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# Accelerator Mass Spectrometry for Measurement of Long-Lived Radioisotopes

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Particle accelerators, such as those built for research in nuclear physics, can also be used together with magnetic and electrostatic mass analyzers to measure rare isotopes at very low abundance ratios. All molecular ions can be eliminated when accelerated to energies of millions of electron volts. Some atomic isobars can be eliminated with the use of negative ions; others can be separated at high energies by measuring their rate of energy loss in a detector. The long-lived radioisotopes <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>129</sup>I can now be measured in small natural samples having isotopic abundances in the range  $10^{-12}$  to  $10^{-15}$  and as few as  $10^5$  atoms. In the past few years, research applications of accelerator mass spectrometry have been concentrated in the earth sciences (climatology, cosmochemistry, environmental chemistry, geochronology, glaciology, hydrology, igneous petrogenesis, minerals exploration, sedimentology, and volcanology), in anthropology and archeology (radiocarbon dating), and in physics (searches for exotic particles and measurement of halflives). In addition, accelerator mass spectrometry may become an important tool for the materials and biological sciences.

ADIOISOTOPES HAVE LONG BEEN USED AS AN IMPORTANT (and sometimes the only) source of information regarding the chronology of geological processes, the history of meteorites and cosmic rays, human evolution, and the dynamics of biological systems. Short-lived radioisotopes, with half-lives less than about 1 year, can usually be measured with high sensitivity by conventional techniques, in which decay products are counted efficiently. Primordial radioisotopes with half-lives greater than

about 10<sup>9</sup> years are relatively abundant naturally (since they have not completely decayed over the life of the solar system) and are used for dating by measuring the buildup of stable decay products. Radioisotopes with half-lives in the intermediate range of  $10^3$  to  $10^8$ years are difficult to measure with decay counting since only a small fraction of the atoms decay over a reasonable counting period of a few months or less. Many of the interesting processes that occur on the earth and in the solar system have time scales that fall in this interval. Over 30 elements have radioisotopes with half-lives in this range; five of these (Table 1) have now been measured with the new technique of accelerator mass spectrometry (AMS) with enough sensitivity for detection at natural levels. Although in some cases decay counting of large natural samples has been possible, sample sizes have been reduced by several orders of magnitude. For example, <sup>36</sup>Cl can be measured by conventional decay counting with 40 g of chloride (1) extracted from thousands of liters of contemporary water, whereas AMS requires only 1 mg of chloride and has backgrounds 10 to 100 times lower than those of conventional techniques.

Accelerator mass spectrometry can also be used to measure trace elements directly in unprocessed materials with backgrounds far lower than is possible with conventional techniques. Microprobe ion sources have been developed with 1-µm spatial resolution; these should be very useful in conjunction with AMS (2). Stable-isotope ratios of trace elements can also be measured with high sensitivity (3)

Although AMS had its beginnings in 1939 with the measurement of <sup>3</sup>He in helium at natural abundances by Alvarez and Cornog (4),

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recent work was triggered in 1977 by Muller, who suggested that a cyclotron could be used for detecting <sup>14</sup>C, <sup>10</sup>Be, and other long-lived radioisotopes (5), and independently by the Rochester group (6) who demonstrated that <sup>14</sup>C could be separated from the isobar <sup>14</sup>N by relying on the instability of the negative ion <sup>14</sup>N<sup>-</sup>. Publications reporting accelerator measurements of natural <sup>14</sup>C at McMaster University (7) and the University of Rochester (8) appeared later in 1977, and these were soon followed by reports on <sup>10</sup>Be measurements at Grenoble (9) and <sup>36</sup>Cl measurements at Rochester (10). Presently AMS measurements are being made at 20 accelerator laboratories around the world, and one-third of these are dedicated to AMS measurements of long-lived radioisotopes.

In this review we first point out the key advantages of AMS over conventional (low-energy) mass spectrometry, and then we describe major components of the apparatus, the analytical procedure, and the limitations. We limit most discussion to one type of accelerator (the tandem electrostatic accelerator) and to five radioisotopes (Table 1). The second part of the review outlines current scientific research with AMS. Earlier reviews and conference proceedings contain additional detail on various aspects of AMS such as history of the technique (11), fundamentals (12–16), <sup>14</sup>C (13, 17–20), <sup>10</sup>Be (21), and <sup>36</sup>Cl (22). The references in Table 1 give details of the detection methods for the various isotopes.

## **Principles of Operation**

Mass spectrometry (MS) is an analytical method that makes use of the face that each atomic and many molecular species have unique masses. MS typically involves four separate steps: (i) the formation of a charged atomic or molecular ion; (ii) acceleration through an electrostatic (ES) potential of a few kilovolts; (iii) separation of the ions based on their mass-to-charge ratio using some combination of the four basic analysis stages (Table 2); and (iv) measurement of the number of ions per second in a detector (or the electric current in a Faraday cup). AMS uses these four steps but includes an acceleration to MeV (megaelectron volt) energies. As we will show, high energies together with the analysis of negative ions allow the separation and measurement of isotopes at abundance ratios far lower than possible at low (kiloelectron volt) energies.

Although MS is a general, powerful, and widely used analytical method (23), its sensitivity is severely limited by the need for high mass resolution. Mass differences between isobars (atoms or molecules having the same mass number M) can be very small (the required mass resolution is  $M/\Delta M = 84,000$  for  $^{14}C^{-14}N$ ). In such cases isotopes can be resolved with sector MS (24) only when very narrow slits are used, and this results in low efficiency for the transmission of ions through the instrument. This approach is self-defeating if the isotope is very rare; AMS obtains much better sensitivity by opening the slits wide for maximum transmission and finding other ways to separate atomic and molecular isobars. AMS must deal with the following difficulties when mass resolutions are in the range  $M/\Delta M = 200$  to 400:

Scattering. Interference from ions that scatter from surfaces and ions that change charge in the residual gas of the vacuum system can be reduced to any desired level by adding multiple stages of mass selection as listed in Table 2. Acceleration to high energy, although helpful for reducing the angular divergence of the beam (12), is not necessary to remove this type of interference.

Molecular interference. There are many types of mass spectrometers, some with excellent vacuum and very clean sample handling

**Table 1.** Information about radioisotopes measured routinely at natural levels with AMS. Details are given in the references as follows:  ${}^{10}Be$  (32),  ${}^{14}C$  (95),  ${}^{26}Al$  (93),  ${}^{36}Cl$  (10),  ${}^{129}I$  (29). AMS has also been demonstrated for the following radioisotopes:  ${}^{32}Si$  (88, 89),  ${}^{41}Ca$  (34),  ${}^{59}Ni$  (35),  ${}^{60}Fe$  (41), and the stable isotopes isotopes (3), Pt, Ir (83), B, P, and Sb (52). The background value is the radioisotope ratio obtained for a blank: a sample with negligible radioisotope content that is prepared identically to the unknown sample. Ion-source efficiency is the fraction of atoms in the sample that are extracted as negative ions and measured in a Faraday cup at the image of the injector magnet for a sample that is completely consumed. The overall efficiency for a sample completely consumed is the product of the ion-source efficiency, stripper yield, and accelerator efficiency. The run time is the time necessary to count 100 radioisotope events in the detector for a sample that has an isotope ratio several times the background value. The decay counting interval is the time required to count 100 radioactive decay events for the same number of atoms placed in a detector that is 100% efficient.

	Radioisotope				
	<sup>10</sup> Be	<sup>14</sup> C	<sup>26</sup> Al	<sup>36</sup> Cl	<sup>129</sup> I
Half-life (years)	$1.6 \times 10^{6}$	5730	$7.05 \times 10^{5}$	$3.0 \times 10^{5}$	$1.57 \times 10^{7}$
Stable isotopes	<sup>9</sup> Be	$^{12}C, ^{13}C$	<sup>27</sup> Al	<sup>35</sup> Cl, <sup>37</sup> Cl	<sup>127</sup> I
Stable isobars	<sup>10</sup> B	<sup>14</sup> N*	<sup>26</sup> Mg*	<sup>36</sup> Ar*, <sup>36</sup> S	<sup>129</sup> Xe*
Chemical form <sup>†</sup>	BeO	C‡	$Al_2O_3$	AgCl	AgI
Terminal voltage (MV)	7.3	2	7.5	8	5
Charge state	3	3	7	7	5
Energy (MeV)	24.7	8	60	64	30
Sample size (mg)§	0.2	0.25	3	2	2
Background ( $\times 10^{-15}$ )	3	7	1	2	20
Ion source current (µA)	12	7	1.5	8	3
Ion source efficiency	$5  imes 10^{-3}$	≈0.05	$2.5 imes10^{-3}$	≈0.03	≈0.01
Stripper yield	0.54	0.42	0.35	0.32	0.10
Accelerator efficiency	0.28	0.71	0.43	0.18	0.23
Overall efficiency	$7.5 imes10^{-4}$	$1.5 imes10^{-2}$	$3.8 imes10^{-4}$	$1.7  imes 10^{-3}$	$2.3 imes10^{-4}$
Run time (minutes)	10	7	40	30	20
Atoms per sample¶	$2 imes 10^5$	$2 imes 10^5$	$4  imes 10^5$	$5  imes 10^5$	$2 imes 10^{6}$
Decay counting interval (years)	1100	3	250	86	1130

\*These isotopes do not form atomic negative ions. †Atomic ions are accelerated except in the case of <sup>10</sup>Be where the molecular ion BeO<sup>-</sup> is accelerated to the terminal (BeO<sup>-</sup> is produced more efficiently than Be<sup>-</sup>). ‡Negative ions of carbon are formed most efficiently when the sample is in the form of graphite or CO<sub>2</sub> gas. Several methods (96) have been developed for converting small carbon samples into forms suitable for the cesium sputter ion source. <10% precision. Carrier material of the same element, with negligible radioisotope, is added if needed to bring the sample weight up to that given in the table. Sample size larger than those listed are easier to handle and may result in better precision. In some cases a binder such as silver powder is mixed with the sample but this is not included in the weight given. IlSomewhat more counting time is needed to obtain 10% precision since uncertainties in the background and standard must be included. A modern sample (<sup>14</sup>C/ <sup>14</sup>C  $\approx 10^{-12}$ ) can be measured to within 1% in 1 hour (95). The number of atoms of radioisotope in the original sample needed for 10% precision. equipment. However, all mass spectrometers show a background of peaks at nearly every integral mass unit that arise from unavoidable molecular contamination. Heroic attempts have been made in special cases to reduce molecular interference to below the  $10^{-12}$  level without success (25). Acceleration to high energies offers the possibility of removing molecular interference entirely. When ions at MeV energies pass through a thin foil or gas, several electrons are stripped off. This process is violent enough to dissociate most but not all molecules (26). Molecules that survive in charge states higher than 2+ are unbound and decay rapidly; if the stripper is followed by a mass/charge analyzer that selects ions in a charge state of three or greater, only atomic ions will pass.

The number of electrons stripped off is described by a statistical model that predicts a bell-shaped distribution of ion intensities with a width of two to three electrons. The energy needed for maximum yield depends on mass: for a gas stripper, 3-MeV carbon and 6-MeV uranium ions will yield, respectively, 50% and 25% in charge state 3+. For reasons noted below it is often desirable to choose a higher charge state, requiring higher energies. Thin (2 to 5  $\mu$ g/cm<sup>2</sup>) carbon foil strippers are sometimes used to obtain higher charge states, but problems for projectiles with high atomic number include multiple scattering (increases angular divergence), unstable transmission, and short foil lifetimes.

Since ion-beam analysis can select only the mass-to-charge ratio (Table 2), with moderate mass resolution ( $M/\Delta M = 100$  to 1000) molecular fragments with lower mass but nearly equivalent mass-to-charge ratio may not be rejected. For example,  ${}^{12}C^{1+}$  would interfere with detection of  ${}^{36}Cl^{3+}$ . For this reason charge states greater than 3+ must sometimes be selected in order to find one such that the mass number and charge state have no common factors. At MeV energies the total energy of the ion can be measured in the detector, and this, together with selection of the energy-to-charge ratio (Table 2), allows determination of the charge state. This is sufficient for separating  ${}^{7}Li^{2+}$  from  ${}^{14}C^{4+}$  but in some cases (such as for  ${}^{36}Cl^{3+}$ ) count rates of the interfering ions would be excessive.

*Isobars.* Discrimination against stable atomic isobars (Table 1) is the most difficult problem for AMS, and a combination of techniques must be used. The first is chemical preparation of the sample. Several milligrams of material in the chemical form listed in Table 1 are purified to remove as much of the isobar as possible. Relative impurities of  $10^{-6}$  to  $10^{-8}$  are a practical limit, and at this level there are usually significant contributions in the ion source from the residual gas or apertures. Additional separation methods are needed to reach the  $10^{-12}$  to  $10^{-15}$  level.

As noted below, tandem accelerators require negative ions in the first acceleration stage. Mass spectrometry of negative ions is a powerful method for rejecting isobars. The negative ion is not stable for some atoms and molecules (including N, Mg, KH<sub>3</sub>, and noble gases), and this has made possible AMS of <sup>14</sup>C (8), <sup>26</sup>Al (27), <sup>41</sup>Ca (28), and <sup>129</sup>I (29). NH and MgH form negative ions so the limitation in rejection of <sup>14</sup>N and <sup>26</sup>Al is determined by the number and quality of MS stages prior to stripping.

At MeV energies, particle identification detectors that have become standard tools in nuclear physics can be used for isobar discrimination. These detectors rely on the fact that the rate of energy loss in matter (dE/dx) depends on the atomic number (Z) of the projectile (30). At 80 MeV the isobar pair <sup>36</sup>S-<sup>36</sup>Cl can be separated with a ratio of better than 10<sup>6</sup>:1 (10). For higher Z, more energy is needed (31).

For light elements where the unwanted isobar has a higher Z than the radioisotope, the thickness of an absorber can be adjusted to stop the isobar while allowing the radioisotope to pass into a detector. This has the advantage that counting rates in the detector remain low, but it works well for <sup>10</sup>Be-<sup>10</sup>B separation only (32).

**Table 2.** Four types of analysis stages used in mass spectrometers. Any two of these elements placed in series will select particles that have unique mass-to-charge and energy-to-charge ratios (12).

Type of analysis	Quantity selected		
Magnetic field	Mass × energy/charge <sup>2</sup>		
Electrostatic (ES) field	Energy/charge		
Velocity selector*	Mass/energy		
Cyclotron resonance	Mass/charge		

\*A velocity selector, also called a Wien or  $E \times B$  filter, consists of perpendicular magnetic and ES fields that provide zero net force on particles of the selected velocity. Time-of-flight detectors also select ions based on their mass-to-energy ratio.

The gas-filled magnet (33) is a powerful isobar separation instrument developed only recently for AMS (34). An ion passing through a gas changes its charge frequently by electron capture and loss. If this charge changing occurs frequently enough in a magnetic field region, the trajectory is determined by the average charge state of the ion, which depends on Z. The scatter in ion trajectories, which limits the ability to separate two isobars, is determined at low pressure by the statistics of the charge-changing processes and at higher pressures by small-angle scattering and energy-loss straggling. The magnet can be followed by a normal *dE/dx* detector with the added advantage that the unwanted ions, which now arrive at a separate location on the focal plane, can be blocked before entering. Further development of the gas-filled magnet should make possible AMS of <sup>36</sup>Cl at lower energies and should aid in AMS detection of other isotopes such as <sup>53</sup>Mn, <sup>79</sup>Se, <sup>92</sup>Nb, and <sup>126</sup>Sn.

Several other isobar separation techniques are possible. Stripping all electrons from an ion at very high energies allows complete separation from an isobar of lower Z (35, 36). Laser-induced resonance ionization spectrometry (37) is isobar specific and could be used in combination with accelerators. Surface ionization ion sources (38) are also isobar specific, but recent tests (39) have shown that this property may not be helpful for <sup>36</sup>S-<sup>36</sup>Cl separation, where it had great promise.



**Fig. 1.** Schematic diagram showing apparatus used for AMS. Not all of the elements shown are used at each installation: in particular the injector electrostatic (ES) analyzer (46), the velocity selector (32), and the gas-filled magnet (34) are not used at most laboratories. The positive-ion ES analyzer is sometimes located immediately after the accelerator. An offset Faraday cup is sometimes used after the injector magnet to monitor the source output while the radioisotope is being counted (32). At several installations rapid mass selection in the injector is accomplished by varying the potential of an insulated magnet vacuum chamber (45, 53). Offset cups are often utilized after the first positive-ion magnetic analyzer so that the field in that magnet does not have to be cycled.

### The Instrument

Figure 1 shows the various components of a typical AMS system. Tandem electrostatic accelerators are generally believed to be the best accelerators for measuring the isotopes listed in Table 1, although there have been some important results with cyclotrons (9, 40) and linear accelerators (34, 36, 41), and a small cyclotron is being developed for AMS (42).

The ion source. A cesium sputter ion source (Fig. 2) is used for most AMS work. This is essentially a secondary ion mass spectrometry (SIMS) instrument that has been refined to produce high currents of negative ions. Generally, solid samples are used; gas samples can give intense beams (43) but the problem of contamination from the previous sample ("memory") is difficult to overcome. For radioisotope studies, sample sizes are 1 to 10 mg of processed material and beam currents of 1 to 10  $\mu$ A are typical, depending on the element (see Table 1). Some sort of multiple sample changing system is used at most AMS installations. The construction of reliable, efficient ion sources is still something of an art; continuous developmental effort is under way at several laboratories (39, 43– 45).

The injector. Mass analysis of the negative-ion beam with a resolution sufficient to separate isotopes of heavy elements is needed prior to acceleration. An ES analyzer is used at the University of Toronto to sharpen the energy distribution of ions produced from cesium sputter ion sources (46). A pre-acceleration of the negative ion beam to 100 to 400 keV is used with the larger tandem accelerators to ensure that the injected ion beam is focused at the central terminal where the stripper canal is located.

The tandem accelerator. The name "tandem" refers to a dual acceleration design. The negative ions are accelerated to the terminal of the accelerator, which is held at a constant positive voltage (Table 1) typically in the range 2 to 10 MV. The electron stripper at the terminal removes several electrons as described above; positive ions are then accelerated a second time to ground potential. The Tandetron (47) operates reliably below 3 MV using a solid-state power supply, and tandem Van de Graaff accelerators (48) use a rotating belt or chain to charge the terminal up to 25 MV in some models. Tandem accelerators have the following characteristics: (i)

the ion source and detector are located at ground potential; (ii) they do not require pulsed beams; (iii) the electron-stripping step needed to eliminate molecules is an integral step in the operation of the accelerator; and (iv) transmission through the accelerator and subsequent analyzers can be made independent of small changes in the terminal voltage.

Positive-ion analysis. Analyzers positioned after the accelerator remove scattered particles accepted by the injector analyzer, molecular fragments, and unwanted charge states. Magnetic analyzers alone are not sufficient: an ES analyzer or velocity selector is necessary to remove particles that have different mass but would otherwise have the correct mass-energy product to pass through the magnetic analyzers (see Table 2). Furthermore, laboratories that have only one magnetic analyzer after the accelerator have troublesome interference from scattering and charge-changing and so two magnets are often used. The University of Pennsylvania (32) and several other laboratories use a velocity selector instead of an ES analyzer; although poorer in resolution, it has the advantages of variable resolution and the possibility of installation within existing beam lines since the beam is not deflected. We should point out that, for AMS, the resolution of an analyzer is less important than the intensity of the baseline signal between mass peaks. Another consideration is the bending power of the magnets: large radius of curvature (132 cm) magnets are used at full field (13.7 kG) for <sup>129</sup>I detection at Rochester (29).

Detection system. The time-of-flight detector serves as an additional positive-ion mass analysis stage (Table 2). It is most useful for the heaviest (slowest) ions such as <sup>129</sup>I (29) and consists of two timepickoff detectors with time resolutions of a few hundred picoseconds (49). The gas-filled magnet, discussed above, has been used for AMS only at Argonne (34). The dE/dx and total energy measurements are made with either gas-ionization detectors (Fig. 3) (50) or silicon surface-barrier detectors or a combination of these (32).

*Measurements.* Isotope ratios are obtained by alternately selecting each stable isotope and measuring its beam current in a removable Faraday cup and then by measuring the radioisotope counting rate in the detector (see caption to Table 1). Standards (samples with a known isotope ratio) are run periodically for normalization, and blanks (samples containing no detectable radioisotope) are used to



Fig. 2. Cesium sputter negative ion sources used for AMS. (A) Here the primary  $Cs^+$  beam is produced in a gun external to the extraction region and focused onto the sample from the front. Seven samples can be mounted in the holder and positioned with an X-Y manipulator. (B) In a high intensity version (97), a more intense  $Cs^+$  beam is formed directly in front of the sample on a cylindrical ionizer and focused onto the solid sample. The negative ions are extracted through the center of the ionizer.



**Fig. 3.** The gas ionization detector used at Rochester. Ions enter through a thin Mylar window into a region filled with isobutane gas at sufficient pressure to stop the ions under the fourth anode plate (about 50 torr for 80-MeV chlorine). Ionization electrons move from the cathode box (-500 V) up through the Frisch grid (-300 V) and are collected by the split anode plates (0 V) providing four  $\Delta E$  signals. This produces a profile for the energy loss that is distinctly different for each element. The ionization chamber, enclosed by the total energy.

measure background. Ratios are corrected for time-varying linear mass fractionation when more than one stable isotope is measured and for nonlinear fractionation, which arises from the stripping process and from stray magnetic fields in the accelerator, by comparison to the standard (51).

*Performance.* Table 1 gives the performance achieved to date in U.S. laboratories. The minimum sample size and minimum isotope ratio needed for 10% precision are listed. Samples of this size take less than 1 hour to measure but would take years (if background were low enough) with the decay-counting method. Table 1 emphasizes factors that determine sensitivity, not those for precision and capacity for measuring large numbers of samples. For samples with higher activity and longer run times it is possible to obtain better than 1% precision, and capacities can range as high as 20 to 30 samples per day. A report of the performance of the 20 operating AMS laboratories is beyond the scope of this review; however, it is safe to say that AMS facilities in Canada and Europe exceed those in the United States in both precision and capacity.

## Limitations of AMS

Sensitivity is a poorly defined term that reflects overall efficiency and background limitations. We will cover these effects separately and quantitatively.

*Efficiency.* Overall efficiency is the ratio of the number of (radioactive) atoms actually detected to the number of (radioactive) atoms in the sample. This ratio is equal to a product of (i) the fraction of sample used, (ii) the efficiency for producing negative ions, (iii) the yield in the particular charge state selected, and (iv) the efficiency for transmission through the accelerator and subsequent analyzers. The variation in ion source efficiency among the different radioisotopes is due primarily to the differences in electron affinity. For some samples, such as <sup>26</sup>Al in terrestrial materials and <sup>36</sup>Cl in seawater, there is plenty of sample material available but the concentrations are low and the real limitation is the length of time one is willing to run the sample. In this case, efficiency of the ion source is less important than the magnitude of the beam current.

The overall efficiency of AMS (Table 1) is actually lower than that possible from low-energy MS and some other techniques such as resonance-ionization spectrometry and neutron activation analysis. AMS excels in the suppression of isotopic backgrounds, thus allowing measurement of very small isotope ratios. The sample utilization and accelerator efficiency could each be improved somewhat, and, with new ion sources, improvements can be expected for the efficiency of producing negative ions. An ion source being developed for <sup>14</sup>C, which uses CO<sub>2</sub> gas, has an efficiency of 7% (43).

*Background.* Events that cannot be distinguished from the radioisotope being measured we collectively call background. Background can arise from (i) contamination from the environment before or during sample preparation, (ii) cross contamination and memory from other samples in the ion source, (iii) the residual gas and apertures in the ion source, and (iv) unresolved isotopes and isobars in the detector. The background isotope ratios listed in



Table 1 result from various combinations of these. For the multiparameter detectors used with AMS there is no background from cosmic-ray events. Cross contamination varies between  $10^{-3}$  and  $10^{-5}$ , depending on the geometry of the ion source. It is possible with adequate mass analysis to avoid background from neighboring isotopes. Isobaric interference can contribute to the background for <sup>10</sup>Be and <sup>36</sup>Cl, but only when the chemical preparation has not adequately removed the isobars <sup>10</sup>B and <sup>36</sup>S. Backgrounds for <sup>14</sup>C are not well understood; although there is no interference from <sup>12</sup>C and <sup>13</sup>C, <sup>14</sup>C is relatively abundant in the atmosphere and contamination is difficult to avoid. Once gas-filled magnets are used for <sup>10</sup>Be and <sup>36</sup>Cl detection, contamination and overall efficiency (that is, the need for sufficient statistics) will be the only limitation of AMS for these five radioisotopes. For trace element measurements ultrahigh vacuum ion sources are needed if ratios below the  $10^{-9}$  level are to be measured (52).

Precision. Factors that affect precision are counting statistics, isotope fractionation effects, frequency of cycling, and effectiveness of the blank and standard corrections. Radioisotope measurements of natural samples are usually limited in precision to about 3 to 10% by counting statistics, with the exception of <sup>14</sup>C samples younger than about 20,000 years. For <sup>14</sup>C it has been possible to obtain precision better than 1% with AMS by increasing the cycling frequency to about 10 Hz (45, 53) (see Fig. 1). The accuracy of fractionation corrections noted above depends on having a standard with identical chemical and physical characteristics and measuring it in the same way as the unknown sample (46, 51). The precision for measuring isotope ratios with conventional MS can be considerably better than with AMS because (i) there are usually many orders of magnitude more ions so statistical fluctuations are much less significant and (ii) the ion optics are much simpler so that fractionation corrections depend less critically on the focusing conditions.

### Applications

Cosmogenic nuclides are produced by the interaction of cosmic rays and their secondaries with terrestrial and extraterrestrial matter; since 1945 there have been anthropogenic (man-made) contributions from weapons testing and nuclear power plant releases. The five radioisotopes considered here (Table 1) are produced mainly in the atmosphere, enter the geologic cycle usually through the hydrosphere, and become incorporated in organic materials, ground water, and sediments. Carbon-14 is tied up in atmospheric CO<sub>2</sub> for several years after production, which allows it to become uniformly mixed with <sup>12</sup>C and distributed over the surface of the earth; this makes <sup>14</sup>C ideally suited for dating organic materials up to about 75,000 years old. The other radioisotopes are removed from the atmosphere by precipitation on a time scale of the order of 1 year, and thus dating is more subject to climate and production fluctuations. Dating methods are nevertheless being developed for these isotopes since their longer half-lives extend to a time interval not well covered by other methods. Geochemical tracer applications are important for all of these long-lived isotopes, and studies are under

way of in situ production on and below the earth's surface.

The following sections outline some of the AMS contributions to several current research areas. Additional information can be found in reviews by Brown (21), Phillips and Elmore (22), and in conference proceedings (15, 20, 54, 55).

Radiocarbon studies. Work in the past 30 years with the decay counting technique has established radiocarbon dating as an essential tool for dating and tracing organic materials in archeology and geochemistry. AMS has now reduced the required sample size by nearly a factor of 1000 from grams to milligrams (it has not yet significantly improved precision or extended the time range), which opens up a new realm of opportunity. AMS can now be used to date small or precious objects such as works of art, individual seeds, amino acids specific to bone, water, ice-core samples, and individual tree rings (20). AMS has tremendously increased the applicability of measuring <sup>14</sup>C as a tracer for carbon particulates in the atmosphere; these particulates may have profound effects on health and climate, and <sup>14</sup>C measurements can be used to trace their origin (56). Carbon-14 measurements of dissolved and particulate organic materials transported by rivers have been an aid in understanding the dynamics of organic materials within terrestrial ecosystems and their



**Fig. 4.** (**A**) The <sup>10</sup>Be concentration measured for the Milcent ice core (central Greenland) using AMS (73). The age of the ice was determined from annual variations of <sup>18</sup>O. (**B**) Comparison of  $\Delta^{14}$ C measured by decay counting in tree rings (71) with calculated  $\Delta^{14}$ C (dotted line) based on the measured <sup>10</sup>Be concentrations. The  $\Delta^{14}$ C (dotted line) was calculated by means of a carbon cycle model on the assumption that the <sup>10</sup>Be concentration reflects the atmospheric production rate of both isotopes. The three maxima correspond to the so-called Sporer, Wolf, and Maunder minima. During these periods it is thought that the solar activity (number of sun spots) was low. It is well known (71) that the solar activity modulates the galactic cosmic rays with an inverse relationship. The discrepancy between the measured and the calculated <sup>14</sup>C curves is probably due to climatic effects.

effects on the composition of the corresponding marine reservoirs (57). In oceanography a potential exists for application of highprecision, small sample radiocarbon measurements. It should be possible to reconstruct past ocean ventilation rates by measuring age differences of foraminifera shells between surface dwelling and bottom dwelling species that coexist in sediment cores (58). Other applications include improving the chronology of marine sediments and studying ocean circulation history, rapid climatic events, geochemistry of organic matter, and deep-sea vent organisms.

Hydrology. Chlorine-36 is an ideal isotope for dating and tracing old ground water because of its suitable half-life  $(3 \times 10^5 \text{ years})$ , simple geochemistry, conservative behavior in ground water, and a general absence of subsurface sources at levels comparable to the atmospheric input. AMS is sensitive enough to measure levels reached after residence in the subsurface for a million or more years. The <sup>36</sup>Cl dating method has now been demonstrated in a large, wellunderstood aquifer, the Great Artesian Basin in Australia (59); the water ages given by the measured <sup>36</sup>Cl/Cl ratios agree well with independent hydrodynamic simulations. This demonstration has paved the way for applications of <sup>36</sup>Cl dating to problems of highlevel nuclear waste repository siting (60), geothermal systems (61), and origins of brines (62).

Similarly, measurements of pre–atomic bomb <sup>129</sup>I have shown variations in hydrologic systems that are a predicable function of age and lithology (63). The longer half-life  $(15.7 \times 10^6 \text{ years})$  of <sup>129</sup>I may lead to an important dating method for ground water (63), marine sediments (64), and petroleum (65). Because of the high mobility of iodine in its inorganic form, <sup>129</sup>I can probably be used for tracing the slow movement of seawater through sediments in response to hydrothermal convection in the underlying oceanic crust. Determination of flow rates in long-lived, low-temperature convection cells is essential both for understanding the elemental budgets of the oceans and for assessing potential sites for the seabed disposal of radioactive waste.

Anthropogenic <sup>36</sup>Cl has important applications as a ground water tracer. The pulse of <sup>36</sup>Cl released into the environment from nuclear weapons testing in the mid-1950s peaked at two to three orders of magnitude above cosmogenic levels and returned to near preatomic bomb levels in the mid-1960s (*66*). This pulse is now an excellent tracer for measuring net infiltration and solute dispersion in desert soils and for studying the movement of solutes from waste repositories in the unsaturated zone. Comparison between perturbed and unperturbed levels of <sup>36</sup>Cl will provide new insight into degradation of water quality and salinization of soil from dry-land farming, an important global issue.

Geology and geochemistry. Accelerator mass spectrometry has been used to study the formation of manganese nodules, marine and lacustrine sedimentation, soil evolution, soil erosion, and rock weathering (21). Beryllium-10 studies of soils and continental sediments have proved useful in determining the long-term erosion history of landscapes covered by clay-rich *p*H-neutral-to-basic soils (67) and in determining the time scale of elemental transport through acidic clay-poor soils (68). The depth profiles of <sup>10</sup>Be in ferromanganese nodules from the world's oceans have yielded growth rates of 1 to 4 mm per 10<sup>6</sup> years (69).

The magmas from island-arc volcanoes, which are associated with a lithospheric plate being subducted at a few centimeters per year, may be derived from sediments along the top of the down-going slab. Measurements of <sup>10</sup>Be in 122 samples from 65 volcanoes (70) indicate that different mechanisms for sediment incorporation may act in different arcs. Future work may show what characteristics affect the isotope's concentration: plate geometry, structure of the overlaying material, seismicity, or geochemistry.

Radioisotopes are measured in ice cores, marine sediments, lake

sediments, and continental saline sediments for dating and for measuring production-rate variations. Tree rings show <sup>14</sup>C production rate variations over the past 7000 years, which are attributed to changes in solar activity (71). Polar ice cores contain an undisturbed record of fallout from the earth's atmosphere over the past 100,000 years or more. Measurements of <sup>10</sup>Be (72, 73) and <sup>36</sup>Cl in ice show some correlation with <sup>14</sup>C and provide information on the sun and climate over longer time scales than is possible from tree rings (Fig. 4). Continental saline sediments can be dated with <sup>36</sup>Cl as shown for Searles Lake in California (74): correlation of cores from associated basins is expected to provide a new and detailed reconstruction of paleoclimatic fluctuations over the past million years. Studies of <sup>10</sup>Be in marine sediment cores can be used to determine sedimentation rates over a time range up to 10 million years (75) and, at the time of geomagnetic reversals, give information on the form and length of the geomagnetic intensity changes (76).

Although AMS is primarily useful for detection of long-lived radioisotopes, sensitive analysis of stable isotopes is also possible. The <sup>187</sup>Re-<sup>187</sup>Os system has been shown (77) to be an indicator of extraterrestrial material and a tracer for the origin of crustal material. The very low (<1 part per billion) concentrations of osmium in geological samples and the large spread of isotopic compositions make AMS well suited for measurement of osmium isotopes. AMS measurements of the <sup>186</sup>Os/<sup>187</sup>Os ratio have been made in samples having osmium concentrations as low as 0.01 ppb and having as little as 0.2 ng of osmium (3).

Measurable quantities of <sup>10</sup>Be, <sup>26</sup>Al (78), and <sup>36</sup>Cl (79) are produced in surface rocks by in situ interactions with cosmic rays. These isotopes build up predictably with time and can be used for dating the exposure time. For very long exposures, equilibrium concentrations provide information on erosion. There are widespread applications to Quaternary chronologic problems as diverse as determining the age of glacial moraines, the erosion of strath terraces, the date of ancient floods, the formation of intervals of soils, and rates of erosion and deposition.

Cosmochemistry. Studies of long-lived radioisotopes in meteorites can help us to understand the history of the cosmic-ray flux and the exposure history of the meteorites themselves (80). Meteorites contain the fossil record of the cosmic radiation, and these studies represent a rich possibility for studying variations in that record. Radioisotopes tell us about the exposure age of meteorites in space, the extent of ablation of material as they enter the earth's atmosphere, and the time since they fell (their terrestrial age). Irradiation records identified by measuring several radioisotopes help us to understand the origin of extraterrestrial material. For example, <sup>10</sup>Be and <sup>26</sup>Al measurements (81) in  $\approx$ 500 µm in diameter "cosmic spherules" recovered in marine sediments confirm an extraterrestrial origin and indicate that they may represent cometary debris.

More than half of the meteorites recovered to date have come from ice fields in the Antarctic. Terrestrial ages have been measured with <sup>36</sup>Cl in many of these and range up to  $2.7 \times 10^6$  years (82). These measurements can provide sensitive tests of models for the flow and emergence of ice in blue ice regions and the resulting concentration of meteorites.

*Mineral exploration*. Accelerator mass spectrometry has been used for measuring trace levels of platinum group elements (PGE) in rocks and minerals. Bulk concentrations of less than 1 ppb have been measured with AMS for platinum and iridium sample targets prepared by the NiS (fire assay) concentration method (83). Sensitivity for iridium by this method is comparable to that by neutron activation analysis, whereas for platinum it is several times better, and thus the method might be useful in the search for commercial platinum ore deposits. In addition, qualitative assays of these elements may be obtained on "raw" samples (crushed powders, cut surfaces) for identifying PGE distribution at the individual mineral grain level (2, 44). This technique may eventually permit detailed analysis of PGE mineralogical association and provide information necessary for developing mineral separation processes to optimize recoveries of precious metals.

*Materials sciences.* Metallic impurities in semiconductor materials limit the development of integrated circuit devices because of the unwanted formation of conductive paths and the creation of deep trapping sites. Concentrations in the range of  $10^{11}$  to  $10^{13}$  atoms per cubic centimeter ( $10^{-11}$  to  $10^{-9}$  g per gram) have observable effects, and currently available analytical techniques cannot detect most impurities at this level (*84*). AMS equipped with an appropriate (ultrahigh vacuum) ion source can probably meet the necessary analytical requirements (*52*).

*Physics.* Big Bang cosmology implies that many exotic elementary particles were present in large numbers during the earliest moments of creation. Although most of these have decayed into stable matter as we know it (neutrons, protons, and electrons), current theories (85) predict that there should exist today stable or nearly stable particles free in nature that could be identified as having either fractional ( $\pm 1/3$  or  $\pm 2/3$ ) charge or integral charge with anomalous mass. With its high sensitivity, AMS is a useful tool for searching for such particles (86). All-electrostatic analysis (Table 2) has been used to avoid dependence on the unknown mass (87).

Half-lives for short-lived radioisotopes can be measured by recording the decay rate as a function of time. This is not possible for lifetimes over about 100 years; here one can use the relation  $dN/dt = -\lambda N$ , which requires measurements of the number of atoms, N, and the activity, dN/dt. (The decay constant,  $\lambda$ , is related to the half-life by  $\lambda = \ln 2/t_{1/2}$ .) For some radioisotopes, it is difficult to produce enough activity to measure N with conventional mass spectrometry and AMS must be used. Half-lives have been measured for <sup>32</sup>Si (88, 89), <sup>44</sup>Ti (90), and <sup>60</sup>Fe (41).

*Biological sciences.* Radioisotopes are used extensively in studies of living systems by introducing labeled compounds as tracers. AMS can enhance detection of long-lived tracers by reducing the measurement time (giving a shorter turnaround), by reducing the dose (which decreases radiation damage and the cost), and by providing direct two-dimensional scans of sections (similar to autoradiography) (91). Elements that have no radioisotopes with half-lives between 1 hour and 1000 years are carbon, aluminum, and chlorine. Of these, <sup>14</sup>C is widely used (and has much to gain in sensitivity and convenience with AMS detection) but <sup>26</sup>Al has not yet been tried as a tracer. The biological effects of aluminum are not well understood (92), and <sup>26</sup>Al can already be detected with sufficient sensitivity with AMS (93).

#### **Concluding Remarks**

After nearly 10 years of development, AMS has been proven to have sensitive detection capability for five long-lived radioisotopes: <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>129</sup>I. With the tremendous reduction in sample size compared with conventional decay-counting methods, entirely new applications in the earth sciences have been added to older, and now revitalized ones. We anticipate that earth scientists will increasingly adopt AMS in studies requiring dating capability or natural tracers that cover time spans of hundreds to millions of years. We expect that the AMS technique will continue to expand beyond these five radioisotopes and that research will move into new areas including materials sciences and biological sciences.

AMS is an example of a new technique that was developed within one discipline (nuclear physics) but has primary applications in another (geosciences). It is reassuring that the funding system has allowed this. AMS would never have developed if earth scientists had no choice but to develop the technique themselves nor would it have developed if some nuclear physicists were not interested in archeology and the geosciences (94).

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