

## NEW AND IMPROVED AMS FACILITIES

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### Abstract

Accelerator Mass Spectrometry (AMS) provides instrumentation originally developed by nuclear physicists more than 30 years ago to measure long lived cosmogenic radionuclides such as  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{129}\text{I}$ , U, Pu and Pa at natural levels. In the past ten years impressive progress in the measurement technique has been made and with the appearance of compact low energy radiocarbon AMS systems, a new category of AMS instruments has been introduced [1,2,3]. This has resulted in a boom of new AMS facilities with more than 20 new installations over the last five years. But low energy AMS is not limited to radiocarbon only and there is a great potential for  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and actinides measurements at compact AMS systems. The latest developments towards the low energy limit of AMS resulted in two new types of systems, the NEC [4] Single Stage AMS (SSAMS) and ETH mini carbon dating system (MICADAS) operating with terminal voltages of about 200 kV only. In addition, systems like the HVEE [5] 1 MV Tandetron or the compact ETH 600 kV system are capable to extend the range of applications at compact systems beyond radiocarbon. These systems will have enormous impact, not only on the use of AMS in biomedical research and on radiocarbon dating but also for research applications with  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{129}\text{I}$  and actinides.

### THE EARLY DAYS OF AMS

The fundamental principles of AMS as they were developed in 1977 in connection with tandem accelerators [6, 7] are still the basis of state-of-the-art AMS instrumentation. In the early days, three different concepts had been applied: first, a dedicated spectrometer generation based on 2.5 MV Tandetron accelerators from General Ionex Corporation was introduced by Kenneth Purser (Arizona, Oxford, Gif-sur-Yvette); second, transformation of medium sized tandem accelerators such as High Voltage EN type into dedicated AMS spectrometers (e.g. Zurich, Utrecht, Uppsala, Erlangen) and third, the gentle modification of large sized tandem facilities to accommodate the needs of AMS experiments (e.g. Rochester, Munich, Canberra). Triggered by the need of highly precise radiocarbon dates and a tremendous measurement capacity in connection with the World Ocean Circulation Experiment WOCE in the early 1990's an AMS-system based on a 3 MV Tandetron accelerator was proposed by Kenneth Purser and installed in 1991 at the Woods Hole Oceanographic Institution [8]. Followed by similar systems in Groningen, Kiel, Nagoya, JEARI, Seoul, Jena and Oxford, this kind of instrumentation became the benchmark for high precision radiocarbon analysis over many years.

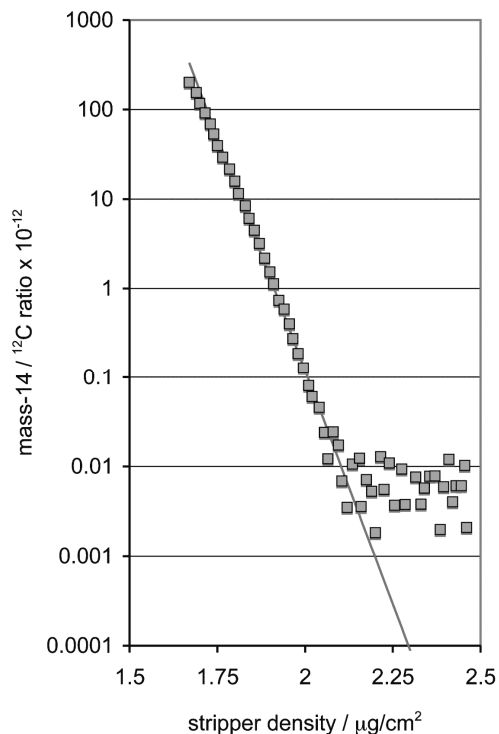


Figure 1: Intensity of molecules of mass 14 amu. At sufficient density the interference can be eliminated and radiocarbon dating measurements become possible.

### COMPACT $^{14}\text{C}$ AMS INSTRUMENTS

To measure long-lived radionuclides at natural concentrations mass interferences have to be filtered out. In several cases isobaric nuclides can be suppressed already in the ion source and only remaining molecular ambiguities have to be removed. These interferences are eliminated exploiting the stripping process inside the tandem accelerator. There are no bound states of small molecules with charge state 3+ and higher, and thus, such molecules simply decompose as soon as they lose their binding electrons in collisions with gas atoms or molecules. To reach sufficient charge state yields, the ions require relatively high energies, e.g. the maximum yield for 3+ and 4+ is reached for  $^{14}\text{C}$  ion at 2.5 MeV and 6.5 MeV respectively in Ar stripper gas. Consequently, quite large accelerator systems are required to provide ions in the necessary energy range.

However, there is a certain probability of molecular ions to decompose in collisions process with stripper gas even if there are bound states. Consequently, the intensity of the molecular component of an ion beam will decrease exponentially with increasing area density of the stripping medium and the interfering molecular component of the ion beam can be reduced down to a level where isotope

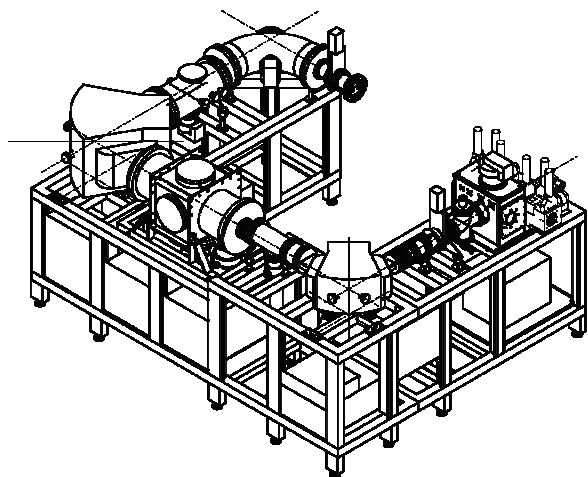


Figure 2: Design view of the next generation AMS spectrometer for radiocarbon dating and for biomedical research. The overall dimensions of the are 2.5 m x 3 m.

ratio measurements at natural concentrations become possible. This new way to eliminate molecular interferences has the great advantage that the maximal yield for  $^{14}\text{C}$  in charge exchange to 1+ can be reached at energies of about 500 keV. Consequently, AMS systems based on this principle are more compact with smaller accelerators and can be produced and operated at much lower cost than traditional systems. The development of compact AMS systems for radiocarbon was undertaken at ETH in collaboration NEC. NEC has used these developments to commercialize AMS systems based on the new technology. The obvious advantages of the compact systems have also been realized by HVEE which now has proposed its own compact AMS system based on a 1 MV Tandatron accelerator [9]. The first instrument of this type was installed at University of Seville in Spain in 2005 [10]. In contrast to the compact NEC systems, which are optimized for radiocarbon, the compact HVEE system has the option to analyze beams up to plutonium.

### Next Generation of Compact Systems

Shortly after proof of principle of the new method to eliminate molecular interferences, the question of the low energy limit of AMS arose. Detailed measurements of molecule dissociation cross sections revealed that even at energies as low as 200 keV, the cross sections are sufficiently high to achieve suppression of molecular interference down to levels where dating measurements can be performed. Stripping yields for the 1+ charge state are still approximately 50% and thus high enough for efficient radiocarbon detection. Traditional accelerator systems are not necessary anymore to obtain ion energies in these ranges. At ETH we have designed a vacuum insulated high-voltage platform in a tandem configuration fed from a commercial high voltage power supply and hosting a charge exchange channel that provides sufficient area density of a stripper gas to eliminate molecular interferences. Based on initial tests of this

device, a prototype dedicated radiocarbon dating system (MICADAS, Fig. 2) was built and became operational in 2004 [11]. The MICADAS type system has a footprint of  $2.5 \times 3 \text{ m}^2$  only and is therefore the smallest AMS instrument capable of high performance radiocarbon measurements.

### Frontiers of Radiocarbon Dating

Measurements at the 1‰ level require counting statistics beyond 1 million  $^{14}\text{C}$  events per sample. Even with modern high current ion sources this requires long measurement times. In general, the measurement system must provide stable and reproducible measurement conditions over much longer times because appropriate normalization procedures and assessment of machine internal variability should be evaluated in parallel with the high precision measurements of samples.

The ETH MICADAS system is based in its design more on a conventional mass spectrometer than a traditional AMS system. This should be a good qualification to achieve highly precise  $^{14}\text{C}/^{12}\text{C}$  measurements. At the MICADAS system, we have measured homogenized material of oxalic acid I standard material. A set of four standard cathodes was analyzed together with blank samples in a 24 h measurement, where for each standard cathode the total measurement time was 2.5 h resulting in 600'000 detected  $^{14}\text{C}$  events equivalent to 1.3‰ statistical precision. The  $^{14}\text{C}/^{12}\text{C}$  ratios of the 120 runs are shown in Fig. 3. Note that none of the total 1200 data blocks taken during the 24 hours of measurement have been discarded. The scatter of the individual measurements is consistent with their internal uncertainty based on counting statistics. The relative standard uncertainty of each of the four standards is between 1.12‰ and 1.35‰ and is reproduced at sub-per mil level. This shows that MICADAS-type systems, in general, are capable to provide measurement conditions necessary for ultra-high precision measurements.

### AMS system for Biomedical Research

Despite the great potential to use AMS in connection with biomedical research a widespread biomedical use of AMS had been impeded by the complexity of the measurement technology and the related high cost of the

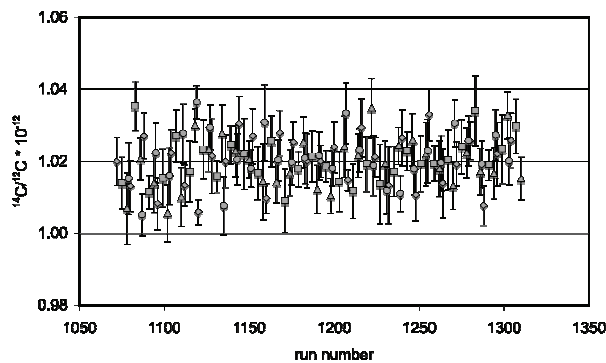


Figure 3:  $^{14}\text{C}/^{12}\text{C}$  measurements of different targets over 24h of operation.

analyses. Biomedical applications have, compared to radiocarbon dating, different demands. Analytic accuracy of typically 5% is sufficient, a much wider dynamic range (1000 Modern to a few % Modern) must be covered and a high sample throughput is required for the efficient use of the installations and for a significant reduction in effective cost for the analyses. In order to provide the advantages of latest type of AMS systems for biomedical research, we have developed at ETH a dedicated biomedical system based on the MICADAS concept (BioMICADAS) in collaboration with Vitalea Science, a commercial service provider for biomedical AMS services. In particular, the sample handling and the capability to digest huge counting rates of the rare isotope have been improved. The present system has the capability to analyze more than 20000 samples per year under conditions, as they are required in biomedical research projects. In parallel, NEC has invented a single stage AMS system that can fit the needs of biomedical research as well. Thus, the latest developments in AMS with advent of compact and more recently of SSAMS as well as the dedicated BioMICADAS systems substantially pushed forward the use of AMS in biomedical research. This should be kept in mind in connection with changes in government regulations regarding the concept of micro-dosing where the new systems enable novel and very attractive applications with pharmaceutical drug developments.

### MULTI-PURPOSE AMS SYSTEMS

There has always been the question of which type of accelerator would be optimal for dedicated AMS systems. A pure radiocarbon dating system has different requirements than a multi-purpose AMS system covering a range of AMS nuclides for which substantial isobar suppression capabilities are needed. FN accelerators reaching terminal voltages of 10 MV seemed to be well suited as basis of versatile multi-purpose AMS systems. At Lawrence Livermore National Laboratory, at Purdue University and at the Australian Nuclear Science and Technology Organisation AMS facilities were built around FN tandems. Large Pelletron and MP tandem accelerators with terminal voltages ranging from 14 to 25 MV (Canberra, Munich, Oakridge) are still used for AMS measurements, predominantly for heavier nuclides which suffer from abundant nuclear isobar interferences. In general, the effort to operate and maintain such large accelerator facilities is too high to justify it with an AMS program.

During the early times of AMS the Toronto group has pioneered the analysis of heavy nuclides at fairly small Tandatron based AMS systems and the AMS system in Vienna has demonstrated that a versatile multi-purpose AMS instrument can be built around a 3 MV accelerator. But more recently, AMS systems based on larger accelerators show quite a renaissance. A number of new facilities have been installed recently or are under construction/commissioning. With these systems measurements of  $^{36}\text{Cl}$  are possible because at higher beam

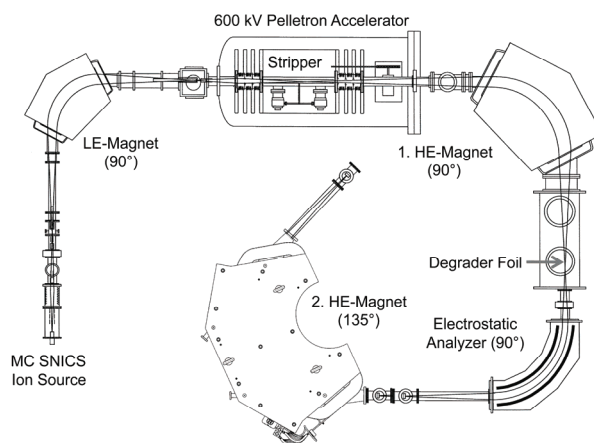


Figure 4: Extension of the high-energy spectrometer of the ETH 600 kV AMS system. By adding a second magnetic spectrometer to the beam line interferences from scattered ions causing background events can be reduced significantly.

energies the interfering isobar  $^{36}\text{S}$  can be suppressed sufficiently using  $dE/dx$  energy loss difference of ions with different nuclear charge.

### Possibilities of Compact Multipurpose AMS Systems

Compact AMS facilities are not limited to radiocarbon only. In particular,  $^{10}\text{Be}$  can be sufficiently separated from its isobaric counterpart  $^{10}\text{B}$  and identified at terminal voltages well below 1 MV [12, 13]. This has become possible because of significant improvements in the detection technique of low energetic ions with gas ionization detectors [14] due to the use of i) the now available very thin and homogenous SiN detector windows [15] and ii) due to the improved detector resolution of the detector electronics by using CoolFET preamplifiers [16] reducing the contribution of the electronic noise to the resolution of the detector signals. Applying the degrader foil technique [12, 17, 18] at beam energies as low as 700 keV and eliminating background from scattered molecular fragments yields competing performance with larger AMS systems. Detection limits for  $^{10}\text{Be}/^9\text{Be}$  ratios in the range of a few times  $10^{-15}$  and  $^{10}\text{Be}$  transmission efficiencies of about 8 % have been demonstrated [19]. The total potential for  $^{26}\text{Al}$  at compact AMS facilities has not yet been sufficiently explored. Performance measurements at the Zurich compact AMS system using 1+ have shown 25% overall beam transmissions and background levels of the order of  $10^{-14}$  [20]. Further improvements seem possible with the upgrade of the high-energy mass spectrometer (Fig. 4) and the expected higher suppression of scattered molecular fragments.

$^{41}\text{Ca}$  measurements are interfered by the atomic isobar  $^{41}\text{K}$  and are therefore predominantly performed at larger AMS systems e.g. PRIME Lab [21], CAMS/LLNL [22]. However, the performance achievable at compact systems has been demonstrated to be sufficient for biomedical

studies [23] and due to the reduced analysis cost at smaller AMS systems, these systems can be regarded as a viable alternative for those applications. With systems having about 5 MV terminal voltage, a much better sensitivity of a few times  $10^{-14}$  can be reached [23] due to the quite impressive potassium suppression of optimized gas ionization detectors. A similar performance has been reached at the VERA system with 3 MV terminal voltage for  $\text{CaF}_2$  samples and of  $10^{-15}$  and below using  $\text{CaH}_2$  [24].

Actinides like U, Pu and Pa are not interfered by stable atomic isobars and the atomic isobar of  $^{129}\text{I}$  does not form negative ions. Therefore, these nuclides can be measured with low energy AMS systems, if molecular isobars can be eliminated. In the case of  $^{129}\text{I}$ , Zhao et al. [25] have shown that using 3+ with stripping efficiency of 10% at 1 MeV  $^{129}\text{I}/\text{I}$  ratios as low as  $10^{-14}$  can be measured. This is sufficient for almost all applications of  $^{129}\text{I}$ . At the Zurich compact AMS system,  $^{129}\text{I}$  measurements have been made using the 4+ charge state [26]. More recently, the performance of these measurements could be increased using 3+ at 500 kV terminal voltage [27]. Because charge state 3+ is used, the stripper density does not have to be increased beyond the equilibrium region, but molecular fragments in charge state 2+ with the same m/q ratio as  $^{129}\text{I}$  can survive the stripping process and can under unfavorable circumstances limit proper identification of  $^{129}\text{I}$  ions. Apart from these minor obstacles, the performance of compact AMS for  $^{129}\text{I}$  will fit the needs of the applications and these systems can be regarded as true alternatives to systems with larger accelerators.

Ultra-sensitive detection of plutonium isotopes have been demonstrated at an accelerating voltage of 300 kV at the Zurich compact AMS system [28]. The observed transmission efficiencies were surprisingly high (15% for Th) and detection levels competitive to larger systems have been demonstrated. Detection limit for  $^{236}\text{U}/\text{U}$  of  $10^{-9}$  have been shown [29] and further progress can be expected adopting better high-energy spectrometers with improved suppression capabilities against scattered molecular fragments. AMS of  $^{231}\text{Pa}$  measurements became possible with a performance comparable to modern mass spectrometers [30] and thus a new radionuclide was introduced into the AMS world of compact AMS systems. The HVEE 1 MV compact AMS system in Seville has also demonstrated impressive capabilities of actinide measurements [31]. As pointed out by Fifield [32], the much better transmission efficiency at low energies make compact systems very attractive for actinide measurements. Further improvements of the mass spectrometer at the high-energy end of these systems, in particular an improved rejection of scattered molecular fragments will make future compact systems even better suited for actinide AMS.

## CONCLUSIONS

AMS has evolved to a powerful analytic technique. Worldwide more than 80 AMS facilities contribute to a wide variety of research fields. Whereas during the early

days of AMS research laboratories optimized their accelerator systems and advanced the technology, today, the two competing companies NEC and HVEE provide a wide variety of AMS instruments with acceleration stages from 0.2 MV to 6 MV terminal voltage. Practically, all the new installations of AMS facilities over the last five years were equipped with instrumentation already available or especially tailored to the intended application program by these two companies. Despite the fact that the collaborative efforts between commercial AMS providers and various research groups has fruitfully improved the available AMS instrumentation, the ultimate goal of having available AMS instruments as compact and as easy to operate and maintain as conventional mass spectrometers has not yet been reached. Thus, there is still a strong need for independent technological and methodical developments at universities and research institutions to advance AMS instrumentation to meet future requirements and to promote a more widespread use of AMS. The availability of small and compact AMS systems has already launched a boom of new laboratories with AMS capabilities. During the past 5 years more than 20 new facilities became operational using accelerators systems with terminal voltages of 1 MV or less. Low energy systems do have the capability to provide competitive performance not only for radiocarbon, but also for other important AMS nuclides such as  $^{10}\text{Be}$ ,  $^{129}\text{I}$  and actinides, although this potential has not been fully exploited yet. In addition, concentrated efforts should be made to sustain the capabilities of AMS of more exotic nuclides requiring large accelerator system and high beam energies.

## REFERENCES

- [1] M. Suter, S. Jacob, H.A. Synal, Nucl. Instr. and Meth. B 123 (1997) 148.
- [2] H.A. Synal, S. Jacob, M. Suter, Nucl. Instr. and Meth. 161-163 (2000) 29.
- [3] H.A. Synal, S. Jacob, M. Suter Nucl. Instr. and Meth. B 172 (2000) 1.
- [4] National Electrostatics Corporation, Middleton, WI, USA.
- [5] High Voltage Engineering Europa B.V., Amersfoort, The Netherlands.
- [6] C.L. Bennett, R.P. Beukens, M.R. Clover, H.E. Gove, R.B. Liebert, A.E. Litherland, K.H. Purser, W.E. Sondheim, Science 198 (1977) 508.
- [7] D.E. Nelson, R.G. Korteling, W.R. Stott, Science 198 (1977) 507.
- [8] K.H. Purser, T.H. Smick, R.K. Purser, Nucl. Instr. and Meth. B 52 (1990) 263.
- [9] M.G. Klein, D.J.W. Mous, A. Gott dang, Nucl. Instr. and Meth. B 249 (2006) 764.
- [10] E. Chamizo, J.M. Lopez-Gutierrez, A. Ruiz-Gomez, F.J. Santos, M. Garcia-Leon, C. Maden, V. Alifimov, Nucl. Instr. and Meth. B 266 (2008) 2217.
- [11] H.-A. Synal, M. Stocker, M. Suter, Nucl. Instr. and Meth. B 259 (2007) 7.

- [12] M. Grajcar, M. Döbeli, P.W. Kubik, H.-A. Synal, L. Wacker, M. Suter, Nucl. Instr. and Meth. B 259 (2007) 173.
- [13] A.M. Müller, M. Christl, M. Döbeli, P.W. Kubik, M. Suter, H.-A. Synal, Nucl. Instr. and Meth. B Proceedings of the AMS-11 Conference (2009).
- [14] M. Suter, M. Döbeli, M. Grajcar, A. Müller, M. Stocker, G. Sun, H.-A. Synal, L. Wacker, Nucl. Instr. and Meth. B 259 (2007) 165
- [15] M. Döbeli, C. Kottler, M. Stocker, S. Weinmann, H.-A. Synal, M. Grajcar, M. Suter, Nucl. Instr. and Meth. B 219-220 (2004) 415.
- [16] Amptek, Inc., D.A. Drive, M.U.S.A. Bedford.
- [17] G.M. Raisbeck, F. Yiou, D. Bourles, J. Lestringuez, D. Deboffe, Nucl. Instr. and Meth. B 5 (1984) 175.
- [18] W. Kutschera, W. Henning, M. Paul, R.K. Smither, E.J. Stephenson, J.L. Yntema, D.E. Alburger, J.B. Cumming, G. Harbottle, Physical Review Letters 45 (1980) 592.
- [19] A.M. Müller, M. Christl, M. Döbeli, P.W. Kubik, M. Suter, H.A. Synal, Nucl. Instr. and Meth. B 266 (2008) 2207.
- [20] M. Stocker, M. Döbeli, M. Grajcar, M. Suter, H.-A. Synal, L. Wacker, Nucl. Instr. and Meth. B 240 (2005) 483.
- [21] G.S. Jackson, D. Elmore, F.A. Rickey, S.M. Musameh, P. Sharma, D. Hillemonds, L. Coury, P. Kissinger, Nucl. Instr. and Meth. B 172 (2000) 899.
- [22] M.L. Roberts, et al., Nucl. Instr. and Meth. B 123 (1997) 57.
- [23] T. Schulze-König, C. Maden, E. Denk, S.P.H.T. Freeman, M. Stocker, M. Suter, H.-A. Synal, T. Walczyk, Nucl. Instr. and Meth. B Proceedings of the AMS-11 Conference (2009).
- [24] A. Wallner, O. Forstner, R. Golser, G. Korschinek, W. Kutschera, A. Priller, P. Steier, C. Vockenhuber, Nucl. Instr. and Meth. B Proceedings of the AMS-11 Conference (2009).
- [25] X.L. Zhao, A.E. Litherland, W.E. Kieser, C. Soto, Nucl. Instr. and Meth. B 259 (2007) 265.
- [26] M. Stocker, M. Döbeli, M. Grajcar, M. Suter, H.-A. Synal, L. Wacker, Nucl. Instr. and Meth. B 240 (2005) 483.
- [27] V. Alfimov, H.A. Synal, Proceedings of the AMS-11 Conference (2009).
- [28] L.K. Fifield, H.A. Synal, M. Suter, Nucl. Instr. and Meth. B 223-224 (2004) 802.
- [29] L. Wacker, E. Chamizo, L.K. Fifield, M. Stocker, M. Suter, H.A. Synal, Nucl. Instr. and Meth. B 240 (2005) 452.
- [30] M. Christl, L. Wacker, J. Lippold, H.-A. Synal, M. Suter, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 262 (2007) 379.
- [31] E. Chamizo, S.M. Enamorado, M. García-León, M. Suter, L. Wacker, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 266 (2008) 4948.
- [32] L.K. Fifield, P.P. Pavel, Radioactivity in the Environment, Elsevier, 2008, pp. 263.