO_2 from the Kapuni natural-gas well in New Zealand. The average value of 0.16 ± 0.01 pMC, which equates to 51,500 years, is lower than for the coal and calcite samples that required a combustion or hydrolysis step in addition to graphitisation. The reasons for this are still under investigation. The bottom panel shows unprocessed "blank" graphite (a natural graphite that is pressed unprocessed into the sample holders), which has an average value of 0.09 ± 0.01 pMC or 55,600 years.

Results from known value standards (ANU sucrose, oxalic acid-II, oxalic acid-I, Tiri-K turbidite, and ICS2-cellulose) are shown in Fig. 2. In general there is very good agreement with the accepted values: the ANU SSAMS estimates of OX-I (average 104.1 ± 0.02 pMC, ~104 accepted), OX-II (average 134.5 ± 0.3 pMC, 1.341 accepted) and ANU sucrose (average 150.3 ± 0. 3 pMC, 150.6 accepted) are all within 3‰ (Fig. 2). The ICS2-cellulose and the Tiri-K turbidite are both within 50 years of their accepted value of 2250 and 18,155 years respectively (Fig. 2).

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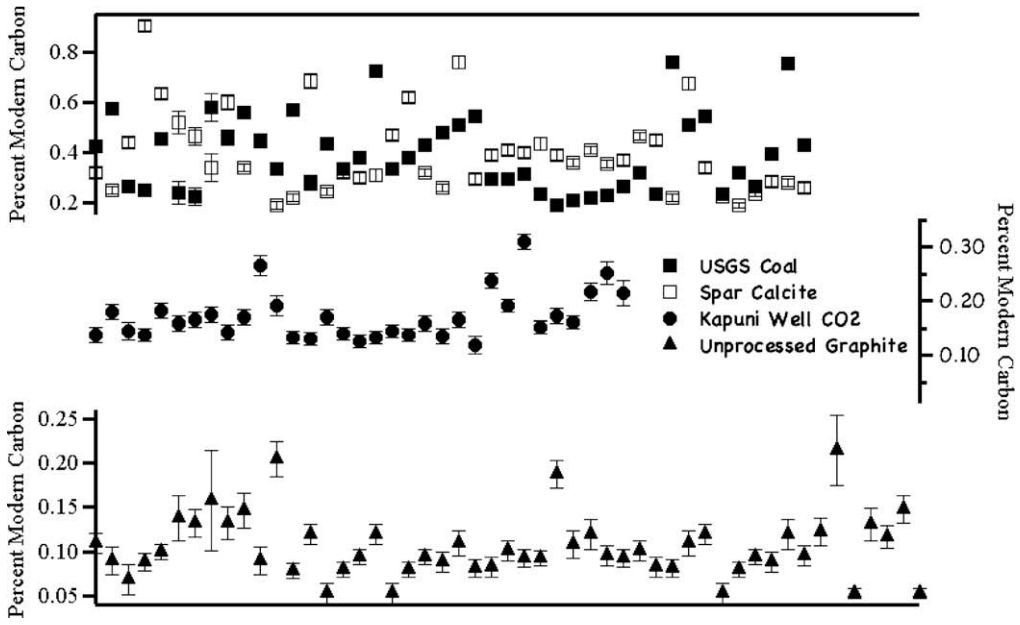

Fig. 1. Blank graphite samples both processed and unprocessed. The top panel shows the USGS combusted coal and hydrolysed spar calcite; the middle panel shows graphitised Kapuni well CO_2. The bottom panel shows unprocessed graphite.

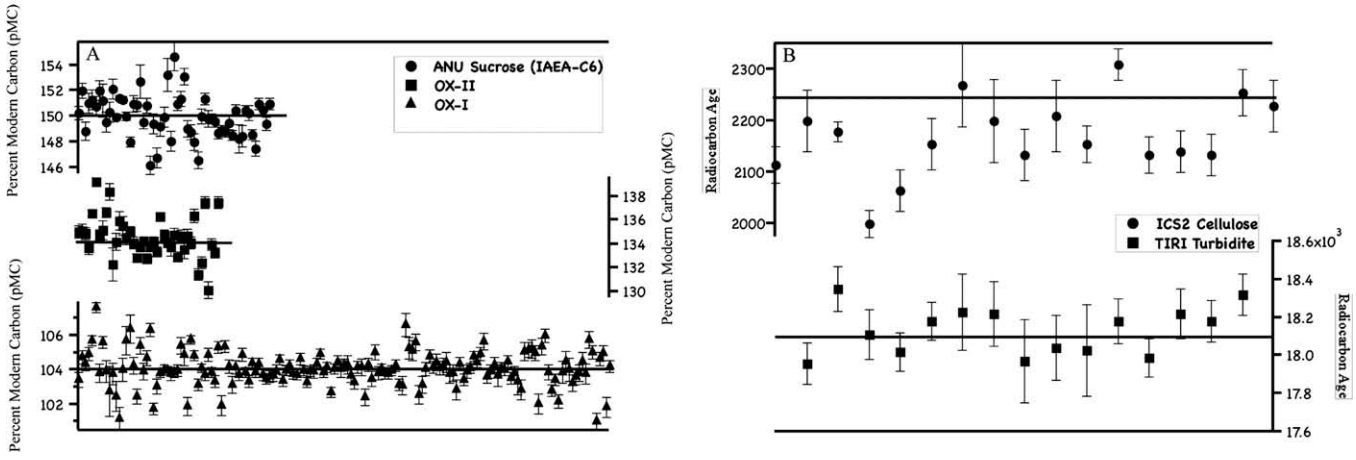

Fig. 2. (A) Results from primary radiocarbon standards (OX-I, OX-II, ANU sucrose) from March 2007 to present. (B) Results from secondary known value standards (ICS2-cellulose, Tiri-K turbidite). Lines show the "accepted" values.

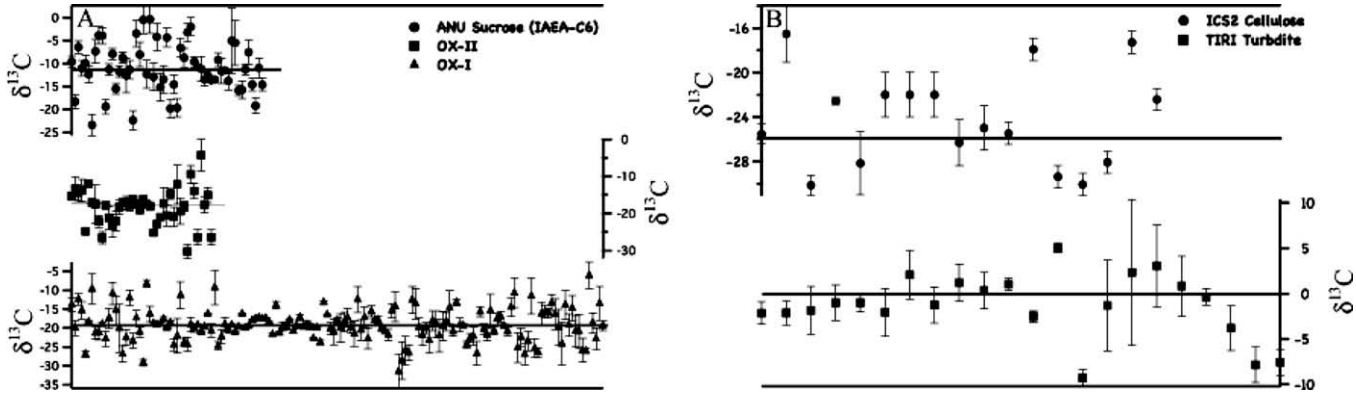

Fig. 3. (A) ANU SSAMS instrumental $\delta^{13}C$ from ANU sucrose, OX-II and OX-I. (B) ANU SSAMS instrumental $\delta^{13}C$ from ICS2-cellulose and Tiri-K turbidite. Lines show the "accepted" values.

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We also measure the $\delta^{13}C$ on-line; the ANU sucrose $\delta^{13}C$ average is $-11.1 \pm 0.9‰$ ($-10.8‰$ accepted), the OX-II average is $-17.5 \pm 1.1‰$ ($-17.5‰$ accepted), the OX-I average is $-19.3 \pm 0.7‰$ ($-19‰$ accepted) (Fig. 3A). The ICS2-cellulose average $\delta^{13}C$ is $-24.2 \pm 2.9‰$ ($-25‰$ accepted) and the Tiri-K turbidite average $\delta^{13}C$ is $1.1 \pm 2.1‰$ (0‰ accepted) (Fig. 3B). These results were obtained during many different sample wheels and tuning regimes. They also encompass a range of high-energy ^{12}C currents from 8 to 20 μA. Some measurements have larger uncertainities than others, this reflects the internal variability of measured $\delta^{13}C$.

Sample size-dependent fractionation was investigated using USGS coal and OX-I standards during two separate runs. Sample sizes varied from 0.02 mg C to 1 mg C. Currents of $^{12}C^-$ beam begin to decrease (Fig. 4) only when samples are less than 0.4 mg C. The $\delta^{13}C$ is stable until ~0.2 mg C when it becomes more positive with decreasing sample size (Fig. 4). If we keep our samples sizes within the 0.4–1 mg C range we do not need sample-specific sized standards to normalize our data. The cause of the increasing $\delta^{13}C$ with decreasing sample sizes (below 0.2 mg C) is still being explored.

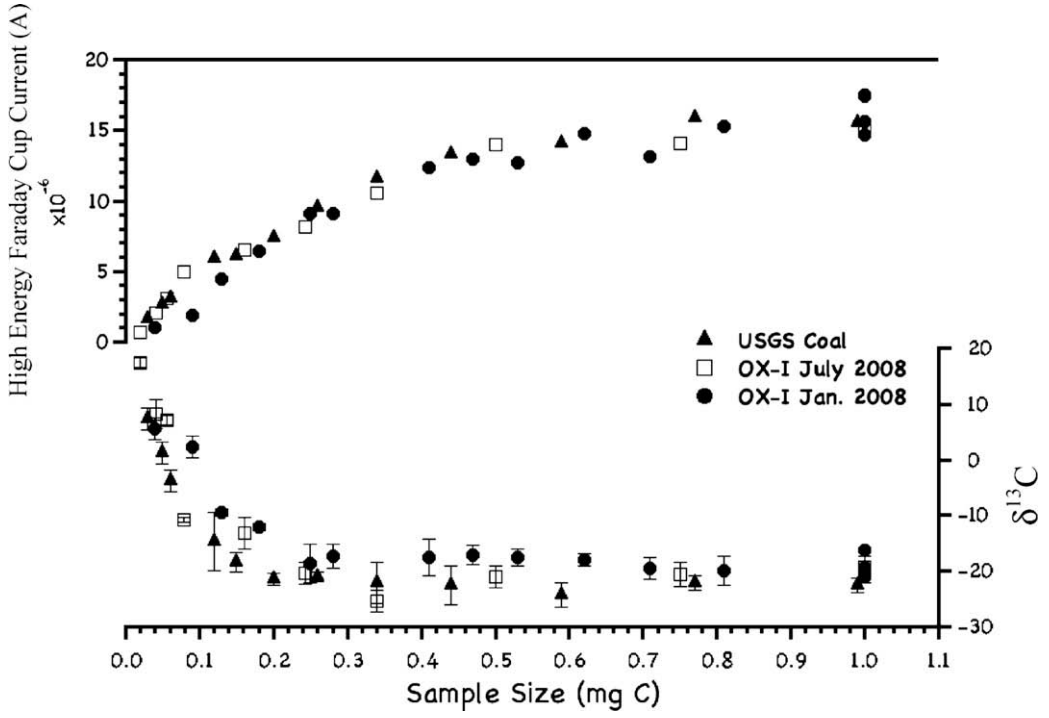

Fig. 4. High-energy $^{12}C^+$ current and $\delta^{13}C$ vs. sample size for USGS coal and OX-I samples.

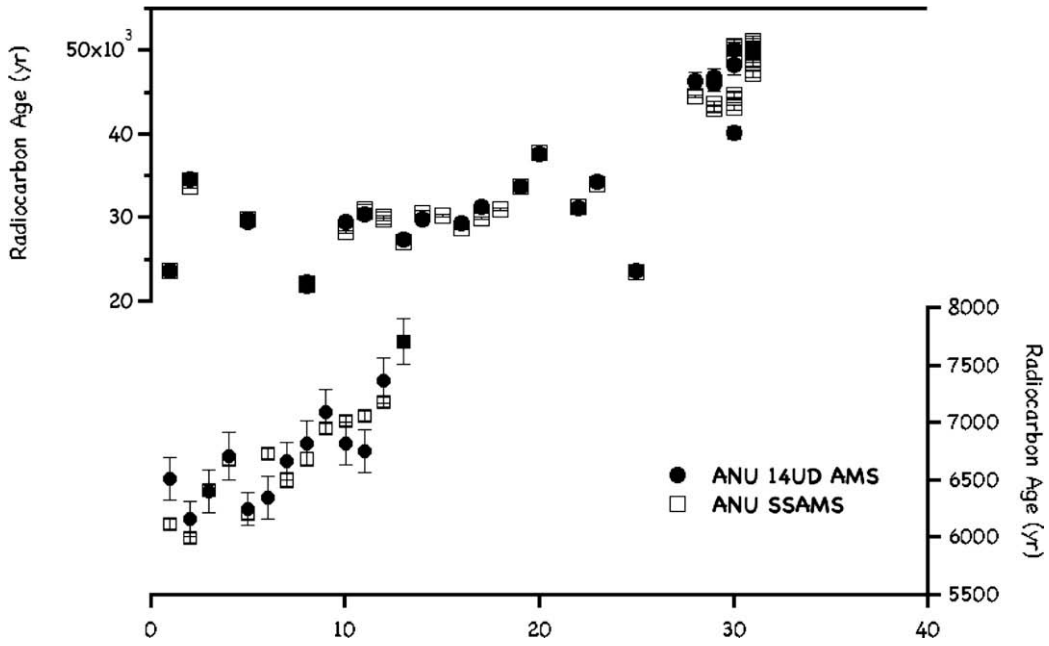

Fig. 5. Comparison of samples between ANU 14UD AMS and ANU SSAMS. In the top panel samples weighing 1.5 mg C were split into two cathodes and run on both machines. In the bottom panel sample cathodes were first run on the 14UD AMS and then the same cathode was analysed by the SSAMS.

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Table 1
Typical source operating parameters.

Cathode voltage	−7 kV
Extraction	−18 kV
Focus (einzel) voltage	1.5 kV
Source bias	−37.5 kV
Cs oven heater voltage	34 V
Cs oven temperature	92–95 °C

Over the past year we have also run comparison samples on the ANU 14UD AMS system. Samples corresponding to 1.5 mg C were graphitised and the graphite was split into two cathodes. These splits were run on both the SSAMS and the ANU 14UD. The agreement between the two machines is good out to 50 kya; backgrounds have not been subtracted from these samples. Only a few sample results lie outside of the 1-σ error envelope (Fig. 5).

3. Conclusions

The ANU SSAMS provides a high-throughput (30 unknowns in 30 h) with ~4‰ reproducibility and counting statistics. We use the instrument $\delta^{13}C$ for sample age correction. Within a sample size range of 0.4–1 mg we obtain accuracies within 3‰ for radiocarbon ages less than 10 ka. Our background for fully-processed samples is ~45,000 years, while the machine blank for unprocessed graphite is ~55,600 years. We plan further investigations to improve our background as well as understanding fractionation at lower/higher beam currents.

Acknowledgement

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