Accelerator Mass Spectrometry – Big and Small

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Abstract. A brief review of the current status of Accelerator Mass Spectrometry is presented, with emphasis on some of the most recent technical developments.

I. INTRODUCTION

Accelerator Mass Spectrometry (AMS) evolved from nuclear physics laboratories some twenty years ago (1-4), when it was realised that long-lived radionuclides, - in particular ^{14}C - can be measured at natural levels by couting atoms directly. It had been noted earlier (5) that during a typical beta decay measurement of ^{14}C lasting two days, only about one out of a million ^{14}C atoms decays (the half-life of ^{14}C is 5730 years). "Waiting around for the decay of these atoms is clearly an inefficient way to count them" (2). With a $^{14}C/^{12}C$ isotopic ratio of 1.2×10^{-12} in modern carbon, one needs a few grams of carbon to obtain enough decays in two days for a statistical uncertainty of 0.5% (corresponding to an age uncertainty of 40 years). In contrast, with a modern AMS facility one can easily obtain counting rates of 50 ^{14}C ions/sec for one hour using only one milligram of carbon in the ion source. This leads in 15 minutes to a counting statistics of 0.5%. In practical terms, the amount of sample material needed is reduced by at least a factor of 1000 and the measuring time by a factor of 100 as compared to beta counting. Such an enormous gain in detection sensitivity ($\sim10^5$) is similar to the gain in light gathering capability of a very large astronomical telescope as compared to the unequipped eye.

Over the years, AMS has developed into an analytic tool of great versatility, with applications in almost every field of science where the measurement of minute traces of long-lived radioisotope is of interest (6-10). Table 1 gives a summary of fields were AMS measurements are performed. In this table our environment is devided into seven "spheres", each constituting a major domain on Earth and beyond. Measurements of long-lived radionuclides provide important clues for the understanding of chemical and physical processes within each sphere. Even more important, interactions between the spheres in the past and in the presence can also be studied by these AMS measurements. Information gathered in this way will be the basis for extrapolating into our future on Earth, although any of these extrapolations have to be treated with utmost care as to their reliability of firm predictions.

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Table 1. The Seven Spheres of the Environment

Sphere	**Areas of interest where AMS measurements of long-lived radionuclides can be performed; the respective radionuclides used are given in parenthesis.**
Atmosphere	Production and distribution of cosmogenic and anthropogenic radionuclides (^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{26}Al, ^{32}Si, ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{85}Kr, ^{129}I) study of trace gases: $C0_2$, CO, OH, O_3, CH_4 (^{7}Be, ^{10}Be, ^{14}C) transport and origin of aerosols (^{14}C)
Biosphere	dating in archaeology and other fields (^{14}C, ^{41}Ca) ^{14}C calibration studies in tree rings, corals and sediments (^{14}C) in-vivo tracer studies in animals and humans (^{14}C, ^{26}Al, ^{41}Ca, ^{79}Se) studies in forensic medicine through bomb-peak dating (^{14}C)
Hydrosphere	dating of groundwater (^{14}C, ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{129}I) global ocean circulation pattern (^{14}C, ^{39}Ar, ^{129}I) paleoclimatic studies in ocean sediments
Cryosphere	dating of ice cores and paleoclimatic studies in glaciers and polar ice sheets (^{10}Be, ^{14}C, ^{32}Si, ^{36}Cl, ^{39}Ar, ^{81}Kr) variation of cosmic ray intensity with time (^{10}Be, ^{36}Cl) bomb-peak identification (^{36}Cl, ^{41}Ca, ^{129}I)
Lithosphere	exposure dating and erosion studies of surface rocks (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl) paleoclimatic studies in loess (^{10}Be) tectonic plate subduction studies through volcanos (^{10}Be) platinum group elements in minerals (stable trace isotopes)
Cosmosphere ^{41}Ca,	cosmic ray record in meteorites and lunar materials (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{44}Ti, ^{59}Ni, ^{60}Fe, ^{107}Pd, ^{129}I); life on Mars ? (^{14}C) evidence for supernova occurrence through extinct and life radionuclides in meteorites and manganese crusts (^{10}Be, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{60}Fe, ^{107}Pd, ^{146}Sm, ^{244}Pu) geochemical solar neutrino detection (^{98}Tc, ^{205}Pb) search for exotic particles (superheavy elements, fractionally charged particles, strange matter)
Technosphere	releases from nuclear industry (^{14}C, ^{36}Cl, ^{85}Kr, ^{90}Sr, ^{99}Tc, ^{126}Sn, ^{129}I) temperature measurement of fusion plasma (^{26}Al) neutron flux of the Hiroshima bomb (^{36}Cl, ^{41}Ca, ^{63}Ni) characterization of fission material (^{236}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{244}Pu) ultra-high purity tests of semiconductor materials (stable trace isotopes)

II. AMS WITH SMALL MACHINES

Measuring cosmogenic radionuclides at natural levels by mass spectrometry means to be capable of measuring radioisotope-to-stable isotope ratios in the range from 10^{-10} to 10^{-16}. For actual applications these extreme isotope ratios have to be measured with a precision of 0.5% for ^{14}C dating purposes, and to a few percent for other radionuclides. This requires to solve three analytical problems: i) separation of the radionuclide from interfering stable atomic isobars (e.g. from ^{14}N for the detection of ^{14}C), ii) separation from interfering stable molecules (e.g. from ^{13}CH and $^{12}CH_2$ for the detection of ^{14}C), and iii) a reliable measurement of extreme isotopes ratios. As it turns out, tandem accelerators offer by far the best conditions for AMS measurements. In particular, the most important long-lived radionuclide in nature, ^{14}C, can be measured with relative ease at tandem accelerators.

In Figure 1 a modern AMS facility is shown, the Vienna Environmental Research Accelerator (VERA)(11,12), which is based on a 3-MV Pelletron tandem accelerator. Since ^{14}N does not form negative ions (1), the otherwise overwhelming background from ^{14}N (2) is completely absent in tandem-based AMS measurements. However, the negative ion spectrum in Figure 2a measured before the entrance into the tandem accelerator shows a very large background of molecular ions of mass 14, which completely masks the ^{14}C signal. An important step in the consecutive acceleration in the tandem is therefore the stripping process in the terminal, which dissociates the $^{13}CH^-$ and $^{12}CH_2^-$ molecules very effectively when sufficient electrons are stripped off. For twenty years it was believed that $^{14}C^{3+}$ ions (or a higher charge states) must be selected to break up the molecules for sure. With the high energy analysing magnet set to select $^{14}C^{3+}$ ions, one observes the energy spectrum shown in Figure 2b. Although the residual ^{12}C and ^{13}C peaks are greatly reduced in intensity, there is still a large number of background peaks which happen to have the same magnetic rigidity as$^{14}C^{3+}$. However, Figure 2c shows that the $^{14}C^{3+}$ ions can be cleanly selected by sending the mix of ions in figure 2b through a Wien filter (see fig. 1) set to the velocity of $^{14}C^{3+}$. For details of $^{14}C/^{12}C$ ratio measurements at VERA the reader is referred to references (12-14).

It is interesting to note that the "dogma" of stripping to at least the 3+ charge state for obtaining a clean ^{14}C signal was only recently revised, although indications for a deviation were reported much earlier (15). Using a sufficiently thick stripper it is possible to destroy the molecules in the 2+ charge state at about 1 MeV (16-18). Even 1+ stripping looked feasible for obtaining a reasonable ^{14}C separation (16). The latter assumption was recently proven to work very well using a 0.5 MV Pelletron tandem in a collaborative effort of NEC and the AMS laboratory of the ETH/PSI Zurich (19). The most surprising result was the measurement of $^{14}C/^{14}C$ ratios down to a level corresponding to a radiocarbon age of 48,000 years. It therefore looks feasible to perform ^{14}C dating measurements with much smaller tandem accelerators than presently in use, approaching essentially the size of table top machines.

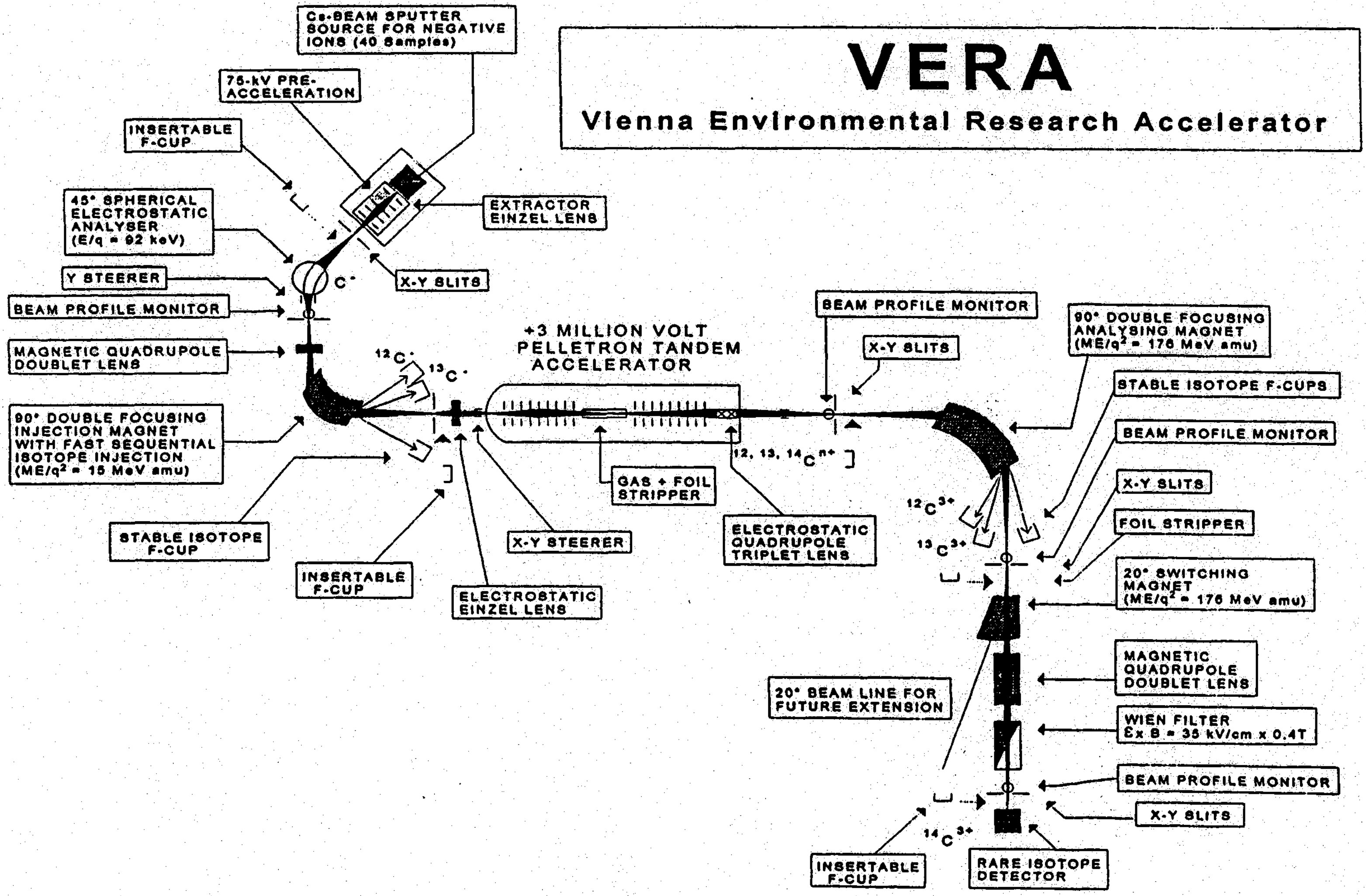

Figure 1. Schematic layout of VERA showing the essential features of the AMS system. ^{14}C measurements are typically performed at a terminal voltage of 2.7 MV.

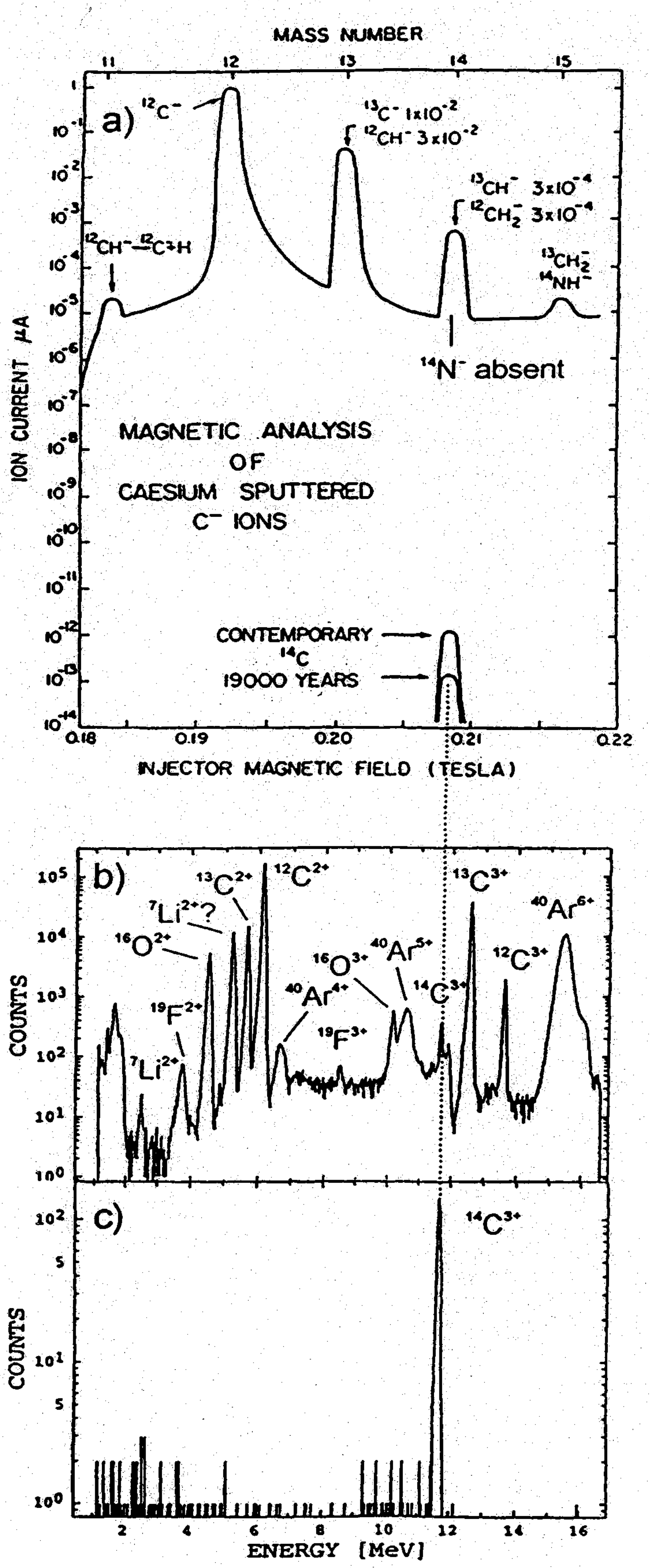

Figure 2. The three steps in the detection of ^{14}C with AMS.

a) In the negative ion mass spectrum after the ion source the ^{14}C signal is buried under an enormous background of mass-14 molecules (15). Most important however, is the absence of $^{14}N^-$ since nitrogen does not form negative ions

b) After acceleration through the tandem and analysis in the high-energy magnet the energy spectrum measured in a Si surface barrier detector still shows many background peaks.

c) After a final analysis through a velocity filter (Wien filter) a clean ^{14}C signal emerges

III. AMS WITH BIG MACHINES

In contrast to the previous section, sometimes very big accelerators are necessary to measure particular radionuclides. This was the case in developing an AMS method for measuring cosmogenic ^{81}Kr ($t_{1/2}$ = 230,000 yr) in the atmosphere (21, 22), and in ground water. Tandem accelerators cannot be used, because Kr does not form negative ions. Therefore, the experiments were performed at a positive ion machine, the K1200 superconducting cyclotron at Michigan State University (22). In order to get rid of the stable isobar ^{81}Br which strongly interfers with ^{81}Kr ($\Delta M/M$ = 3.7x10^{-6}), 17+ ions from the superconducting ECR source were accelerated to an energy of 45 MeV/nucleon (3.65 GeV). At this high energy, 80% of the ^{81}Kr ions can be fully stripped to the 36+ charge state and separated in a magnetic spectrometer from fully stripped ^{81}Br, which can only acquire a maximum charge of 35+. In order to measure small Kr gas samples (~0.4 cm^3 STP), a special gas handling system was developed (21, 22) and a comparison with pre- and post nuclear krypton was performed (Collon 1998). Since no difference between the two Kr sources was found, a first ^{81}Kr dating of groundwater from the Great Artesian Basin in Australia, the largest groundwater system in the world, was attempted. Four samples of 16,000 l of groundwater each were degassed in the field and the extracted gas (320,000 cm^3/sample) were subjected to a rigorous separation procedure at the University of

Table 2. Preliminary results of ^{81}Kr-dating of groundwater from the Great Artesian Basin in Australia

Sample	^{81}Kr/Kr [10^{-13}]	Age [yr]
Atmospheric Krypton	5.20 ± 0.70 [a)]	0
Raspberry Creek	2.63 ± 0.32	225,000 ± 42,000
Duck Hole	2.19 ± 0.28	287,000 ± 38,000
Oodnadatta	1.78 ± 0.26	354,000 ± 50,000
Watson Creek	1.54 ± 0.22	402,000 ± 51,000

[a)] Reference value for natural atmospheric krypton

Bern. This resulted in 0.4 cm^3 Kr /sample containing approximately 3 million ^{81}Kr atoms. Typically, 60 to 100 $^{81}Kr^{36+}$ ions could be counted in the final detection system, resulting in an overall efficiency of ~$2x10^{-5}$ (atoms detected/atoms in the sample). In Table 2,preliminary results for the measured groundwater ages are listed. Although the overall efficiency is a factor of 1000 lower than the typical one achieved for ^{14}C measurements, it was possible to obtain a definite result for very old groundwater samples. Clearly, a substantial improvement in efficiency would be desirable to start "routine" measurements for groundwater samples.

IV. AMS FACILITIES WORLD-WIDE

As mentioned above, AMS originally developed at accelerators in nuclear physics laboratories. A few years after the initiation of AMS, the first generation of small dedicated AMS facilities (Tandetrons) appeared on the market (23). Eventually , a second and third generation of small machines (3 MV terminal voltage) were developed, which became the workhorse for ^{14}C measurements. Recently, several new AMS facilities based on 5-MV Pelletron tandems were established. Parallel to this development a number of larger nuclear physics tandem accelerators were upgraded for AMS measurements. Sometimes, these tandem accelerators were shipped around the world to be assembled as dedicated AMS facilities in a new location: the EN tandem from Oxford went to Peking University, the FN tandem from Rutgers University went to ANSTO in Sydney; the EN tandem from Canberra went to Lower Hutt in New Zealand, and the FN tandem from Washington University went to Livermore. In Table 3, a summary of AMS facilities around the world is given. These 47 facilities measure an estimated total of well over 100,000 samples per year, approximately 90% of it for ^{14}C. Although ^{14}C is by far the most used radionuclide with AMS, many others are gradually increasing in importance (see Table 1). It is foreseeable that eventually all long-lived radionuclides with half-lives longer than approximately 100 years will be subject to AMS measurements.

Comparing big and small AMS facility, there are a few points to be mentioned:

- ^{14}C can be well measured with tandem accelerators at TV = 2 -3 MV. The newest development (19) indicates that it is possible to use also much lower terminal voltages. True radiocarbon dating seems feasible at TV = 0.5 MV.. It may even be possible to measure ^{26}Al ($t_{1/2} = 7.1x10^5$ yr) and ^{129}I ($1.7x10^7$ yr) with these mini-tandems because the respective stable isobars, ^{26}Mg and ^{129}Xe, do not form negative ions. In addition, actinides seem to be another group of radionuclides suited for small tandem accelerators because there are no stable isobars in this mass region (24).

- Isobar separation is the dominant analytic problem in AMS measurements, whenever the stable isobars do form negative ions. Here, higher energy helps greatly, and larger tandem accelerators can more easily perform measurements for interesting radionuclides such as ^{10}Be ($t_{1/2} = 1.5\times10^6$ yr), ^{32}Si (135 yr), ^{36}Cl (3.0×10^5 yr), ^{41}Ca (1.0×10^5 yr), ^{44}Ti (59 yr), ^{53}Mn (3.7×10^6 yr), ^{59}Ni (9.2×10^4 yr), ^{63}Ni (100 yr), ^{60}Fe (1.5×10^6 yr), ^{90}Sr (29 yr), ^{98}Tc (4.2×10^6 yr), ^{126}Sn (2.3×10^5 yr), ^{205}Pb (1.5×10^7 yr), and others.

- Noble gases can only be measured with positive-ion accelerators. As discussed above, a very big machine was necessary to remove the stable isobar ^{81}Br for the ^{81}Kr measurements.

- Finally, a possible solution to the *isobar* separation problem in connection with small accelerators may come from combining the power of elemental separation through laser ion sources (25) with a small accelerator (e.g. a cyclotron) supplying the necessary *isotope* separation.

Table 3. Facilities for Accelerator Mass Spectrometry (1998)

Country	No.	Accelerator	Location
North America			
Canada	1	2.5 MV Tandetron	University of Toronto, Toronto
USA	8	ATLAS Linac	Argonne National Laboratory, Chicago a)
		2.5 MV Tandetron	University of Arizona, Tucson
		9.5 MV FN Tandem	Lawrence Livermore Nat. Lab, Livermore
		K1200 Cyclotron	Michigan State Univ., East Lansing a)
		3 MV Pelletron	Naval Research Lab, Washington D. C. a)
		3 MV Pelletron	University of North Texas, Denton
		9 MV FN Tandem	Purdue University, West Lafayette
		2.5 MV Tandetron	Woodshole Oceanographic Institution
Europe			
Austria	1	3 MV Pelletron	University of Vienna, Vienna
Denmark	1	6 MV EN Tandem	University of Aarhus, Aarhus
England	2	2.5 MV Tandetron	University of Oxford, Oxford
		5 MV Pelletron	University of York, Sand Hutton b),
France	1	2.5 MV Tandetron	Nat. Sci. Research Center, Gif-sur-Yvette,
Germany	4	6 MV EN Tandem	Univ. of Erlangen-Nuernberg, Erlangen
		3 MV Tandetron	University of Kiel, Kiel
		14 MV MP Tandem	Tech. Univ. & Univ. of Munich, Garching
		3 MV Tandem	Forschungszentrum Rossendorf a),
Israel	1	14 MV Pelletron	Weizmann Institute of Science, Rehovot
Italy	1	3 MV Tandem	University of Napels a)
Netherlands	2	3 MV Tandetron	University of Groningen, Groningen
		6 MV EN Tandem	University of Utrecht, Utrecht

Table 3. (continued)

Country	No.	Accelerator	Location
Sweden	2	6 MV EN Tandem	University of Uppsala, Uppsala
		3 MV Pelletron	University of Lund, Lund
Switzerland	2	6 MV EN Tandem	Swiss Federal Inst. of Technology Zürich
		0.5 MV Pelletron	Swiss Federal Inst. of Technology Zürich b),
Asia			
China	4	14 MV MP Tandem	Chinese Inst. of Atomic Energy, Bejing
		6 MV EN Tandem	Peking University, Beijing
		6 MV Tandem	Shanghai Inst. of Nucl. Res., Shanghai
		Mini Cyclotron	Shanghai Inst. of Nucl. Res., Shanghai b)
Japan	9	3 MV Tandetron	Japan Atomic Energy Res. Inst., Mutsu b)
		5 MV Pelletron	Japan Nucl. Cycle Develop. Inst., Toki b)
		8 MV Pelletron	Kyoto University, Kyoto a)
		10 MV Tandem	Kyushu University, Fukuoka a)
		2.5 MV Tandetron	Nagoya University, Nagoya
		3 MV Tandetron	Nagoya University, Nagoya b)
		5 MV Pelletron	University of Tokyo, Tokyo
		5 MV Pelletron	Nat. Inst. for Environ. Studies, Tsukuba
		12 MV Pelletron	University of Tsukuba a)
India	1	3 MV Pelletron	Institute of Physics, Bhubaneswar a),
Korea	1	3 MV Tandetron	Seoul National University, Seoul b),
Australia & New Zealand			
Australia	3	8 MV FN Tandem	Nucl. Sci. and Technol. Organ., Sydney
		2.5 MV Tandetron	Comm. Sci. and Industr. Res. Organ., Sydney
		14 MV Pelletron	Australian National University, Canberra
New Zealand	1	6 MV EN Tandem	Inst. of Geolog.& Nucl. Sci., Lower Hutt
South America			
Argentina	1	20 MV Pelletron	Nat. Atomic. Energy Comm., Buenos Aires a)
Brazil	1	9 MV Pelletron	University of Sao Paulo, Sao Paulo a)

a) AMS development at existing accelerators
b) New dedicated AMS facilities in test operation

V. CONCLUSIONS

Table 3 clearly shows that tandem accelerators dominate the field of AMS. The size of these machines vary from very small (TV = 0.5 MV) to very large (TV = 20 MV). New machines are generally on the small side of the spectrum (TV = 3 to 5 MV). Often, AMS facilities developed around accelerators which simply were available. It is probably fair to say that with enough technical upgrading and modification, almost any accelerator can be used for AMS. This makes AMS a universally available technique.

The breadth of information which can be gathered with AMS in the seven spheres of our environment is enormous. Combined with high-precision stable isotope measurements, this constitutes the "isotope language", which may allow us one day to disentangle even the most complex processes in the environment. Since we can reasonably expect that the power of both AMS and stable isotope MS will increase with time, a bright future for this field lies ahead of us.

REFERENCES

1. Purser, K. H., Liebert, R. B., Litherland, A. E., Beukens, R. P., Gove, H. E., Bennet, C. L., Clover, H. R., and Sondheim, W. E., "An attempt to detect stable N^- ions from a sputter ion source and some implications of the results for the design of tandems for ultrasensitive carbon analysis", *Revue de Physique Appliquée* **12**, 1487-1492 (1977).

2. Muller, R. A., "Radioisotope dating with a cyclotron", *Science* **196**, 489-494 (1977).

3. Nelson, D. E., Korteling, R. G. and Stott, W. R., "Carbon-14: Direct detection at natural concentrations", *Science* **198**, 507-508 (1977).

4. Bennet, C. L., Beukens, R. P., Clover, M. R., Gove, H. E., Liebert, R. B., Litherland, A. E., Purser, K. H.,and Sondheim, W., "Radiocarbon dating using electrostatic accelerators: negative ions provide the key", *Science*, 198, 508-510.

5. Oeschger, H., Houtermans, J., Loosli, H., and Wahlen, M., "The constancy of cosmic radiation from isotope studies in meteorites and on the Earth", in *12th Nobel Symposium on Radiocarbon Variations and Absolute Chronology*, ed. Olssen, I. U., New York: John Wiley & Sons, 1970, pp. 471-498.

6. Litherland, A. E., "Ultrasensitive mass spectrometry with accelerators", *Ann. Rev. Nucl. Part. Sci.* **30**, 437-473 (1980).

7. Elmore, D. and Philiips, F. M., "Accelerator mass spectrometry for measurement of long-lived isotopes", *Science*, 236, 543-550 (1987).

8. Kutschera, W. and Paul, M., "Accelerator mass spectrometry in nuclear physics and astrophysics", *Ann. Rev. Nucl. Part. Sci.*, **40**, 411-438 (1990).

9. Finkel, R. C. and Suter M., "AMS in the earth sciences: techniques and applications", *Advances in Anal. Geochem.* **1**, 1-114 (1993).

10. Tuniz, C., Bird, J. R., Fink, D., and Herzog, G. F., *Accelerator Mass Spectrometry: Ultrasensitive Analysis for Global Science*, Boca Raton: CRC Press, 1998, pp. 1-371.

11. Kutschera, W., Collon, P., Friedmann, H., Golser, R., Hille., P., Priller, A., Rom, W., Steier, P., Tagesen, S., Wallner, A., Wild, E. and Winkler, G., "VERA: A new AMS facility in Vienna", *Nucl. Instr. Meth.* B **123**, 47-50 (1997).

12. Priller, A., Golser, R., Hille, P., Kutschera, W., Rom, W., Steier, P., Wallner, A. and Wild, E., "First performance tests of VERA", *Nucl. Instr. Meth.* B 123, 193-198 (1997).

13. Rom, W., Golser, R., Kutschera, W., Priller, A., Steier, P. and Wild, E., "Systematic investigations of ^{14}C measurements at the Vienna Environmental Research Accelerator", *Radiocarbon*, 40/1, 255-263 (1998).

14. Wild, E., Golser, R., Hille, P., Kutschera, W., Priller A., Puchegger, S., Rom, W. and Steier, P., "First ^{14}C results from archaeological and forensic studies at the Vienna Environmental Research Accelerator", *Radiocarbon*, 40/1, 273-281 (1998).

15. Litherland, A. E., "Accelerator mass spectrometry", *Nucl. Instr. Meth.* B5, 100-108 (1984).

16. Suter, M., Jacob, St. and Synal, H. A., "AMS of ^{14}C at low energies", *Nucl. Instr. Meth.* B 123, 148-152 (1997).

17. Mous, D. J. W., Purser, K. H., Fokker, W., van den Broek, R. and Koopmans, R. B., "A compact ^{14}C isotope ratio mass spectrometer for biomedical applications", *Nucl. Instr. Meth.* B 123, 153-158 (1997).

18. Mous, D. J. W., Fokker, W., van den Broek, R. and Koopmans, R. B., "An ion source for the HVEE ^{14}C isotope ratio mass spectrometer for biomedical applications", *Radiocarbon* 40/1, 283-288 (1998).

19. Suter, M., Huber, R., Jacob, S. A. W., Synal, H. A. and Schroeder, J. B., "A new small accelerator for radiocarbon dating ", to be published in the Proceedings of the Denton Conference 1998.

20. Kutschera, W., Paul, M., Ahmad, I., Antaya, T. A., Billquist, P. J., Glagola, B. G., Harkewicz, R., Hellstrom, M., Morrissey, D. J., Pardo, R. C., Rehm, K. E., Sherrill, B. M., and Steiner, M., "Long-lived noble gas radionuclides", *Nucl. Instr. Meth.* B 92, 241-248 (1994)

21. Collon, P., Cole, D., Davids, B., Fauerbach, M., Harkewicz, R., Kutschera, W., Morrissey, D. J., Pardo, R., Paul, M., Sherrill, B. M. and Steiner, M., "Measurement of the long-lived radionuclide 81Kr in pre-nulcear and present-day atmospheric krypton", to be published in Radiochimica Acta (1999).

22. Collon, P., Antaya, T. A., Davids, B., Fauerbach, M., Harkewicz, R., Hellstrom, M., Kutschera, W., Morrissey, D. J., Pardo, R. C., Paul, M., Sherrill, B. M., and Steiner, M., "Measurement of ^{81}Kr in the atmosphere ", *Nucl. Instr. Meth.* B 123, 122-127 (1997)

23. Purser, K. H., Schneider, R. J., Dobbs, J. McG. and Post, R., "A preliminary description of a dedicated commercial ultra-sensitive mass spectrometer for direct atom counting of ^{14}C", *Proc. Sypmosium on* National Laboratory Report ANL/PHY-81-1, 1981, pp. 431-462.

24. Zhao, X.-L., Nadeau, M.-J., Kilius, L. R. and A. E. Litherland, "The first detection of naturally-occurring ^{236}U with accelerator mass spectrometry", *Nucl. Instr. Meth.* B 92, 249-253 (1994).

25. Van Duppen, P., "Laser ion sources for on-line isotope separators ", *Nucl. Instr. Meth.* B 126, 66-72 (1997).