High throughput, high precision ^{14}C AMS with a small accelerator

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Abstract. The costs of Accelerator Mass Spectrometry (AMS) systems for radiocarbon measurements have dropped sharply in recent years, but at $1M to $1.3M, these instruments still represent major investments. However, these costs are not excessive for a facility producing several thousand high precision ^{14}C analyses per year. Here we describe methods developed at the AMS laboratory at University of California Irvine (UC Irvine) for achieving high throughput at high precision. Key factors include: large-scale parallelism in sample preparation plus streamlining of sample pretreatment procedures; development of a robust and easily maintainable high output ion source; removal of data analysis bottlenecks; and training of laboratory personnel to ensure that several staff members can carry out critical functions.

1. Introduction

The Keck Carbon Cycle AMS laboratory at University of California Irvine (UC Irvine) [1] was set up in 2001 - 2002 to use radiocarbon AMS to help understand the carbon cycle and its interactions with the climate system. The AMS system (Fig 1) is based on a National Electrostatics Corp. (NEC) 1.5SDH-1 0.5MV pelletron with 40-sample MC-SNICS ion source. Three years after installation, the system routinely measures 500 unknowns per month, measurement precisions are typically 2-3‰, and backgrounds for graphitized coal and calcite samples are equivalent to radiocarbon ages in excess of 55,000 years.

1. SAFETY SCREEN
2. INJECTOR TERMINAL
3. MC SNICS ION SOURCE
4. EXTRACTOR FOCUS
5. PREACCELERATION TUBE
6. ISOLATION VALVE
7. CRYO PUMP STATION, FARADAY CUP & X-Y STEERER
8. BEAM PROFILE MONITOR
9. MAGNET CHAMBER INSULATOR
10. DRIFT TUBE
11. INJECTOR MAGNET ME/z^2=2, 12" R
12. TURBO PUMP
13. INJECTOR OFFSET FARADAY CUP
14. CHAIN DRIVE MOTOR
15. PRESSURE TANK
16. PELLETRON COLUMN
17. ACCELERATION TUBE
18. PELLETRON CHAIN
19. TERMINAL IDLER
20. TURBO PUMPED GAS STRIPPER
21. CRYO PUMP STATION, BPM, Y-STEERER
22. ANALYZING MAGNET ME/z^2=18, .75m R
23. ANALYZING OFFSET FARADAY CUP
24. FARADAY CUP
25. SPHERICAL ELECTROSTATIC ANALYZER .75M RADIUS WITH CRYO PUMP STATION
26. PARTICLE DETECTOR
27. HIGH VOLTAGE TERMINAL
28. BELLOWS
29. Y STEERER
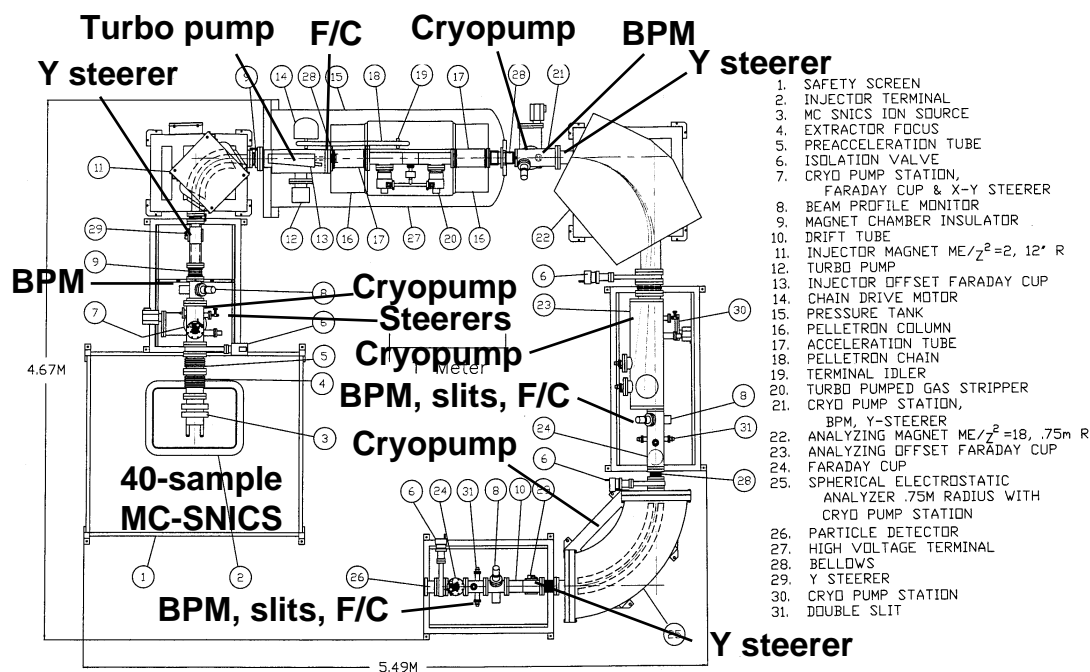30. CRYO PUMP STATION
31. DOUBLE SLIT

FIG. 1. The UC Irvine AMS spectrometer. The overall dimensions of the system are 4.7 x 5.5 m. (BPM = Beam Profile Monitor, F/C = Faraday Cup).

Before our system was purchased, other researchers [2,3,4] had shown that these small commercial spectrometers were capable of high precision measurements, and importantly, that the NEC hardware and software systems would run reliably overnight unattended. However, as described below, several additional factors are critical for satisfying the twin requirements of high throughput and high precision.

2. Sample Preparation

Large-scale parallelism in sample preparation coupled with streamlining of sample pretreatment procedures is essential for high throughput and rapid sample turnaround. We currently operate three sample preparation laboratories at UC Irvine; one associated with the AMS laboratory itself plus two others that support major programs using the spectrometer. Each laboratory has an area of 100 - 150 m^2, comparable with the size of the room housing the AMS spectrometer itself.

Present sample processing capabilities include simple acid-alkali-acid treatment and sealed tube combustion of organics; leaching and hydrolysis of carbonates, cellulose extractions, collagen extraction from bone, and extraction of CO_2 from air samples and from seawater. Other methods currently under development by researchers and students include Dissolved Organic Carbon extraction from seawater (UV oxidation) and separation of simple compound classes (lipids, carbohydrates, etc) from soils and other organic materials. A preparative capillary gas chromatograph plus cryogenic trapping system for isolating single compounds from sample matrices will be purchased shortly.

We have simplified and streamlined the preparation procedures for routine samples wherever possible. For example, samples are processed in parallel in large batches, reagents are purchased ready-mixed, and all acid and base treatments are carried out in disposable 13mm test tubes. Time-consuming centrifugation can be avoided in many cases by careful use of ultra-fine tip pipettes for removing supernatant liquids. Carbonate samples are leached and hydrolyzed in disposable septum-sealed 3ml Vacutainer blood collection vials, using disposable hypodermic needles to introduce and remove gases [5]. Sample packing procedures are simple, avoiding the use of presses and other elaborate loading facilities: prepared graphite samples are tamped by hand into sample holders using a piece of drill rod which is then hammered to compress the graphite.

Our twin 12-reactor hydrogen reduction lines for graphite production from CO_2 (Fig 2) can routinely produce 48 samples per day, for samples from 1mg down to a few micrograms. An unusual feature of the reactor design is the use of magnesium perchlorate to absorb water from the graphitization reaction [5], avoiding the need for elaborate cryogenic or thermoelectric cooling apparatus or frequent filling of cold traps. The perchlorate is used three times and then discarded, well before saturation.

We also produce graphite by zinc reduction in sealed quartz tubes in a batch processing mode [6], with a capacity of about 40 samples per day. In our laboratory, this method is used primarily for near-Modern samples, but it is noteworthy that
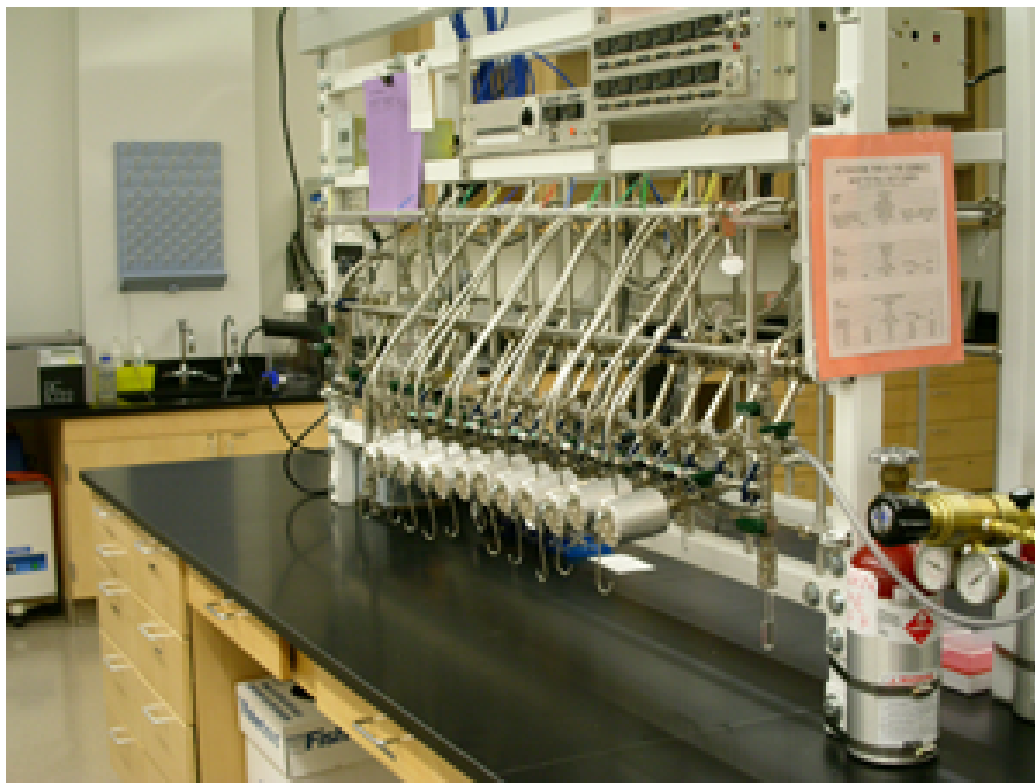
FIG. 2. One of two 12-reactor graphitization lines in the sample preparation lab. Reactor temperature controllers and pressure monitoring readouts are mounted in the rack above the line.

backgrounds for 1 mg samples (without on-line zinc distillation or other elaborate pretreatment) are 50,000 ^{14}C years BP. Precisions are as good as those achieved with hydrogen–reduced graphite, and ion source outputs are only slightly lower.

In addition to the samples produced in-house, we measure an additional 1500-2000 samples per year (including standards and blanks) that are received as prepared graphite in sample holders from two other universities and a commercial radiocarbon laboratory. Provided throughput at external sample facilities is sufficiently high that processing continuity and therefore quality are maintained, the use of external graphite preparation laboratories is a very cost-effective way of ensuring that an AMS spectrometer is used efficiently.

3. Ion Source

In modern AMS spectrometers, the beam transport efficiency (ions detected as a fraction of ion source output) is determined primarily by the electron stripping efficiency to a given charge state, plus beam losses in the high energy acceleration tube due to scattering in the stripper. Typical radiocarbon spectrometer efficiencies range from about 35% to 50% [7]: in other words, transport efficiencies span a rather narrow range of about 1.5:1. In contrast, ion source outputs of C$^-$ reported recently [7] vary by more than a factor of 30, from 5-10 μA to over 200 μA, indicating the overwhelming importance of the source for overall system performance. Ion source upgrading has therefore been a major part of our in-house development program.
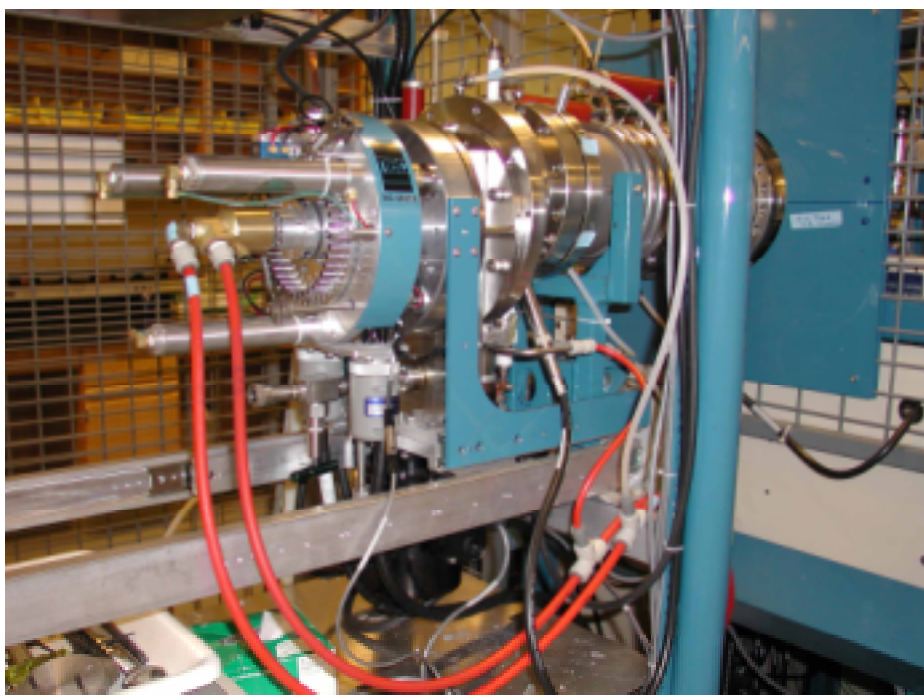
FIG. 3. The MC-SNICS ion source, plus the track system built into the source high voltage rack for in-place servicing.

3.1 Reliability issues

Our initial upgrades to the NEC source were primarily for improving reliability and serviceability [8]. A sliding track system was built into the existing power supply rack to allow the source to be rolled back from the beamline for servicing in-situ (Fig 3). The Cs oven and delivery tube were redesigned so that the delivery line was heated by conduction from the ionizer assembly and was vacuum-insulated throughout. This reduced Cs consumption by 50% and completely eliminated clogging of the delivery tube. Changes to the geometry of the ionizer assembly and extraction electrodes eliminated most of the arcing problems in those regions; and cooling the downstream end of the source housing trapped more stray Cs in the source body and further reduced extractor arcing. Problems of sparking from Al sample wheels at source startup were eliminated, first by improved cleaning procedures and subsequently by adoption of a new copper and stainless steel sample wheel design.

3.2 Vacuum improvement

The standard NEC extraction/einzel lens assembly limits the pumping conductance between the source and downstream vacuum pumps. We rebuilt the internal extractor and lens electrodes with a more open structure, and replaced all aluminum parts with stainless steel for ease of cleaning. We also replaced the NEC preacceleration tube downstream of the extraction assembly with a larger fully shielded Ceramaseal insulator. The conductance increased by a factor of three, leading to a large reduction in the pumpdown time required after sample wheel changes. Previously, high outputs could only be achieved after 30-60 minutes of pumping (turning the ionizer and Cs
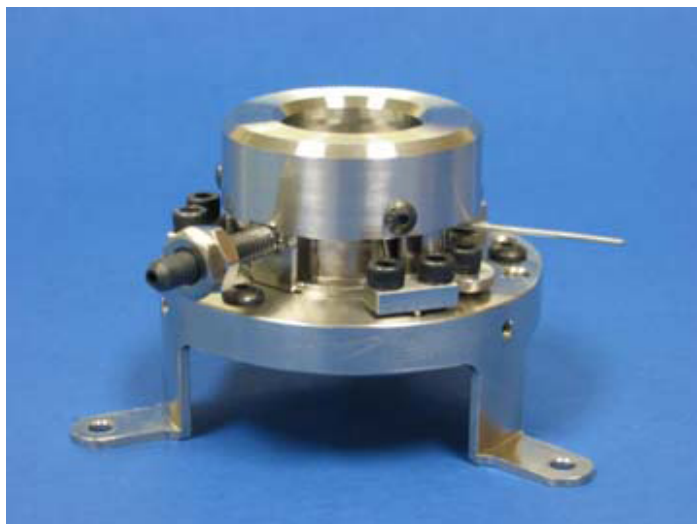
FIG. 4. Housing for the Spectramat ionizer. Cs vapor enters through the hollow stud on the left and fills a circular gallery at the top of the housing. Six apertures direct jets of vapor on to the spherical ionizing surface in the interior of the assembly. The ionizer heater lead appears at the right. The overall diameter of the base is 6.5 cm.

oven on earlier had little effect). After these changes, the source can be turned on almost immediately.

3.3 Spherical ionizers

Spherical ionizers provide inherently better Cs focusing than the standard NEC conical design [9,10]. We have replaced the conical ionizer with a new assembly based on the spherical Heatwave/Spectramat ionizers used in the Lawrence Livermore [10] and High Voltage Engineering Europa (HVEE) sources (Fig 4). The Cs focus electrode in the standard source was replaced by an immersion lens at cathode potential, eliminating the Cs focus power supply and providing improved local pumping as well as increasing clearances between electrodes in the critical central region of the source. We have also begun testing spherical ionizers built by NEC, with electrode geometries similar to those of Hausladen et al. [9], but incorporating a cathode immersion lens. C⁻ currents of over 150μA have been observed in initial tests, suggesting that performance will be similar to that achieved with the Spectramat assembly.

3.4 Source performance

The MC-SNICS/Spectramat combination routinely delivers C⁻ outputs of 150 μA at sputtering voltages of 7.5kV, sufficient to measure a wheel of 40 samples to 2-3‰ in less than 24 hours. Turnaround times after a wheel change are less than one hour, and a complete source cleaning (required every few weeks) takes 6 hours or less, including pumpdown and outgassing time.

We have found that the well-focused Cs beam from the spherical ionizer drills a remarkably straight-sided flat-bottomed hole into the sample, so that source output is maintained until essentially all of the sample material is consumed. On large (1mg) test samples run to extinction, we have measured over 10% of the ^{14}C atoms present in

the samples, corresponding to source efficiencies above 20%. Importantly, source efficiencies are 10-15% for 5-10μg carbon samples, indicating that high precision (<1%) measurements on ultra-small samples may be possible, though subject to resolution of serious sample preparation and background issues.

4. Data analysis

Removal of data analysis bottlenecks is another prerequisite for high throughput. We use the Lawrence Livermore "Fudger" AMS analysis code (T.Ognibene, pers. comm.) as our primary analysis tool, but the stripchart recorder module within NEC's equivalent "abc" software is used routinely, for detailed scanning of data from individual measurement runs to determine the causes of anomalies.

Typically, the 40 samples in a wheel (25 unknowns plus primary and secondary standards and process blanks) are each measured 8-10 times over 24 hours. Data analysis begins with a quick visual scan of the data from all 300 – 400 measurement runs to detect anomalies due to accelerator or ion source sparks or other problems. These runs are considered lost and are eliminated: no attempt is made to cut out "bad" regions within a given run. The results are then corrected for isotopic fractionation and normalized to a set of six or more aliquots of the OX1 radiocarbon standard, using a running mean of the standards to normalize out any time-dependent drifts. The normalized results are then pasted into an Excel spreadsheet where backgrounds are subtracted to produce the final data. The entire process is repeated by a second analyst and the results (which are usually consistent within 1‰ on modern samples) are compared before any data is released. In favorable cases the total analysis time can be well under an hour, so that in many cases, results are available just one to two days after the samples are run.

4. Staff training

A fourth major component leading to high throughput has been personnel training to ensure that most laboratory functions can be carried out by at least two, and preferably several, staff members. We currently have ten people (including students) who can prepare samples, eight who can analyze data, and six who can change sample wheels and tune and operate the spectrometer. This duplication has allowed us to keep up with the high sample throughput while maintaining quality control procedures such as independent analyses of all data by two researchers. Similarly, two members of the research group have sufficient expertise to independently supervise complete ion source rebuilds and repairs for all but the most severe spectrometer breakdowns.

5. Conclusions

We have shown that a small commercially available AMS system can produce thousands of high precision ^{14}C analyses per year. Other than ion source improvements, relatively few hardware changes are required. However, extensive sample preparation facilities plus a relatively large group of well trained personnel are essential for using such an instrument efficiently.

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