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Review

Progress in isotope analysis at ultra-trace level by AMS

Walter Kutschera*

Vienna Environmental Research Accelerator (VERA), Institut für Isotopenforschung und Kernphysik,
Universität Wien, Währingerstrasse 17, A-1090 Wien, Austria

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Abstract

This paper attempts to convey a flavor of the progress in Accelerator Mass Spectrometry (AMS) since its initiation in 1977. During this period, AMS evolved into the most powerful analytic technique to measure long-lived radioisotopes at natural isotopic abundances, which typically range from 10^{-12} to 10^{-16}. As such, it covers a section of isotope ratio measurements, which is hardly accessible by any other mass spectrometric means.

Keywords: Accelerator mass spectrometry; Long-lived radioisotope analysis

Contents

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* Tel.: +43 1 4277 51700; fax: +43 1 4277 9517.
E-mail address: walter.kutschera@univie.ac.at.

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1. Introduction

In order to speak about progress, we have to define a beginning. In the context of this review, it may be interesting to describe briefly the very beginning of the use of accelerators for mass spectrometry. The dawn of the field we now call Accelerator Mass Spectrometry (AMS) goes back to the early days of accelerators. In 1939, Alvarez and Cornog used the newly built 60-in. cyclotron in Berkeley to discover accidentally ^{3}He in nature [1]. The discovery of ^{3}He as a natural constituent of helium was surprising because at that time ^{3}H was expected to be the stable isobar of mass 3, and therefore ^{3}He was supposed to be radioactive [2]. Alvarez and Cornog went on to measure ^{3}He/^{4}He ratios in helium from gas wells (10^{-8}) and from atmospheric helium (10^{-7}), and they also found that ^{3}H produced in the irradiation of deuterium with deuterons was radioactive [3]. These experiments showed already the unique power of using accelerators as mass spectrometers: to measure extremely low-isotopic abundances, which are beyond the capability of low-energy mass spectrometers. Much later, Alvarez recalled these first AMS experiments in a reminiscent talk at the second AMS conference at Argonne National Lab in 1981 [4]. A write-up of this story can also be found in *Physics Today* [5].

After this first AMS experiment, almost 40 years passed without using accelerators as mass spectrometers. During this period, the main purpose of accelerators was to provide beams for experiments in nuclear and particle physics. While particle physics was aiming at beams with the highest possible energies of light particles (electrons and protons), nuclear structure studies were interested in isotopic beams of essentially all elements with precisely defined energies. The demands of nuclear physics thus nurtured the development of accelerator technology, which eventually would be become very useful for AMS. For example the Cs-beam sputter source, developed to produce negative-ion beams for tandem accelerators from a large variety of elements [6], turned out to become an indispensable part of AMS facilities.

In 1977, it was again Luis Alvarez who revived the use of a cyclotron as a very sensitive mass spectrometer to search for quarks with unit charge, i.e., essentially for anomalously heavy isotopes of hydrogen [7]. In the course of this experiment, Alverez’ collaborator Richard Muller quickly realised the power of AMS to measure rare radioisotopes, such as ^{3}H ($t_{1/2} = 12.3$ a), ^{10}Be (1.5×10^{6} y), ^{14}C (5730 a), and ^{26}Al (7.1×10^{5} a) [8]. As often in science, there were parallel efforts going on to use tandem accelerators for an AMS detection of ^{14}C. The first important step was to demonstrate that ^{14}N does not form stable negative ions [9]. It was then shown by two groups at the tandem accelerators of McMaster University in Hamilton [10] and of the University of Rochester [11] that ^{14}C can indeed be detected at the extremely low, natural isotope ratio of $^{14}\text{C}/^{12}\text{C} = 10^{-12}$. The non-existence of certain negative ions gave tandem accelerators a distinct advantage over accelerators operating with positive ions only. In addition, the stripping process in the tandem terminal efficiently breaks up interfering molecules (e.g., $^{12}\text{CH}_2^-$ and $^{13}\text{CH}^-$). Therefore, ^{14}C, the most important radioisotope for dating in archaeology, can be easily detected with tandem accelerators. Similarly, the detection of ^{26}Al and ^{129}I (1.7×10^{7} a) benefits from the instability of $^{26}\text{Mg}^-$ and $^{129}\text{Xe}^-$, respectively. Already in the first year of AMS, attempts to detect superheavy elements in nature with this method were undertaken at the tandem accelerator of Brookhaven National Laboratory [12]. Such experiments were driven by the ability of tuning an accelerator to uncharted territory and to identify and detect traces of extremely rare isotopic species.

The rapid development of AMS in the first couple of years [13] clearly demonstrated the advantage of tandem accelerators over cyclotrons. Nevertheless, an interesting project at the Shanghai Institute of Nuclear Studies eventually led to a demonstration of ^{14}C dating measurements with a mini-cyclotron, operating also with negative ions to suppress ^{14}N [14]. On the other hand, for AMS of noble gases positive-ion accelerators must be used, since noble gases do not form negative ions [15]. This leads to particularly severe background problems from stable-isobar interferences, e.g., of ^{81}Br for ^{81}Kr detection [16], and of ^{39}K for ^{39}Ar detection [17].

It was also evident early on, that small dedicated tandem facilities would be the most efficient machines for AMS, particularly for ^{14}C measurements. Pioneering efforts in this direction were pursued for many years by Purser [18,19]. The development of AMS in the past 25 years has been reviewed extensively [13,20–28]. It is recommended to inspect these reviews for an in-depth study of AMS. In addition, the proceedings of the tri-annual AMS conferences starting in 1978 provide ample evidence for this steadily growing field [29–37].

In the following, the basics of AMS will be briefly discussed in Section 2, followed by recent technical developments in the field in Section 3. Applications of AMS are so numerous that only a flavor of their prospects can be discussed in Section 4. The outlook in Section 5 will discuss some remote possibilities of AMS for the future, rather than trying to make ‘hard’ predictions.

2. Basics of AMS

AMS is a method to measure long-lived radioisotopes by counting atoms rather than decays. The advantage of the former can be most easily demonstrated by the numerical example given in Table 1. It can be seen that one gains a factor of about one million in detection sensitivity with AMS as compared to beta (β)-counting. To envision such a huge improvement in detection sensitivity, let us compare it with the gain in light collection efficiency looking at the stars with our bare eyes (aperture ∼5 mm) as compared to a very large telescope (aperture ∼5 m). Since the amount of light collected scales with the square of the aperture one gets a factor of $(5000/5)^2 = 1 \times 10^6$. This enormous gain revolutionized ^{14}C dating, since it is now possible to perform ^{14}C measurements with a thousand times smaller sample material (milligrams instead of grams) and a hundred times shorter counting times (0.5 h instead of several days). Although this gain in detection sensitivity is already very impressive, it increases further for longer half-lives and/or decays where the radiation is difficult to observe. In fact, there are radioisotopes, such as ^{41}Ca ($t_{1/2} = 1.04 \times 10^5$ a), which have never been detected at natural levels ($^{41}\mathrm{Ca}/^{40}\mathrm{Ca} = 10^{-14}$ to 10^{-15}) prior to AMS [38–40]. It is interesting to note that ^{41}Ca was proposed for dating long before there was hope to detect it in nature [41], and independently again shortly after AMS was invented [42].

Measuring isotope ratios down to a level of 10^{-15} and below requires a method capable of handling a very large dynamic range of isotope measurements. This comes naturally with the AMS technique, since the abundant stable isotopes (e.g., ^{12}C, ^{13}C) are measured as ion currents in suitable Faraday cups, whereas the rare isotope (e.g., ^{14}C) is measured by ion-counting in a solid state or gas detector (Table 1). In order to demonstrate the selectivity required to measure isotope ratios in the range from 10^{-12} to 10^{-15}, a student at the Vienna Environmental Research Accelerator (VERA) came up with an interesting comparison: if we take an average house with 100 m^2 floor space and a height of 10 m, we get a volume of 1000 m^3. Filling this house from bottom to roof with sugar of 1-mm^3 grain size, we can fill in exactly 10^{12} grains. A ^{14}C measurement of modern organic carbon with AMS is thus comparable of finding one sugar grain with different mass in the house filled with sugar. Furthermore, a ^{14}C measurement in a 57,300-year-old sample is equivalent of finding one sugar grain with different mass in 1000 houses filled from bottom to roof with sugar.

3. Technical developments in recent years

3.1. Accelerator developments

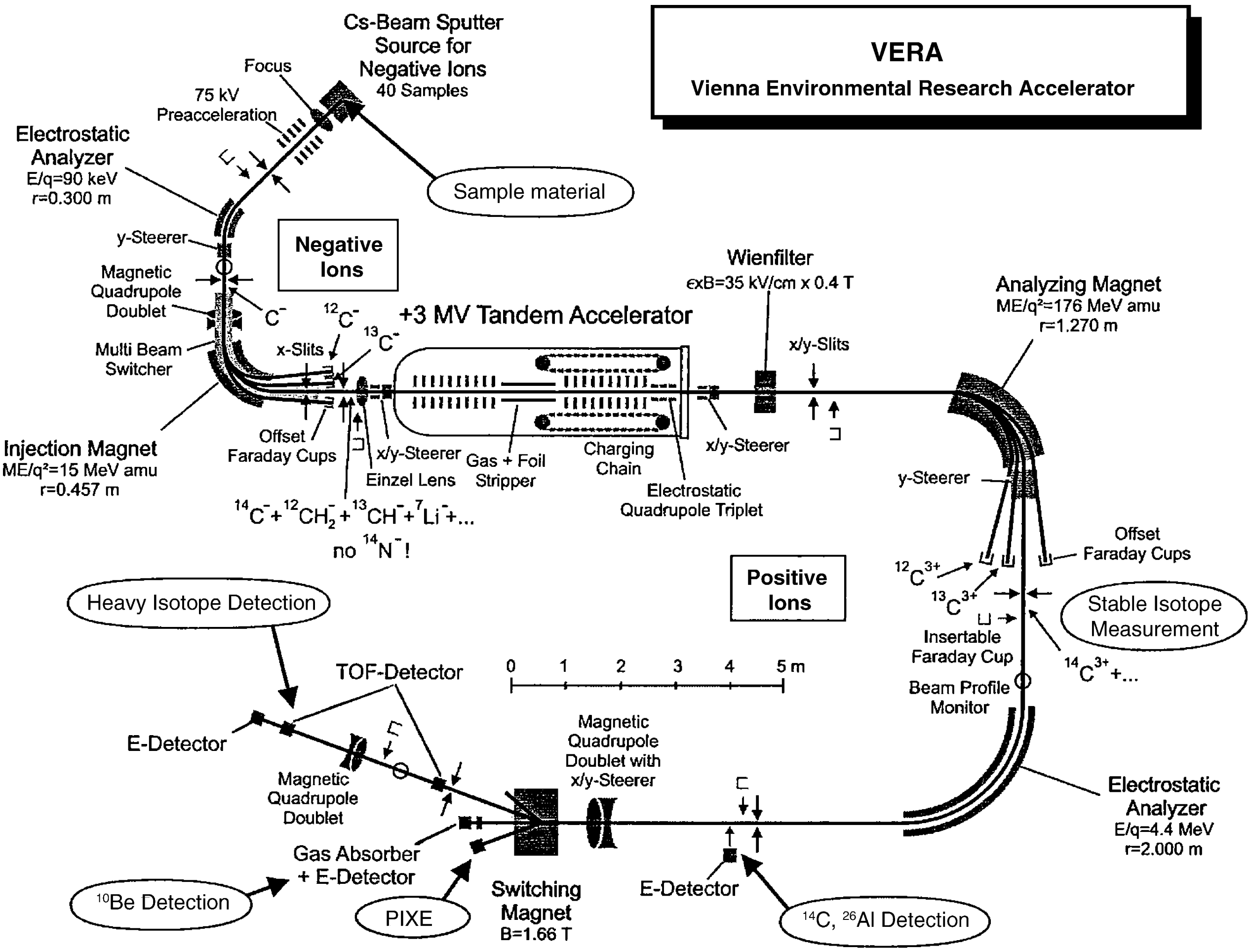
AMS started out by using a cyclotron [8] and tandem accelerators [9–11] available at nuclear physics laboratories. It

Table 1
Comparison of ^{14}C detection by beta-counting vs. atom-counting

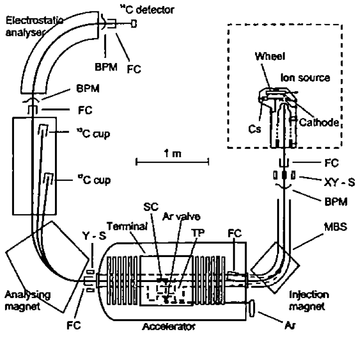
Characteristics of sample material	
Amount of material	1 mg organic carbon
Isotope ratios	$^{13}\mathrm{C}/^{12}\mathrm{C} = 1.1 \times 10^{-2}$ $^{14}\mathrm{C}/^{12}\mathrm{C} = 1.2 \times 10^{-12}$
Number of atoms	5.0×10^{19} ^{12}C atoms 5.5×10^{17} ^{13}C atoms 6.0×10^{7} ^{14}C atoms
Beta-counting with gas proportional detectors or liquid scintillation detectors	
Beta decay	$^{14}\mathrm{C} \rightarrow {}^{14}\mathrm{N} + e^- + \bar{\nu}_e$, E_β (max) = 158 keV
Half-life	$t_{1/2} = 5730 \pm 40$ a
Decay constant	$\lambda = \ln 2/(t_{1/2}) = 1.210 \times 10^{-4}$ a^{-1}
Decay rate	$\mathrm{d}(^{14}\mathrm{C})/\mathrm{d}t = -\lambda^{14}\mathrm{C} = 7260$ decays/a = 0.828 decays/h
β-counting rate	0.50 counts/h (∼60% detection efficiency)[a]
Ion-counting at the VERA 3-MV tandem AMS facility	
^{12}C ion currents	$^{12}\mathrm{C}^- = 50\ \mu\mathrm{A} = 3.1 \times 10^{14}$ $^{12}\mathrm{C}^-$ ions/s $^{12}\mathrm{C}^{3+} = 75\ \mu\mathrm{A} = 1.6 \times 10^{14}$ $^{12}\mathrm{C}^{3+}$ ions/s (50% 3+ stripping efficiency)
^{14}C counting rate	190$^{14}\mathrm{C}^{3+}$ ions/s = 684,000$^{14}\mathrm{C}^{3+}$ ions/h 0.190$^{14}\mathrm{C}^{3+}$ ions/s = 684$^{14}\mathrm{C}^{3+}$ ions/h for 57,300-year old carbon
Detection efficiency	(Ions counted)/(atoms in the sample) = 2%[b]
Comparison of ^{14}C detection sensitivity	
Ion-counting/beta-counting = 684,000/0.50 = 1.4×10^6	

[a] Beta-counting actually requires about 1 g of carbon (500 counts/h), otherwise the background counting rate (mainly from cosmic-rays) in the detector far exceeds the ^{14}C decay signal. Still, this counting rate is a factor of 1000 smaller than the ^{14}C ion-counting rate measured with AMS from 1 mg of carbon.

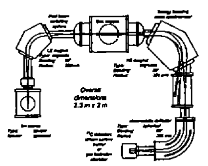
[b] This assumes that 1 mg of carbon is used up (sputtered away) in 1 h, which is a typical consumption rate in the Cs-beam sputter source, but may vary somewhat.



Standard AMS, TV = 3 MV
16 mx 12 m = 192 m^2
VIENNA



Compact AMS, TV = 0.5 MV
6 m x 5 m = 30 m^2
POZNAN



Mini AMS, TV = 0.2 MV
3.0 m x 2.3 m = 6.9 m^2
ZURICH

was quickly realised, however, that ^{14}C dating needed high precision for isotope ratio measurements in order to compete with existing beta-counting facilities – never mind the enormous gain in sample reduction for AMS (Table 1).

Tandem accelerators at nuclear physics facilities were not built to be used as mass spectrometers for the required precision (5‰) of $^{14}C/^{12}C$ ratios measurements, and were unnecessarily large (terminal voltages of 8–10 MV). Therefore, small dedicated tandem AMS facilities (2–3 MV) were proposed early on [18], with first results reported at the AMS-3 conference in 1984 [31]. Instead of using belts (and later Pelletron chains) to generate the high voltage of Van-de-Graaff type accelerators, the charging system of the small machines was based on a Cockroft–Walton type power supply (Tandetron).

In 1990, Purser presented a precision ^{14}C accelerator mass spectrometer [19] based on this principle. In recent years, two companies offer complete AMS facilities: High Voltage engineering Europe (HVEE) in Amersfoort (Netherlands) manufactures Tandetron-type AMS facilities, and National Electrostatic Corporation (NEC) in Middleton, WI (USA) manufactures Pelletron-type AMS facilities. Both machines have their pros and cons, but both have reached a high level of sophistication allowing ultra-trace isotope analysis with high precision and accuracy.

With the rapid increase in computing power, automisation became common ground for the second and third generation of dedicated AMS facilities. When a higher energy is needed to solve the separation of stable-isobar background from the radioisotope of interest, computerisation also helps to run larger and more complex accelerators in a reasonably quantitative way for AMS experiments. Depending on the scientific question, isotope ratio measurements with modest precision (5–15%), achievable at almost any large accelerator, can lead to relevant answers. For example, $^{81}Kr/^{80}Kr$ ratios in the range of 10^{-13} were measured at the superconducting cyclotron of Michigan State University for dating very old groundwater for the first time with ^{81}Kr ($t_{1/2} = 2.3 \times 10^5$ a) [43]. At the ATLAS linear accelerator at Argonne National Laboratory, $^{39}Ar/^{40}Ar$ ratios down to the extremely low range of 10^{-16} were measured to develop a technique for studying oceanic circulation characteristics [17]. In both cases, it was necessary to use positive ions from the ion source, since noble gases do not form stable negative ions. Another example of high-energy requirement was the measurement of $^{60}Fe/^{56}Fe$ ratios in the range of 10^{-15} in deep-sea manganese crusts at the 14-MV tandem accelerator at Munich, resulting in the first indication for a supernova explosion 'close' to Earth some 3 million years ago [44,45].

3.2. The standard AMS machines

Notwithstanding the high-energy applications mentioned above, the 3-MV tandem accelerator turned out to become the 'standard' facility for AMS. Approximately half of all AMS facilities worldwide (i.e., about 30 out of 60) fall into this category. From those, most of them concentrate on ^{14}C measurements only. However, continuous improvement on peripheral instrumentation led to facilities of high versatility, capable of covering a large range of isotopes. For example, at the Vienna Environmental Research Accelerator (VERA) (Fig. 1) the radionuclides ^{10}Be, ^{14}C, ^{26}Al, ^{129}I, ^{182}Hf, ^{210}Pb, ^{236}U, and ^{244}Pu have been measured [46,47]. It is also important to note that the universality of VERA does not compromise on the precision of ^{14}C measurements [48], reaching essentially the same performance level as the dedicated ^{14}C machines [49]. In fact, a recent work at VERA on ^{14}C calibration of a high-altitude stone-pine dendrochronology required a precision of $^{14}C/^{12}C$ ratio measurements at the 2‰ level [50]. One has thus reached the precision of beta-counting facilities, which from the beginning was a major goal for AMS.

Even though the standard-size machine of 3 MV terminal voltage turned out to be a highly versatile tool for most AMS needs, 5-MV machines have a certain advantages if ^{36}Cl measurements are of interest [51,52]. This is due to the difficulty of separating the stable isobar interference of ^{36}S at lower energies. As mentioned above, even higher energies and other accelerator types are sometimes required to solve the stable-isobar interference problem in particularly difficult cases, and to perform AMS experiments for noble gas radionuclides, respectively.

3.3. The 'small-is-beautiful' development

The most significant technical development in recent years is displayed in Fig. 1, which shows the trend towards smaller AMS facilities. The greatly reduced floor space for these new facilities and the reduction of the number of elements to be controlled are the most attractive features – not to speak about the reduction in overall cost. The significantly lower energies, however, introduce a variety of problems (stripper thickness, molecular dissociation, scattering, ion-optical beam quality, vacuum requirements, etc.), which had to be thoroughly investigated, and are summarized in a recent paper by Suter [28]. After some early, exploratory work at Toronto [53], and first ideas about small AMS systems for biochemical and environmental applications [54], the essential break-through came at the PSI/ETH AMS facility in

Fig. 1. Comparison of the size of modern AMS facilities. On top of the figure, the Vienna Environmental Research Accelerator, VERA, is shown in its present form after an upgrade for heavy-isotope AMS [46]. The locations for the measurement of the three carbon isotopes for a ^{14}C AMS measurement is indicated. Other light radioisotopes (^{10}Be, ^{26}Al) are measured at the respective detector positions. Heavy radioisotopes (^{129}I, ^{182}Hf, ^{210}Pb, ^{236}U, and ^{244}Pu) are measured at the beamline including a TOF (time-of-flight) set-up [47]. A future position for proton-induced X-ray emission analysis (PIXE) is also indicated. In the lower part of the figure, increasingly smaller AMS facility are shown on the same scale. The compact facility at Poznan [62] has already a substantially reduced floor space, whereas the Mini AMS facility at Zürich [68] has truly reached table-top dimensions.

Zürich [55]. There it was shown unequivocally, that the abundant $^{12}CH_2{}^-$ and $^{13}CH^-$ molecules can be dissociated in the 1+ charge state, provided that the gas-stripper thickness is sufficiently large. This then allowed to lower the terminal voltage from 3 to 0.5 MV, and even lower. Previously, it was "gospel" that 3+ charge states had to be used to dissociate the molecules. Whereas, the 3+ charge state guarantees a Coulomb dissociation, the 1+ charge state requires a collisional dissociation, a process well-known in low-energy mass spectrometry.

A collaboration of the Zürich lab with National Electrostatic Corporation in Wisconsin led to a 'compact' prototype AMS facility with only 0.5 MV terminal voltage [56]. Subsequently, the group performed many detailed studies to improve the system [57–60]. Besides Zürich, compact ^{14}C dating facilities delivered by NEC are now operational at the University of Georgia in Athens [61], at the A. Mickiewicz University in Poznan [62], and at the University of California in Irvine [63]. For a less stringent detection of ^{14}C and ^{3}H in labeled compounds for biomedical investigations, compact facilities at MIT in Boston [64,65], and at Lawrence Livermore National Lab [66] have been constructed with terminal voltages at 1 MV. Meanwhile the upgraded compact facility at Zürich ventured successfully into other radionuclides besides ^{14}C, such as ^{10}Be, ^{26}Al, ^{129}I, and ^{244}Pu [67].

An interesting development towards even smaller AMS facilities is now going on, with the goal to reach a true tabletop facility (Fig. 1). Again, the Zürich group is leading these efforts, and now pushes the terminal voltage of their Mini AMS facility down to 0.2 MV [68]. In addition, NEC has recently constructed a single-stage low-energy facility to be installed at the University of Lund running at 0.25 MV accelerating voltage [69]. Although starting with negative ions like the tandem facilities, there will be no acceleration after the stripping process. The physical size of this latter machine is comparable to the compact machines described above, because it needs a high-voltage platform with proper protection. Both machines avoid the use of SF_6 as insolating gas, simplifying matters further.

3.4. Sample preparation

It is well-known that sample preparation is an integral part of mass spectrometric measurements. No matter how precise the mass spectrometer on hand, without a careful sample preparation, the results may be highly precise but inaccurate. There seems to be no better advice for this problem as the one giving by Willard Libby – the inventor of ^{14}C dating – in his Noble Lecture of 1960: "Radiocarbon dating is something like the discipline of surgery – cleanliness, care, seriousness, and practice." The ability of AMS to make measurements on small samples suggests to push this parameter to the lowest amount feasible. Whereas 1 mg of solid carbon is the standard amount used for a ^{14}C AMS measurement, sample sizes down to 10 μg have also been used occasionally (e.g., [70]). However, due to background contributions the uncertainties of $^{14}C/^{12}C$ measurements increase considerably. If little original sample material is available, diluting it with 'dead' carbon is sometimes the better option in order to get enough carbon (100–200 μg) for a reliable AMS measurement [71].

On the other hand, carrier-free $^{10}Be/^{9}Be$ ratio measurements have been performed recently at the Zürich AMS facility, using a focused Cs-beam sputter source and only 0.1 μg of beryllium extracted from deep-sea ferromanganese crusts [72]. Approximately 10% precision can be reached in such measurements. Another carrier-free approach has been developed at the Gifsur-Yvette AMS facility to measure $^{129}I/^{127}I$ ratios in seawater [73]. Samples of 0.1 l of seawater were used containing approximately 6 μg of iodine, which is converted into AgI for use in the sputter source.

Most AMS facilities are equipped with cesium-beam sputter sources (e.g., [74]), which require solid sample preparation. An important step for solid carbon preparation was the work of Vogel et al. [75] using a catalytic reduction of CO_2 to elemental carbon. The 'Vogel-method' is now used at almost all AMS laboratories for ^{14}C sample preparation. The interest in biochemical investigations with labeled compounds, and also various environmental applications, require a compound-specific analysis of the original sample material. It would thus be advantageous to combine biochemical separation techniques, such as Gas Chromatography (GC) and High Performance Liquid Chromatography (HPLC) with AMS. Several AMS laboratories are working on such schemes, e.g., at MIT [65], Oxford [76], and Woods Hole Oceanographic Institution [77]. In this case, one uses special gas ion sources, capable to accept material in the typical time frame given by the GC and HPLC output, and convert those pulses of compounds into a negative-ion beam. It seems that substantial technical developments are still lying ahead, but the on-line coupling of such a sophisticated front end to an AMS facility is likely to open up a new dimension of analytic capability.

4. Applications

The main field of AMS is the measurement of long-lived radioisotopes at natural levels. Since radioisotopes are present in virtually every terrestrial and extraterrestrial material, and AMS made it possible to trace these radioisotopes at the faintest level, an enormous breadth of applications evolved. An overview of the areas of applications is given in Table 2, where our physical world is divided into seven large domains. It can easily be seen that one can cover almost the entire world by measuring long-lived trace isotopes. It is also apparent that one cannot describe the many applications in any detail in this review. Thus, a random walk through the table will be performed, picking out a few raisins from the cake.

Table 2
Overview of AMS applications in the seven large domains of the environment

Domain	Area of application[a]
Atmosphere	Cosmogenic and anthropogenic radionuclides in the atmosphere (^{3}H $t_{1/2} = 12.3$ a), $^{3}\underline{\mathrm{H}}$, ^{7}Be (53 d), ^{10}Be (1.5 × 10^{6} a),^{14}C (5730 a), $^{14}\underline{\mathrm{C}}$, ^{26}Al (7.1 × 10^{5} a), ^{32}Si (140 a), ^{36}Cl (3.01 × 10^{5} a), $^{36}\underline{\mathrm{Cl}}$, ^{39}Ar (269 a), ^{81}Kr (2.3 × 10^{5} a), $^{85}\underline{\mathrm{Kr}}$ (10.8 a), ^{129}I (1.7 × 10^{7} a), $^{129}\underline{\mathrm{I}}$) Study of trace gases: CO_2, CO, OH, O_3, CH_4 (^{14}C) Transport and origin of carbonacous aerosols (^{14}C, $^{14}\underline{\mathrm{C}}$) and loess (^{10}Be) Exchange of stratospheric and tropospheric air (^{7}Be, ^{10}Be)
Biosphere	Dating in archaeology and other fields (^{14}C, ^{41}Ca (1.04 × 10^{5} a)) Calibration with tree rings, corals, lake and ocean sediments, spaleothems (^{14}C) Studies in forensic medicine through bomb-peak dating ($^{14}\underline{\mathrm{C}}$) In vivo tracer studies in plants, animals, and humans ($^{14}\underline{\mathrm{C}}$, $^{26}\underline{\mathrm{Al}}$, $^{41}\underline{\mathrm{Ca}}$, $^{79}\underline{\mathrm{Se}}$ (3.0 × 10^{5} a), $^{99}\underline{\mathrm{Tc}}$ (2.11 × 10^{5} a), $^{129}\underline{\mathrm{I}}$)
Hydrosphere	Dating of groundwater (^{14}C, ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{129}I) Global ocean circulation pattern (^{14}C, $^{14}\underline{\mathrm{C}}$ ^{39}Ar, ^{99}Tc, $^{129}\underline{\mathrm{I}}$) Paleoclimatic studies in lake and ocean sediments (^{14}C)
Cryosphere	Paleoclimatic studies in ice cores from glaciers and polar ice sheets (^{10}Be, ^{14}C, ^{26}Al, ^{32}Si, ^{36}Cl, ^{39}Ar, ^{81}Kr) Variation of cosmic-ray intensity with time (^{10}Be, ^{14}C, ^{36}Cl) Bomb-peak identification ($^{36}\underline{\mathrm{Cl}}$, $^{41}\underline{\mathrm{Ca}}$, $^{129}\underline{\mathrm{I}}$)
Lithosphere	Exposure dating and erosion studies of surface rocks (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl) Neutron flux monitor in uranium minerals (^{236}U (2.34 × 10^{7} a)) Paleoclimatic studies in loess (^{10}Be) Tectonic plate subduction studies through volcanic rock measurements (^{10}Be)
Cosmosphere	Cosmogenic radionuclides in meteorites and lunar material (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{44}Ti (60 a), ^{59}Ni (7.5 × 10^{4} a), ^{60}Fe (1.5 × 10^{6} a), ^{129}I). Possible live supernovae remnants on Earth (^{60}Fe, ^{146}Sm (1.08 × 10^{8} a), ^{182}Hf (8.9 × 10^{6} a), ^{244}Pu (8.1 × 10^{7} a), ^{247}Cm (1.56 × 10^{7}) Stable trace isotope ratios in presolar grains (Pt, Os) Geochemical solar neutrino detection (^{99}Tc, ^{205}Pb 1.5 × 10^{7} a)) Search for exotic particles (superheavy elements, fractionally charged particles, anomalously heavy isotopes, strange matter, dark matter)
Technosphere	Releases from nuclear fuel reprocessing ($^{85}\underline{\mathrm{Kr}}$, $^{99}\underline{\mathrm{Tc}}$, $^{129}\underline{\mathrm{I}}$) Half-life measurements ($^{32}\underline{\mathrm{Si}}$, $^{41}\underline{\mathrm{Ca}}$, $^{44}\underline{\mathrm{Ti}}$, $^{60}\underline{\mathrm{Fe}}$, $^{79}\underline{\mathrm{Se}}$, $^{126}\underline{\mathrm{Sn}}$) Temperature measurement of fusion plasma ($^{26}\underline{\mathrm{Al}}$) Neutron flux of the Hisoshima bomb ($^{36}\underline{\mathrm{Cl}}$, $^{41}\underline{\mathrm{Ca}}$, $^{63}\underline{\mathrm{Ni}}$ (100 a)) Nuclear safeguards ($^{233}\underline{\mathrm{U}}$ (1.59 × 10^{5} a), $^{236}\underline{\mathrm{U}}$, $^{237}\underline{\mathrm{Np}}$ (2.14 × 10^{6} a), $^{239}\underline{\mathrm{Pu}}$ (2.41 × 10^{4} a), $^{240}\underline{\mathrm{Pu}}$ (6.56 × 10^{3} a), $^{242}\underline{\mathrm{Pu}}$ (3.73 × 10^{5}), $^{244}\underline{\mathrm{Pu}}$)

[a] Radionuclides measured in the respective area of application are listed in parenthesis. Underlined radionuclides indicate an anthropogenic origin. Half-lives are also given when they first appear.

4.1. Radiocarbon

Clearly, ^{14}C is dominating the field among all other radioisotopes. Although ^{14}C has by now been used 50 years for applications, new applications still emerge. As a result, about 90% of all AMS measurements worldwide are devoted to ^{14}C, with several facilities dedicated to ^{14}C measurements only. ^{14}C dating for archaeology is, of course, the classical application, and this field has benefited enormously from the reduction in sample size of AMS as compared to beta-counting.

4.1.1. 'Sensational' archaeology

^{14}C dating was developed by Libby and coworkers in the late 1940s [78–81]. The importance of ^{14}C measurements was quickly recognized, and in1960 Willard Libby was awarded the Nobel Prize in chemistry "for his method to use carbon-14 for age determination in archaeology, geology, geophysics, and other branches of science." It is probably fair to say that the basic applications of ^{14}C were all developed long before AMS appeared on the scene, because after the invention of ^{14}C dating in the late 1940s, beta-counting was the only method available to measure ^{14}C at natural levels. What AMS has really changed was the application to areas one could not touch with beta-counting because too much material was needed. Unfortunately, this sometimes led to dating objects, which, in hindsight, should better not have been touched at all. It is well-known that the ^{14}C measurements of the Shroud of Turin by three AMS laboratories resulted in a date of the Middle Ages [82], instead of the expected time around Christ's death. Although ^{14}C experts find it very difficult to imagine how ^{14}C dating could have gone that wrong, a widespread mistrust of the ^{14}C dating method evolved; 15 years later, the issue is still not resolved.

A much less sensitive, yet also quite sensational object was the Iceman 'Ötzi', a frozen man, which was accidentally discovered in 1991 by mountain hikers in a shallow glacial deposit in the European Alps. Radiocarbon dating of small pieces of tissue and bones revealed that Ötzi had lived between 5100 and 5350 years ago [83–85]. It is important to note that despite a high precision of the ^{14}C/^{12}C measurements, the "wiggles" in the ^{14}C calibration curve prevent

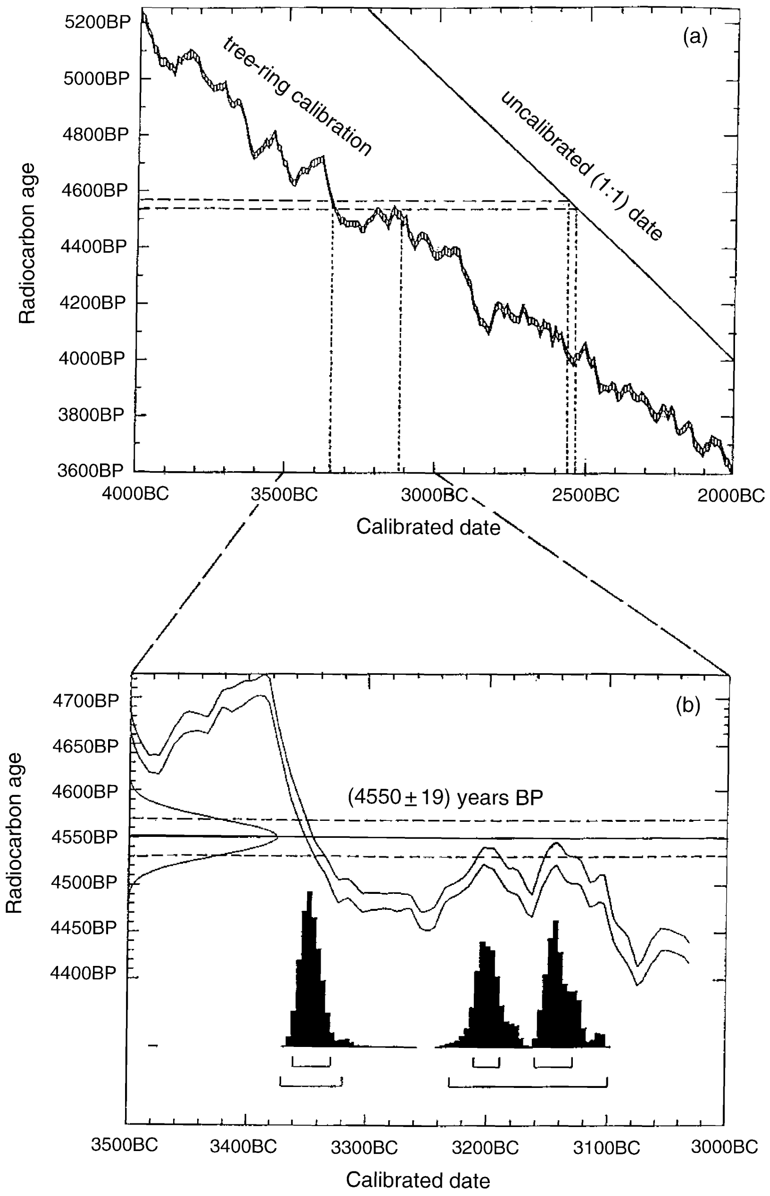


Fig. 2. The determination of the age of the Iceman from ^{14}C measurements at the AMS laboratories of Zürich [83,85] and Oxford [84], as displayed in a recent isotope study about various materials of the Iceman [86]. The combined (uncalibrated) radiocarbon age from these measurements is 4550 ± 19 years BP (Before Present = 1950 A.D.). The error represents the 68.2% (1σ) confidence value. The uncalibrated age is translated into a calibrated age with the help of the computer program OxCal using the INTCAL98 tree-ring calibration curve [87]. (a) Calibration curve from 4000 to 2000 B.C.. The straight line at 45° indicates a 1:1 transformation of the radiocarbon age into an uncalibrated calendar date. The intersection of the radiocarbon age with this line and the tree-ring calibration curve shows that the calibrated date is approximately 650 years older. (b) The enlarged “wiggly” section of the calibration curve leads to three different solutions for the calendar date spanning 250 years. The small rectangular brackets beneath the peaks indicate the distribution of the 68.2% (1σ) confidence ranges into three sections of 3360–3300 B.C. (29.3%), 3210–3190 B.C. (19.8%), and 3160–3130 B.C. (19.1%). The large brackets indicate the 95.4% (2σ) confidence ranges of 3370–3320 B.C. (34.3%) and 3230–3100 B.C. (61.1%).

a more exact and absolute age determination (Fig. 2). Later, several objects from the Iceman and a variety of small botanic remains found at the discovery site confirmed the Neolithic origin, and also gave some clues about the climatic conditions around this time [86].

4.1.2. Mapping the oceans

An example from a different field demonstrates the power of ^{14}C AMS particularly well. The National Oceanographic Sciences AMS (NOSAMS) facility at Woods Hole Oceanographic Institution was set-up in 1990 [19,88], with the goal to map the world oceans with ^{14}C measurements in an unprecedented way. As compared to the first mapping performed in the 1970s with beta-counting, which required 250-l water samples [89], 0.5 l of water now delivers enough carbon for a ^{14}C AMS measurement. As a result, over 13,000 water samples have been measured within the World Ocean Circulation Experiment project (WOCE), literally mapping the

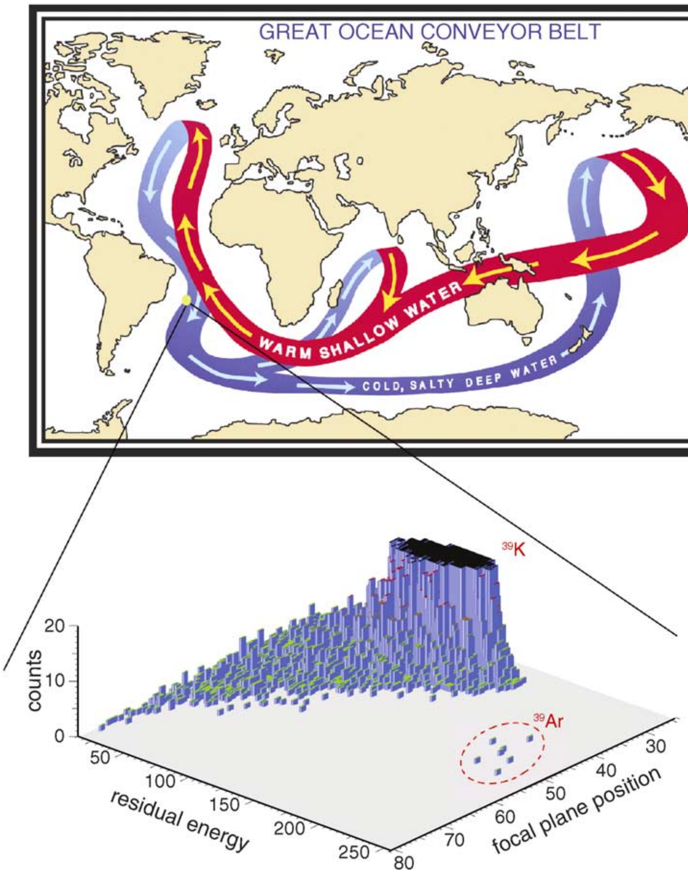


Fig. 3. The top of the figure displays a conceptual illustration of the Atlantic Conveyer Belt as first envisioned by Broecker [92], and derived from relative differences of ^{14}C ages between surface and bottom water [89]. As may be imagined those differences are smallest where the water dives into the abyss in the North Atlantic, and are largest where the aged bottom water up-wells in the North Pacific. Dissolved CO_2 of surface water reflects essentially the ^{14}C content of atmospheric CO_2. Also indicated is the ^{39}Ar measurement from a deep-sea sample (water depth 4700 m) off the eastern coast of Brazil measured with the gas-filled-magnet technique at the ATLAS accelerator of Argonne [17,27]. The clean separation from a huge ^{41}K background was accomplished at 232 MeV with the gas-filled-magnet technique. From a measured ^{39}Ar/Ar ratio of $(2.6 \pm 0.6) \times 10^{-16}$, which is 32% of the modern atmospheric ratio, and the half-life of 269 years, one obtains a ^{39}Ar decay age of 440 years. This is not unreasonable considering the time it takes for the deep water to travel from the North Atlantic to Brazil [89].

world oceans in three dimensions [90,91]. The goal of this global survey is to study the major circulation patterns of ocean currents. A detailed understanding of these patterns will be of utmost importance for global modeling of the climate on Earth. Very recently, AMS of ^{39}Ar mentioned above (Section 3.1) was developed as another oceanographic tracer (Fig. 3). Due to its conservative chemical behavior and a half-life of 269 years matching typical times of ocean currents, it adds an important analytical capacity to disentangle the complex dynamics of the oceans (see also Section 4.2.1 below).

4.1.3. Pushing the accuracy of ^{14}C dating

Although the precision of ^{14}C measurements has improved considerably in the course of the past 50 years, the precision of calibrated dates is often not good enough to solve some of the most burning questions in archaeology. One of those is the accurate timing of the ‘Minoan’ eruption of Thera on the Aegean island of Santorini some 3500 years ago. This vulcanic eruption is an important time marker to synchronise the interaction of cultures in the 2nd millennium B.C. in the East Mediterranean, and to link this date with the absolute Egyptian chronology. As seen in the example of Fig. 2, the main reason to prevent a very precise dating with ^{14}C are the natural fluctuations of the atmospheric ^{14}C content. However, if a set of ^{14}C dates from different times is available, a powerful technique called “wiggle matching” allows one to narrow down the uncertainty in absolute age determination. In essence, sophisticated procedures using Bayesian mathematics have been developed in the ^{14}C calibration programs (e.g., [93]) to combine ^{14}C dates with stratigraphic information of the excavation sites. In the most recent application of this method, one arrives at a more precise date of the Thera eruption, demonstrating how it is possible to ‘cut thin slices with a blunt knife’ [94].

A problem of similar archaeological significance is the exact timing of the Iron-Age chronology between 1000 and 800 B.C., trying to establish a consensus between dates from Egyptian Pharaos with those of Hebrew Kings [95–97]. Again, the extensive use of stratigraphic sequencing (wiggle matching) and inter-laboratory comparison of ^{14}C results squeezes the

^{14}C dating method to its limit. A caveat in these interesting efforts should be mentioned: the improvement of 'bare' ^{14}C dates by wiggle matching depends critically on the stratigraphic assumptions and intricacies of the Bayesian method itself [98]. Hence, it is very important to use samples from a stratigraphically secure context. Much remains to be done, before one can confidently push the precision of ^{14}C dating beyond its inherent uncertainty.

4.2. Anthropogenic radioisotopes

The production of radioisotopes by man can be simply divided into two groups: unintentional and intentional. The first group comprises releases from nuclear weapons testing and nuclear industry. The intentional production of radioisotopes is chiefly concerned with the production of short-lived radioisotopes for medical use, for both diagnostic and therapeutic purposes. In connection with AMS, the sensitive detection of long-lived radioisotope tracers allows unique in vivo studies in both animals and humans. Because of the long half-lives, strong isotope signals add only negligible amounts of radioactivity.

4.2.1. Releases from nuclear weapons and nuclear industry

Probably, the most cataclysmic events created by man were the atomic bomb explosions of Hiroshima and Nagasaki in 1945. AMS measurements of ^{63}Ni ($t_{1/2} = 100$ a) produced by the ^{63}Cu (n, p)^{63}Ni reaction in copper-containing materials (e.g., lightning rods) allowed the assessment of the fast-neutron flux to which materials and humans were exposed from these bomb explosions [99,100]. Within the so-called Life Span Study [101], a cohort of about 120,000 individuals from Hiroshima and Nagasaki including non-exposed controls have been followed to study late effects of ionising radiation. Information on fast- and slow-neutron fluxes are essential to evaluate the relation between neutron dose and possible health effects. An AMS study of ^{41}Ca produced by slow-neutron activation of ^{40}Ca in tooth enamel of atom bomb survivors were recently initiated at the Munich 14-MV tandem [102]. When this study is finished, a more secure basis for both the fast- and the slow-neutrons dose for the above mentioned Life Span Study will be established.

Between 1950 and 1963, the intense atmospheric nuclear weapons testing programs produced the so-called 'bomb peak' of radioisotopes in the atmosphere. The total explosive power of these tests was equivalent to about 25,000 Hiroshima bombs (∼500 Mt TNT as compared to ∼20 kt TNT equivalent). Among the radioisotopes produced, ^{14}C turned out to serve as a particularly useful tracer to study the global carbon cycle dynamics [103]. At the time of the atmospheric Nuclear Test Ban Treaty in 1963, the ^{14}C content in the atmosphere had increased by 100% (factor two) above the natural concentration. The distribution of this surplus to other carbon reservoirs on Earth (biosphere, hydrosphere) can be followed closely to this day. The atmospheric ^{14}C content has meanwhile decreased to about 10% above natural. Please note that the radioactive decay of ^{14}C changes the ^{14}C/^{12}C ratio by only 1% in 80 years; whereas, the carbon cycle dynamics decreases the bomb-peak ^{14}C/^{12}C ratio in atmospheric CO$_2$ by ∼1% per year (like a much faster running ^{14}C clock). Since the ^{14}C/^{12}C ratio in atmospheric CO$_2$ has been closely monitored since the 1950s [103], it provides a calibration curve to date very recent objects with an uncertainty of only 1–2 years. In an application to forensic medicine, the time of the death of two persons could be determined to have occurred in the late 1980s, with a relative time difference of about 1 year [104].

In connection with bomb-peak dating, it is perhaps useful to point out a peculiarity of ^{14}C dating in general. Just as the current example of dating does not depend on the half-life of ^{14}C, also archaeological dating does not depend on the actual half-life value, as long as a calibration of the ^{14}C content is available (compare Fig. 2). In contrast, for absolute dating (see Section 5.2. below) the half-life value need to be known.

Other, semi-intentional sources of radioisotopes are the releases from nuclear fuel reprocessing plants into the ocean. Here, the long-lived fission product ^{129}I ($t_{1/2} = 1.7 \times 10^7$ a) released from La Hague in France and Sellafield in England is being utilised as an oceanographic tracer [105,106]. As mentioned in Section 3.4, ^{129}I/^{127}I ratios can now be measured carrier-free from only 0.1 l of seawater [73], which allows for a fast and efficient way of tracing ^{129}I throughout the oceans. On a global scale, the strong ^{129}I input from the reprocessing plants provides a source well defined in location and time. The AMS measurement of ^{99}Tc ($t_{1/2} = 2.11 \times 10^5$ a), another fission product released into the ocean, has recently been developed at the 14-MV tandem accelerator facility of the Australian National University at Canberra [107]. Here, the goal is to make a measurement on ocean water samples of 0.25 l. Since technetium has no stable isotopes, rhodium is added, and ^{99}Tc/^{103}Rh isotope ratios are measured.

4.2.2. Biomedical tracing

It was recognised early on in the AMS development, that ^{14}C tracer studies might be useful for studies in medicine [108]. From 1990 onward several AMS groups started programs in biomedical application of AMS, with Lawrence Livermore National Laboratory taking the lead [109,110]. Again, ^{14}C is the most-used radioisotope, allowing for a large variety of metabolic studies, e.g., of carcinogenic compounds [111]. In order to speed up drug research, a new 5-MV Pelletron tandem AMS facility was set-up at York University in the UK, dedicated solely to biomedical use [112]. It is interesting to note that this facility, now called Xceleron, was sponsored by funds provided by large pharmaceutical companies, and is operated essentially on a commercial basis only.

One of the most promising radioisotopes measured with AMS for biomedical studies is ^{41}Ca ($t_{1/2} = 1.04 \times 10^5$ a). The very long half-life, a low decay energy, and a very low natural isotopic abundance ($^{41}Ca/^{40}Ca \sim 10^{-15}$, see [102]) allows for a strong isotopic signal to be injected into living humans ($\sim$5 ng of ^{41}Ca). This generates an initial $^{41}Ca/^{40}Ca$ ratio of $\sim 5 \times 10^{-9}$ in urine samples [113]. One can thus follow this tracer over a large dynamic range. Once a human has received a ^{41}Ca dose mentioned above, the skeleton is essentially labeled with ^{41}Ca for life, settling after a few months at a $^{41}Ca/Ca$ ratio in the 10^{-11} range in the analysed urine samples, with a very slow decrease with time [113,114]. This then, makes it possible to follow the metabolism of bone for many years and to study its response to particular drugs [113]. An excessive loss of bone mass with age (osteoporosis) is a widespread phenomenon, and there is great hope that these ^{41}Ca studies will eventually help to find a cure for this problem.

4.3. *Other natural radioisotopes*

As compared to the overwhelming use of ^{14}C, other radioisotopes seem to pale in importance. However, there are very interesting questions which can only be answered with these other radioisotopes.

4.3.1. *Exposure dating*

Although the bulk of cosmogenic radionuclides are produced by cosmic-ray interaction in the Earth's atmosphere [115], secondary cosmic-ray particles (e.g., neutrons and muons) penetrate to the surface and produce radionuclides through nuclear reactions in the surface material [116]. Depending on the material composition, and the altitude and latitude of the location, production rates of ^{10}Be and ^{26}Al in quartz (SiO_2) are in the order of 10–100 radioisotopes per year per gram of exposed material [117]. In a setting where the surface material was once well shielded to prevent radioisotopes production, the time when exposure started can be determined from the accumulated radioisotope concentration. Examples for such measurements are: the date of the retreat of glaciers [118], the date of metoritic impacts [119], river incision rates [120], the date of landslides [121], the exposure time of landbridges [122], and cosmic-ray background for geochemical neutrino detection [123].

A refinement of the method, which is of particular interest to geomorphological studies, is the determination of erosion rates from exposure dating. In essence, the physical half-life is shortened by the erosion to an effective half-life determining the temporal built-up of a particular radioisotope. Given a theoretical concept and knowing the production rate for radioisotopes at the particular location, erosion rates can be deduced [116].

4.3.2. *Ice cores*

Ice cores from the large polar ice sheets of Greenland and Antarctica represent the most detailed paleoclimatic record on Earth, with the longest record extending now back to 740,000 years [124]. Information about the atmosphere in the past is deposited in ice by precipitation and by occluded air bubbles. Once the ice forms, the information is literally frozen in place. In addition to stable isotope signals, such as δD and $\delta^{18}O$, which provide information on paleotemperatures, the record of cosmogenic radioisotopes (^{10}Be, ^{26}Al, ^{36}Cl) allow one to trace variations in cosmic-ray production. These variations are chiefly caused by the variable shielding effects on cosmic-ray primary protons by the solar and terrestrial magnetic fields. In this way, the geomagnetic field between 20,000 and 60,000 years has been reconstructed from ^{10}Be and ^{36}Cl measurements in the GRIP ice core from Greenland [125]. In rare cases concentration changes of radionuclides have been observed which seem to be caused by changes of the primary cosmic-ray intensity. For example, excessive ^{10}Be concentration peaks in the Vostok ice core from Antarctica indicate some possible cosmic events around 35,000 and 60,000 years ago [126]. New ice cores are likely to be drilled in the next couple of years (e.g., [127]), and radioisotope measurements will add important aspects to the wealth of information stored in these archives.

4.3.3. *Supernova remnants on Earth*

One of the most exciting applications of AMS is the search for radioisotopes in the million-year half-life range, which were produced in 'close' supernovae explosions and may be detectable in proper archives on Earth. (Geologists actually call radioisotopes in this time range 'short-lived', because only those which survived the age of the solar system ($\sim$4.6 billion years) are considered to be long-lived.) If such a supernovae occurred within a few half-lives of the respective radioisotope, there is a finite chance to find it. As mentioned already in Section 3.1, the first AMS experiment of this kind was performed at the Munich 14-MV tandem AMS facility. Minute traces of ^{60}Fe ($t_{1/2} = 1.5 \times 10^6$ a) were found in a deep-sea ferromanganese crust, indicating an anomalous increase in ^{60}Fe some 3 million years ago [44]. Improved measurements recently gave a more convincing picture (Fig. 4) [45], suggesting that this event may indeed have been caused by a supernova explosion 2.8 million years ago at a distance of a 'few tens of parsec' ($\sim$100 light years). Other possible candidates for tracing such cosmic events are ^{182}Hf (8.9×10^6 a) and ^{244}Pu (8.1×10^7 a). Experiments to develop the AMS technology for these radioisotopes have started at several AMS facilities, and first searches were initiated for ^{182}Hf [128] and ^{244}Pu [129–131].

5. Outlook

On the one hand, AMS is a well-established technique with efforts to simplify the method by miniaturisation of the facilities, particularly for ^{14}C measurements. On the other hand, AMS is still an evolving technology for many other radioisotopes and their applications. In general, predictions what might happen in a couple of years are not very meaningful. Niels Bohr expressed this once so beautifully by saying:

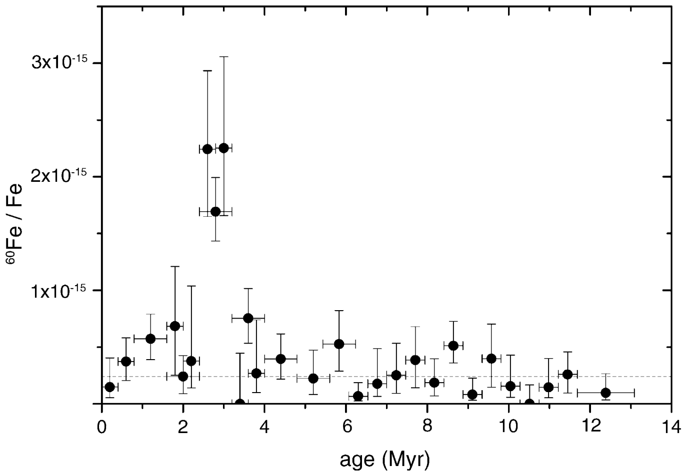


Fig. 4. Depth profile of ^{60}Fe/Fe ratio measurements in a deep-sea ferromanganese crust from the Pacific Ocean (4830 m water depth), measured at the Munich 14-MV tandem facility [45]. The slow-growth rate of the crust (2.5 mm per million years) converts depth to the time scale shown in the figure. A clear ^{60}Fe anomaly above the background level of 2.4×10^{-16} is seen at an age of 2.8 million years. This indicates a possible extraterrestrial event, and according to astrophysical models would be compatible with the deposition of ejecta from a supernova a few tens of parsec (~100 light years) away from the Earth [45].

"Predictions are always difficult, especially of the future." In the following, we will discuss a few ideas, which may initiate some new thinking about AMS.

5.1. The 'ideal' atom-counting device

It is probably not an exaggeration to say that AMS is an analytical technique, which allows one to trace long-lived isotopes with unrivaled versatility and sensitivity. However, Laser Resonance Ionisation Spectroscopy (RIS) demonstrated already during the early years of AMS that single-atom detection is possible in favorable cases [132]. Later, Resonance Ionisation Mass Spectrometry (RIMS) made big strides to reach the isotope selection sensitivity of AMS, but the physical limitation from the tails of the Lorentzian line shape of resonance processes [133] required multi-step isotope enrichments to measure radioisotope concentrations at natural levels. However, by increasing dramatically the observation time of atoms with the magneto–optical-trap (MOT) technique allowed one to reach the necessary isotope selectivity without isotope enrichment [134]. This has been demonstrated recently by measuring ^{81}Kr/Kr ratios in the 10^{-13} range to date old groundwater samples from the Nubian aquifer in Egypt [135]. Measuring ^{81}Kr/Kr ratios in the 10^{-13} range was previously only possible with AMS [43], or by combining laser resonance ionisation mass spectrometry with a rather elaborate multi-step isotope enrichment procedure [136]. The various efforts of laser-based techniques are summarized in a recent review paper by Lu and Wendt [137].

Whereas AMS has to make a big effort to separate stable-isobar background (e.g., ^{81}Br in the case of ^{81}Kr detection), it is very difficult for laser resonance processes to reach the isotopic selectivity required for natural samples. Ideally, one would like to combine the power of lasers to separate isobars (elements), with the power of AMS to separate isotopes. Although a combination of element-selective laser ion sources [138] with AMS seems feasible, such a system has not yet been realised. Here, clearly, lies a challenge for the future.

5.2. On the possibility of absolute ^{14}C and ^{41}Ca dating

Although ^{14}C dating is an extremely useful tool for archaeology and other fields, it has the problem of depending on a calibration. As seen from Fig. 2, the natural wiggles in the calibration curve convert the uncalibrated, high-precision radiocarbon age into a rather unprecise absolute date. In principle, this could be avoided, if one would be able to make an absolute dating measurement [139]. Absolute dating using a radioisotope means that both parent and daughter products are measured. A famous example of this method is the potassium–argon (^{40}K/^{40}Ar*) technique. The * indicates that only the radiogenic ^{40}Ar should be considered. It can easily be shown that the ^{40}K/^{40}Ar* ratio does not depend on how much ^{40}K was initially present. Similarly a ^{14}C/^{14}N* ratio measurement would not require a calibration to know the initial ^{14}C. Because of the overwhelming amount of ^{14}N present in any natural setting, it seems impossible to detect the minute radiogenic contribution of ^{14}N*. However, Szabo et al. [139] discussed the kinematics of the ^{14}C beta decay, which generates a maximum recoil energy of only 7.3 eV for the ^{14}N* atom. This low-recoil energies is comparable to the binding energy of atoms in a molecule, and results in a finite retention probability for the ^{14}N* atom. Starting from some organic molecule containing a ^{14}C atom, the result is a molecule with different chemical and physical properties. It is not entirely unfeasible to think of using the AMS technique developed to identify doubly charged negative ions [140], to find a ^{14}N* ion in the break-up of an organic molecule. For such an experiment, one of the problems one would have to solve is the production of a beam of unaltered molecules from the material. At this point, absolute ^{14}C dating is still a dream.

Similarly, one can dream of developing an absolute dating method for ^{41}Ca [26]. Here the favorable electron-capture decay of ^{41}Ca to ^{41}K* generates an even lower recoil energy of maximal 2.3 eV. Thus, the retention probability should be large. Again, the question of distinguishing the minute ^{41}K* signal from normal ^{41}K is formidable, and perhaps impossible. Yet, the potential of a functioning ^{41}Ca dating method is obvious: a favorable half-life of 104,000 years and the possibility to date bones directly would most likely be of great value in studying the ancestry of our own species. So, the dream goes on.

5.3. Searching for the unknown

AMS can be applied to many fields, as seen from Table 2. It has developed into an invaluable analytical tool to study physical and chemical processes in our environment. Since it is likely that both the number of facilities and their use will grow, AMS has a bright future. But AMS has properties, which make it unique in the sense that one can also use it to search for the hitherto unknown. As mentioned in the introduction, such experiments were performed right after the invention of AMS to search for superheavy elements [12]. A more focussed search of this kind was performed at the tandem accelerator of the University of Phildelphia for the isotope 294 of the platinum-like element with $Z = 110$ [141].

In a different field, a low-energy search for free quarks was performed at the 700-kV Cockroft–Walton proton injector of Fermi Lab [142]. A more extensive search for fractionally charged particles was later performed at the 3-MV Pelletron tandem accelerator of the Kellogg Radiation Laboratory in Caltech [143].

A search for anomalously heavy isotopes of low Z nuclei was performed at the Rochester 10-MV tandem accelerator [144], in the sprit of similar experiments of Luis Alvarez at Berkeley 10 years earlier [7]. The rational for these experiments are only partly driven by theoretical prediction. Another part is simply the ability to tune the AMS machine to white spots on the landscape of nuclides, far away from known territory. With the increasing computer control of accelerators, it may be worthwhile to revive these searches in a more serious and general way. The probability is very faint to find something relevant. Yet it is intriguing to go on with these searches. Someone simply has to do it.

There are a number of other species one could search for with AMS: strange matter containing roughly equal numbers of up, down, and strange quarks may have been formed in phase transitions of the early universe [145–147], and may reveal themselves as low Z, high mass particles. Finally, dark matter is one of the big unsolved problems of our time [148]. Why not searching for it with AMS?

The list of hypothetical particles is large, and somewhere there ought to be something unknown to be discovered. It is perhaps worthwhile to devote a certain fraction of the time of AMS facilities for these searches. The current status of largely computer-controlled AMS facilities allows one to perform automatic searches over a large parameter space. In the true spirit of science, we should simply try to look beyond the horizon.

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