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# Single Atom Counting with Accelerators

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**A new method of measuring small concentrations of long lived radioisotopes is beginning to have a major impact in many areas of science, where they are used for dating or tracing.**

Direct detection of radioisotopes with conventional mass spectrometers is possible when the potential background atoms, in particular stable isotopes of the same mass (isobars) or molecules of similar mass are present in sufficiently low concentrations. Most of the long lived radioisotopes of interest for dating purposes however, occur in such small concentrations that their peak in the mass spectrum is obscured by the stable isobar and molecule distributions. Table 1 gives the half-lives, the approximate ranges of natural concentrations in terrestrial and meteoritic materials, and the interfering stable isobars of some of these radioisotopes. Up to now, none listed in the table has been directly observed by classical mass spectrometry or by the more modern separation-identification technique using lasers.

The key idea of the new AMS technique which allows us to measure directly such small concentrations is the acceleration of the sample atoms to MeV energies and to use various filter processes and particle identification techniques developed for nuclear physics research to eliminate the isobaric and molecular interferences. The detection methods used for each radioisotope depend on the dominant background atoms and these in turn depend on the specific accelerator used.

Physicists in many places have been developing these techniques since the idea was first mooted about six years ago in Berkeley and Rochester. Most interestingly it was found that the sensitivity required to detect such isotopes can be achieved quite easily with virtually any existing particle accelerator combined with a reasonable mass filter and particle detection system. However, it was soon realized that there is an enormous difference between detecting a radioisotope at its natural level and measuring the relevant isotope ratios with the high precision required for dating. For instance, to compete with the classical radiocarbon dating method, introduced by Libby in 1946, the $^{14}C/^{12}C$ ratio has to be measured with a precision of at least 1%. An error of 1% in the ratio measurement corresponds to an uncertainty of 80 years in dating, independent of the sample age.

The problems encountered in transforming an existing particle accelerator into a high precision dating tool are considerable and have been solved only recently for one type of accelerator, notably the tandem van de Graaff. For this reason the following description of the AMS method and some of its applications will be restricted to this type of accelerator only, taking the dating facility of the ETH Zurich as an example.

### AMS with Tandem Accelerators

The schematic layout of the facility is shown in Fig. 1 in the configuration used for radiocarbon dating. It consists of an electrostatic EN tandem accelerator delivering voltages up to 6 MV, an ion source attached to a 90° double focussing inflection magnet at the low energy side and an electrostatic energy selector followed by another double focussing 90° analyzing magnet and a particle detector system at the high energy end. The currents of the abundant stable isotopes are measured directly with conventional Faraday cups, whereas the rare isotopes are identified and counted individually by means of a $\triangle E/E$ gas counter telescope. With such an arrangement both the atomic and the mass

*Fig. 1 — Schematic layout of the ETH accelerator dating facility.*

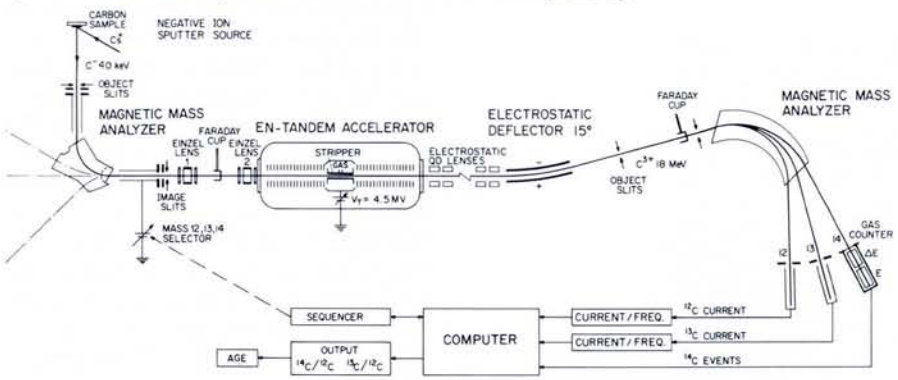


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**Table 1 — Long-lived Radioisotopes**

| Isotope | Half-life (a) | Approx. Range of Terrestrial Conc. [1] | Production Process | Interfering Isobars |
|---|---|---|---|---|
| $^{10}$Be | $1.6 \times 10^6$ | $10^{-8}$ - $10^{-14}$ | Spallation | $^{10}$B |
| $^{14}$C | $5.7 \times 10^3$ | $10^{-12}$ - $10^{-16}$ | $^{14}$N (n,p) $^{14}$C | $^{14}$N [2] |
| $^{26}$Al | $7.4 \times 10^5$ | ~ $10^{-14}$ | Spallation | $^{26}$Mg |
| $^{32}$Si | $1.3 \times 10^2$ | $10^{-15}$ - $10^{-17}$ | Spallation | $^{32}$S |
| $^{36}$Cl | $3.1 \times 10^5$ | $10^{-12}$ - $10^{-14}$ | Spallation | $^{36}$S, $^{36}$A [2] |
| $^{41}$Ca | $1.3 \times 10^5$ | $10^{-14}$ - $10^{-15}$ | $^{40}$Ca(n,γ)$^{41}$Ca | $^{41}$K |

[1]) Compared to the stable isotope of the same element.
[2]) These elements do not form stable negative ions.

number of each registered particle can be identified.

Tandem accelerators require negative ions, which are accelerated to high velocities in the first section through the large potential difference. These ions are converted into multiply charged positive ions and further accelerated through the same potential difference. The energy of the ions emerging from the accelerator is given by $E = eU\ (1 + q)$ where $eU$ is given in eV and $q$ is the charge state. Typically 3-6 MV are required to produce $3^+$ charge states with optimum efficiency. In our case, optimum beam intensity for $C^{3+}$ is obtained with 4.5 MV giving a final beam energy of 18 MeV.

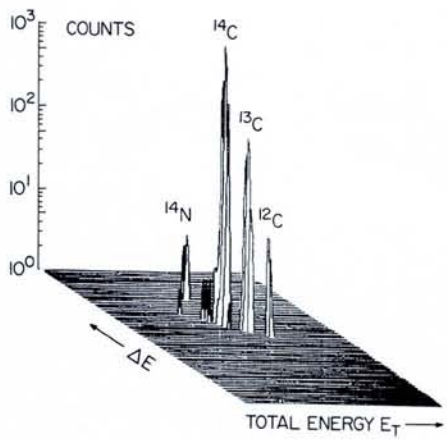
The negative ions are produced by a Cs sputter source. The sample atoms, e.g. carbon, are sputtered and negatively charged, using an intense (0.5 - 1 mA) and well focussed 40 keV Cs beam. The negative ions are extracted, accelerated to the same energy and focussed with an electrostatic einzel lens onto the object slits of the inflection magnet. For graphite-like samples, almost 10% of all atoms are converted into negative ions and more than 40 μA of carbon beam can be obtained. Up to 25 samples can be loaded simultaneously which in turn can then be placed on the sputter-electrode, kept at -40 kV, by means of a computer controlled manipulator.

The 90° inflection magnet provides a first low resolution stage of mass analysis. Only negative ions of similar masses, e.g. isobars, molecules and tails of particles having adjacent masses are injected simultaneously into the accelerator. In the case of carbon, the isobar problem is negligible as $^{14}$N simply does not form negative ions, living long enough to survive the acceleration process. On the other hand, negative molecule ions such as $^{12}CH_2^-$ and $^{13}CH^-$ are observed in abundance. For a modern sample, the molecule to $^{14}$C ratio is found to be of the order of $10^9$. One of the advantages of using a tandem accelerator is that all these molecular components are broken up upon passing through the stripper at the tandem terminal, provided enough electrons are removed from these molecules. Up to now, no triply charged stable or metastable molecules have been observed to exist. Therefore any beam emerging from a tandem accelerator will be contaminated only with fragments of molecules, if charge states of $3^+$ or higher are selected out. A normal mass filter can then be used to separate these fragments from the wanted rare isotopes.

Fig. 2 shows a three dimensional representation of a particle spectrum measured with the counter telescope. The sample was modern carbon with a $^{14}C/^{12}C$ ratio of $1.2 \times 10^{12}$. As can be seen, the $^{14}$C peak is well resolved from the few background peaks. These are produced by molecular fragments which somehow outwitted the high energy mass filter. Similar tests with dead carbon have indicated a dating limit of about 60000 years.

In order to determine the isotopic ratios, required for a $^{14}$C dating with high precision, not only the rare but also the abundant stable isotopes should be accelerated and measured at the high energy side of the accelerator. Most of the existing accelerators, however, in particular electrostatic tandem accelerators are neither designed to handle large beam intensity variations occurring when switching from the rare to the abundant isotope, nor capable of sustaining intense heavy ion beams continuously. Consequently the high beam currents required for a fast dating can only be injected in short pulses, typically of 20 μs length for a 40 μA $|\ ^{12}C^-$ beam. In our case, such short pulses are generated by applying periodic (25 Hz) high voltage pulses to the insulated vacuum chamber of the inflection magnet, as indicated in Fig. 1. Usually the magnet remains set for the rare isotope and the pulsing system is used to change the momentum of the stable isotopes to the values required for injection. This allows not only a quasi-continuous monitoring of all isotopes of interest, but most of the time remains available for the measurement of the rare isotope, for which the statistical error usually is the limiting error.

*Fig. 2 — Three-dimensional representation of $^{14}$C data measured with the $\Delta E/E$ gas counter telescope. $\Delta E$ is the energy loss in the first section, $E_T$ the total ion energy measured in both sections of the counter. The sample was modern carbon, e.g. $^{14}C/^{12}C = 1.2 \times 10^{-12}$. The background peaks represent fragments from negative molecule ions $^{12}C\,H_2^-$, $^{13}C\,H^-$ and $^{14}N\,H^-$.*




Extensive tests have shown that it is indeed possible in this way to measure $^{13}C/^{12}C$ ratios reproducibly and over long time periods with an accuracy of about 0.2%. Using a modern sample, only two minutes of measuring time are required to collect $10^4$ $^{14}$C atoms. In principle, the same arrangement can be used to determine ratios of any rare isotope of interest, provided negative ions are being formed. Our system is presently being used to measure concentrations of $^{10}$Be, $^{26}$Al, and $^{36}$Cl as well.

There are many applications of AMS to problems using these long-lived radioisotopes for dating or as tracers. To illustrate the potential of the method, a few examples using $^{14}$C and $^{10}$Be will now be discussed.

### Radiocarbon Dating

During the past three decades, radiocarbon dating has been developed into a precise, widely used technique, calibrated with tree rings back over 8000 years. $^{14}$C has a half-life of 5730 years and is the most abundant long lived radioisotope in nature. It is continuously produced in the atmosphere by the reaction $^{14}$N (n,p) $^{14}$C, where the neutrons are secondary products of cosmic ray interactions with the atmosphere. $^{14}$C oxidizes to $CO_2$ and is distributed into the reservoirs of the Earth, including the atmosphere, the ocean and the biosphere. The equilibrium specific activity in the atmosphere and biosphere was 13.5 decays per minute and gram of carbon in 1950, the reference year of all radiocarbon dating work.

With the traditional beta decay counting technique, about 12 h of measuring

*Fig. 3 — Front and back faces of some of the twelve recently discovered "pre-mayan" codices. Only the few mg of leather, shown below, were required to date this foil.*

time are required to determine the $^{14}C/^{12}C$ ratio of a modern 1g sample with 1% accuracy. This measuring time would increase by a factor of 1000 for the same sample having an age of 57300 years (10 half-lives). Clearly much more material would be required to reduce the measuring time to an acceptable level. With AMS, this severe restriction is removed. A modern sample can now be dated to 1% accuracy within 2 min, as mentioned above, but even more important, only about 6 µg of carbon are consumed for such a measurement: in practice, 0.2 - 2 mg of carbon are required to prepare a good sample.

This almost destruction free dating method opens up new possibilities in many fields where often only small samples are at hand, particularly in archaeology, geology, oceanography and climatology. Even art objects which otherwise would be completely destroyed, can now be checked for authenticity. Such an example is given in Fig. 3, which shows foils of very thin but stiff leather weighing only a few grams. The full set consists of a total of 12 similar foils which are completely covered with mayan hieroglyphs on the front side and, even more interestingly, with corresponding comments in Spanish on the back, presumed to have been added in the 16th century. This unusual archaeological object which was found in rather mysterious circumstances in Mexico 10 years ago, was believed by the experts to have an age of at least 2000 years. Since the conventional decay counting technique would practically consume a whole foil, an AMS measurement was made, for which only a few mg of leather was required. The result: an incredible and beautifully made forgery, not older than about 20 years. Such a precise assertion can be made in this case, because the observed $^{14}C/^{12}C$ ratio is larger (about 50%) than that of the 1950 standard. This indicates that the animal, from which the leather was taken, must have lived during the period of enhanced $^{14}C$ activity produced by the nuclear weapon tests performed in the atmosphere during the fifties and sixties.

The polar ice shields of our earth are another most interesting subject. All cosmogenic radioisotopes have accumulated here during the past $10^5$ - $10^6$ years in a practically undisturbed stratigraphy and can be used to study changes in the production rates due to solar and terrestrial events. By measuring the $^{14}C$, trapped as $CO_2$ in ice, a time scale for events occurring over possibly the past 500000 years can be established. However, the total amount of trapped $CO_2$ is extremely small: from 20 kg of ice typically only about 1 $cm^3$ $CO_2$ (NTP) can be extracted; from this a solid carbon sample of 0.5 mg can be produced. The technique needed to extract such small amounts of gas from ice and convert it contamination-free into solid carbon samples suitable for AMS is presently being developed by the group of Prof. Oeschger at the University of Bern. First tests with such samples are so encouraging that dating of ice cores recovered from the various deep drilling sites in Greenland and Antarctica now appears to be feasible.

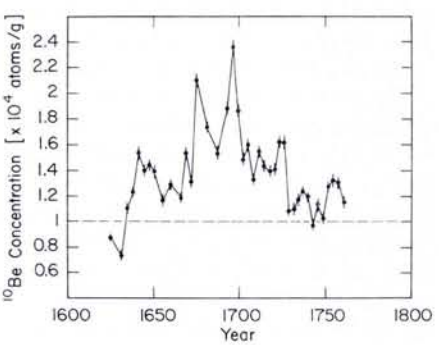
$^{14}C$ can also be used as tracer to study the large scale operation of the oceanographic system. Of the utmost interest is the amount of perturbance of this system by the great warming which accompanied deglaciation 10000 years ago. The difference in the rates of change of the $^{14}C/^{12}C$ ratios between surface and bottom water at that time is a measure of this effect, because it determines the ventilation time of the deep sea. This information is kept by organisms known as benthic and planctonic foraminiferas the first of which is known to live on the sea floor, while the other lives in the surface water only. Both are stored in marine sediments and can be selected out from deep sea drill cores. However the benthic forams are very small in size (250 µm) and very rare. It takes almost a week to select under the microscope the 200 shells required to gain about 1 mg of carbon. Such measurements are presently being carried out in collaboration with Prof. Broecker, Columbia University. First results indicate that the wanted information can indeed be obtained in this way.

**Beryllium-10**

$^{10}Be$ has a half-life of 1.6 Ma and is the second most abundant long lived radioisotope after $^{14}C$. It is produced in the upper atmosphere by cosmic ray spallation of atmospheric nitrogen and oxygen. $^{10}Be$ gets attached to aerosols within a very short time and is removed from the atmosphere by precipitation. It is deposited in snow and ice, in the topsoil, and the biosphere, or it enters the lake and the ocean and accumulates in sediments as well as in manganese nodules growing on the sea floor. The residence time of $^{10}Be$ in the atmosphere is only 1-2 a. This isotope can thus be used to study production rates on a short time scale ($<<$ 1 Ma), as well as for dating on a multi million year time scale.

The importance of $^{10}Be$ as a means of investigating the modulation of cosmic ray intensity by solar activity and geomagnetic effects has been recognized for more than 20 years, but the number of measurements using beta decay counting remained small because of the small decay rate and its low natural concentration, which varies from about $10^4$ atom/g ice to about $10^{10}$ atom/g manganese nodule. A sample containing $10^7$ Be atoms emits only five beta-rays a year, far below the background level of any beta counter system. With AMS, the $^{10}Be$ concentration of such a sample

*Fig. 4 — $^{10}Be$ concentration in units of $10^4$ atoms/g ice as a function of age in the Milcent ice core (Greenland) during the "Little Ice Age". The age was determined by measuring the seasonal variation of $\delta^{18}O$. The dashed line indicates the present value, averaged over the last 80 years.*



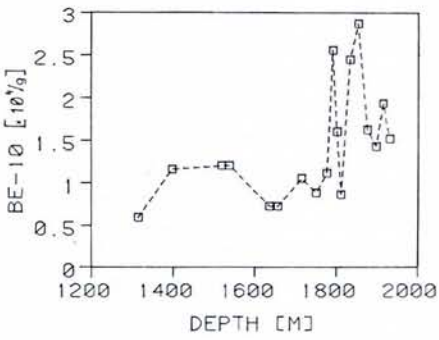



*Fig. 5 — $^{10}$Be concentration profile in the Dye 3 ice core (Greenland). The depth range from 1780 m to 1940 m below the surface corresponds to an estimated time range of 13000-30000 BP. The data indicate a rapid transition from Wisconsin to Holocene.*

can easily be measured within half an hour to an accuracy of a few percent.

The fact, that $^{10}$Be concentrations of up to 50 samples can be measured with high precision in only about 24 hours allows us to study full profiles in ice cores, deep sea sediment cores, manganese nodules and meteorites. Fig. 4 shows such a high resolution measurement made on an ice core recovered from Milcent, in the central part of Greenland. This measurement was made with a time resolution of about three years and covers the time period from 1620 to 1760 AD known as the Maunder minimum which coincided with the temperature minimum of the little ice age in Europe. This period was characterized, as we know from the observation of astronomers, by a very quiet Sun and a harsh climate, as is beautifully illustrated by many famous Netherland painters of the time.

The absolute timescale was established by $\delta^{18}$O measurements, which allowed the seasonal variations within each year back to 10000 years to be determined as with tree rings. It is interesting to note that at this time the production rate for $^{10}$Be not only varied strongly, but was up to a factor of two larger than the present day value averaged over the past 80 years. A $^{10}$Be measurement required 1-2 kg of ice which was cut from the 400 m core. Approximately 1 mg of Be was added to the melted ice and both isotopes were extracted chemically as BeO. The $^{10}$Be/$^{9}$Be ratio of such a sample was typically of the order of $10^{-12}$ - $10^{-13}$.

Another measurement made on a 2000 m long ice core, drilled in the frame of the Greenland Ice Sheet programme at Dye 3, revealed even larger fluctuations as shown in Fig. 5. The depth range investigated covers an estimated time range of about 4000 - 50000 a, and includes the last ice age which ended rather abruptly 1300 years ago. During this time the $^{10}$Be concentration was up to a factor of three larger than the post-glacial values. It is an open question yet whether this unique signal reflects higher production rates or lower precipitation or both.

Manganese nodules are fascinating subjects. They are found in many places on the deep sea floor and are the result of a continuous accumulation mainly of Mn and Fe (as hydroxide) around a nucleus, e.g. a fish tooth or simply a piece of rock. A cut through such a nodule recovered from the southern part of the Pacific at a depth of almost 5000 m is displayed in Fig. 6. These nodules started to grow 15-20 Ma ago. They represent a fortune mainly because of the few percent of very rare metals including Ni and Co which are found in the crusts. Its amount is estimated to exceed that of the known continental deposits by orders of magnitudes. Accordingly their origin and growth rate are of much interest. Again AMS opens the possibility of gaining such information by measuring for instance the $^{10}$Be, which, like the other elements, is continuously accumulated in traces. Only about 100 mg of material has to be processed to obtain enough $^{10}$Be for a measurement.

The observed $^{10}$Be profile through such a nodule, starting from the surface towards the bottom is given in Fig. 7. In this semi-logarithmic plot cleary two straight lines can be fitted into the data, indicating that short term variations observed in ice are averaged out over sufficiently long time periods, so that this isotope can indeed be used as a geochronometer on a multi-million time scale. Futhermore, the slopes of the lines indicate the growth rate of the nodules and it can be seen that it changed by almost a factor of two about 6 Ma ago. Presently we can only speculate what

*Fig. 6 — Typical cross-section through part of a manganese module found in the southern Pacific at a depth of almost 5000 m. The nucleus is a piece of basalt rock.*

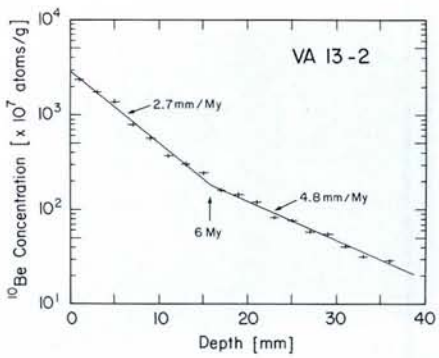




*Fig. 7 — $^{10}$Be concentration profile of the manganese crust from the surface towards the nucleus. 6 Ma ago, the growth rate changed rapidly from 4.8 mm/Ma to 2.7 mm/Ma, probably as a result of fundamental changes in deep sea circulation.*

kind of event might have been responsible of this dramatic change in the chemistry of the deep sea. These investigations are being made in collaboration with a group of Prof. Münnich at the University of Heidelberg.

## Conclusions and Outlook

These few, arbitrarily selected examples clearly illustrate the potential of the single atom counting method, which from the point of view of applications is only at its very beginning. Besides $^{14}$C and $^{10}$Be, all other radioisotopes listed in Table 1 have been observed at their natural concentration and possible applications in hydrology, meteoritic physics and other fields are presently being investigated.

The activity in this field is best illustrated by the fact that many existing nuclear physics facilities are presently being modified so that programmes on ultrasensitive mass spectrometry can be carried out. New centres with accelerators specially designed for AMS are being installed in several countries, including Australia, Canada, England, France, Japan, New Zealand and the USA. The dedicated facilities of these centres are generally based on a 3 MV tandem accelerator optimized for $^{14}$C dating. The problems of using cyclotrons as precise mass spectrometers seems formidable compared with those of tandem accelerators. However there are some important cases such as $^{41}$Ca or the heavy noble gases, which do not form negative ions readily, where the positive ion cyclotron or heavy ion accelerator approach might be of some advantage.

### REFERENCE

The status of AMS, as well as its applications will be discussed in more detail during the 3rd International Symposium on Accelerator Mass Spectrometry which will take place from 10 - 13 April, 1984, in Zurich.