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Accelerator mass spectrometry – from DNA to astrophysics

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Abstract. A brief review of accelerator mass spectrometry (AMS) is presented. The present work touches on a few technical aspects and recent developments of AMS, and describes two specific applications of AMS, the dating of human DNA with the ^{14}C bomb peak and the search for superheavy elements in nature. Since two extended general reviews on technical developments in AMS [1] and applications of AMS [2] will appear in 2013, frequent reference to these reviews is made.

1 Introduction

Direct atom counting with Accelerator Mass Spectrometry (AMS) revolutionized the utilization of long-lived radionuclides, which previously had to be detected through their feeble radioactive decay. This brought about a reduction in sample size by factors of thousand to a million (gram to milligram to microgram), opening up many new applications.

With AMS one usually measures atom ratios of the rare radionuclide to a stable nuclide in the range from 10^{-12} to 10^{-16} with mass spectrometric techniques including an accelerator. It turned out that tandem accelerators have many advantages for such measurements, and consequently almost all AMS facilities around the world (~100) are based on this type of accelerator (Fig. 1, [1]). Although ^{14}C comprise about 90% of all AMS measurements, more than 50 different radionuclides across the nuclear chart – both natural and man-made - are by now being utilized by AMS. This led to a wide variety of AMS applications in almost every domain of our environment at large [2].

In the current work a brief general description of AMS will be given, covering both recent technical developments and applications in the many fields AMS has made an impact. Two special applications will be treated in more detail. The use of the ^{14}C bomb peak to determine the age of human cells by dating DNA extracted from them, and the search for superheavy nuclides in nature.

2 Technical Advances of AMS

In order to measure the ultra-low radionuclide abundances mentioned above, AMS has to efficiently remove interferences from both molecular and atomic isobars of stable nuclides. Without accelerators, this is sometimes impossible and hence these isobars limit standard mass spectrometric methods.

The use of tandem accelerators requires the use of negative ions from the ion source, which has a distinct advantage for the detection of some radionuclides because the corresponding stable atomic isobars (in brackets) cannot form negative ions: ^{3}H (^{3}He), ^{14}C (^{14}N), ^{26}Al (^{26}Mg), ^{36}Cl (^{36}Ar), ^{55}Fe (^{55}Mn), ^{68}Ge (^{68}Zn), ^{129}I (^{129}Xe), ^{202}Pb (^{202}Hg). However molecular isobars with stable nuclides usually form negative ions, e.g. $^{12}CH_2^-$ and $^{13}CH^-$, and therefore must be removed by other means. For AMS with tandem accelerators this is usually accomplished by the stripping process in the terminal. On the other hand, some radionuclides form negative molecular ions while the corresponding molecule with the stable isobar (in brackets) does not, e.g. $^{41}CaH_3$($^{41}KH_3$). The more difficult cases for AMS are radionuclides where negative ions exist for stable atomic and molecular isobars (in brackets), e.g. ^{10}BeO (^{10}BO), ^{36}Cl (^{36}S), ^{53}MnO (^{53}CrO), ^{60}Fe (^{60}Ni). When the radionuclide of interest does not form negative atomic or molecular ions, e.g. ^{39}Ar and ^{81}Kr, positive-ion accelerators have to be used, and strong interference from the stable isobars ^{39}K and ^{81}Br, respectively, have to be dealt with. If the isobar interference cannot be removed by selective ion formation or filtering, the final separation has to happen after the accelerator. For this, higher energy is always an advantage and thus favors larger tandem accelerators.

2.1 Small and large AMS facilities

The most significant technical development of AMS in recent years is the trend to ever smaller AMS facilities, particularly for ^{14}C measurements. This is depicted in Figure 1 reproduced from the recent review by Synal [1]. Together with the reduction of carbon sample sizes from

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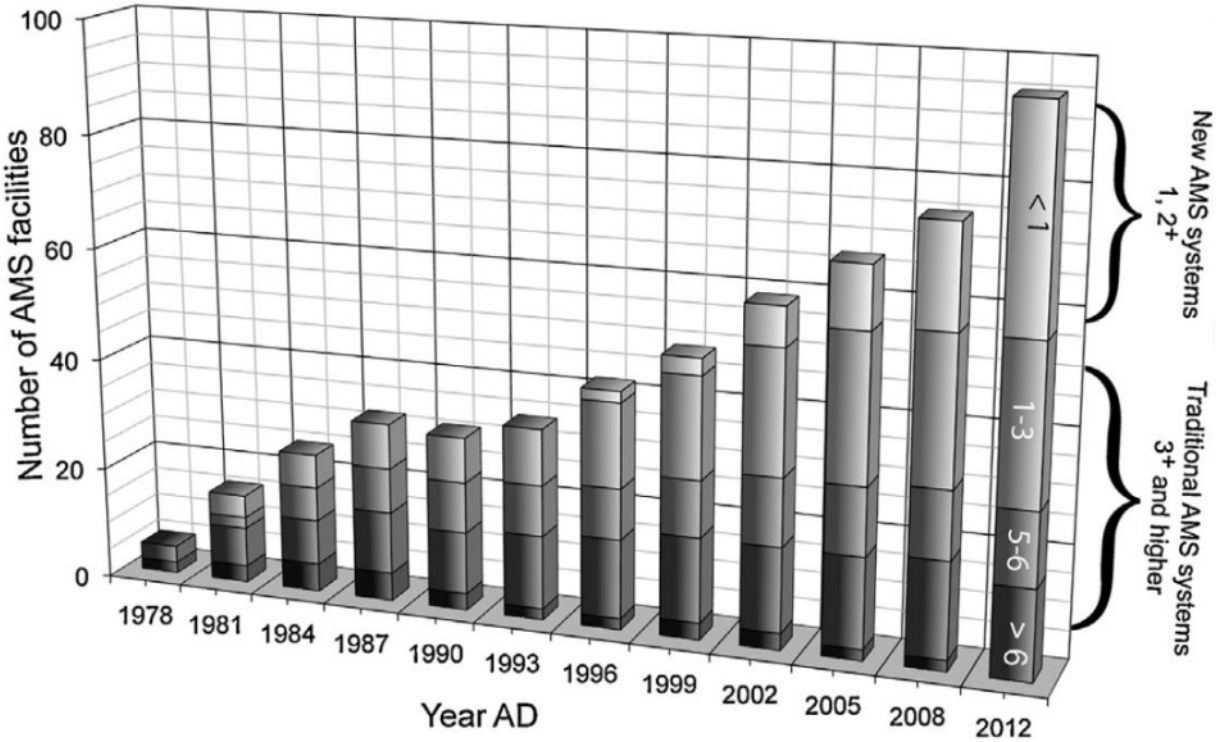

Figure 1. The growth of the number of tandem AMS facilities since 1978, as shown in Figure 4 of Ref. [1]. The different grey shades of the column sections indicate the tandem terminal voltages in MV (see last column in 2012). Cyclotrons (bottom sections) were only in use until 2008. The increase in the number of small tandem AMS facilities in the later years is clearly visible.

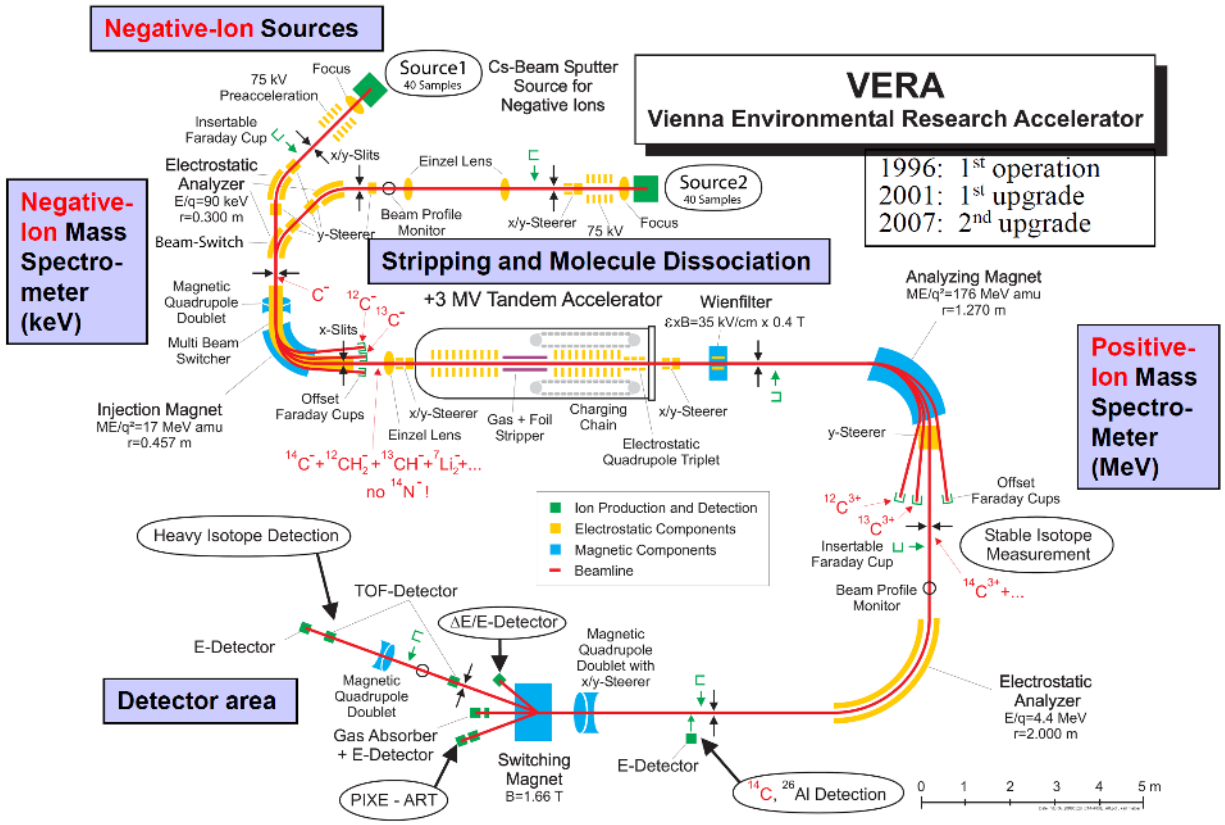

Figure 2. Layout of the Vienna Environmental Research Accelerator (VERA). This dedicated AMS facility if based on a 3-MV Pelletron tandem accelerator. The detection of ^{14}C is schematically indicated. Since its first operation in 1996, the facility was upgraded twice to allow AMS experiments for 'all' isotopes. A third upgrade is planned with a laser – negative ion interaction at the injection side, and an additional 90-degree analyzing magnet in the detector area.

milligram to microgram (e.g. [3]), this may be called the second revolution of radiocarbon dating (the first was atom counting versus decay counting). Small AMS facilities such as the mini radiocarbon dating system MICADAS in Zurich [4] occupy a floor space of less than 10 m^2, which is comparable to the space requirement for a standard mass spectrometer. By contrast, the largest AMS facilities such as the one based on a 14-MV tandem accelerator in Munich occupy a hundred times larger floor space [5]. However, the much higher energies available at this facility allows one to perform isobar separation for AMS experiments not possible at the smaller machines. Examples are ^{53}Mn [6], ^{60}Fe [7], and ^{63}Ni [8]. Somewhat in between the small and large machines is the Vienna Environmental Research Accelerator (VERA), a dedicated AMS facility based on a 3-MV tandem accelerator [9]. A schematic of the upgraded VERA facility is shown in Figure 2. Although a considerably larger floor space than MICADAS is needed, VERA has the advantage to allow one to perform AMS experiments for the entire mass range of nuclides [10], from hydrogen to superheavy elements. It should be noted, though, that in the mass region of actinides there exist no stable atomic isobars and therefore small AMS facilities can also be used for their detection [11]. However, molecular isobar interferences limit the sensitivity to some extent [12].

2.2 New developments of isobar suppression

Two different approaches to suppress stable isobars of interfering negative ions at low energy are under development [13, 14]. When the interfering atomic or molecular anions have an electron affinity lower than the negative ion of the radionuclide of interest, it is possible to selectively neutralize the former by photodetachment with a laser, and therefore remove it from the negative ion beam [15]. In order to do this efficiently, the ions extracted from a Cs-beam sputter source with typically 20 keV have to be slowed down to thermal energies in a linear quadrupole guide by buffer-gas-cooling [16].
Currently, such a system is in the test phase at the VERA lab, and will soon be put online with the AMS facility. This should allow one to utilize also "difficult" radionuclides at the relatively low energies of the VERA facility.

The other method was pioneered at the IsoTrace laboratory in Toronto [14], and uses isobar specific ionic reactions in a gas-filled radiofrequeny quadrupole (RFQ) reaction cell at eV energies. This method looks promising to perform selective on-line ion chemistry with large suppression of unwanted stable isobars [17]. It will eventually also allow one to use small AMS facility where the isobar interferences must be removed at the injection side of the tandem accelerator.

3 Applications of AMS

The broad range of applications of AMS have recently been reviewed for the special volume "100 Years of Mass Spectrometry" [2]. Here we reproduce the table from this review which allows one to get an impression of the multitude of application in the seven domains of our environment at large. The table clearly indicates the importance of ^{14}C measurements in almost every field of application. A detailed discussion of the various applications indicated in the table can be found in ref. [2]. Here only two recent applications of AMS in different fields (biomedicine and astrophysics) will be discussed: ^{14}C bomb peak dating of human DNA and the search for superheavy elements in nature, respectively.

Table 1. Overview of AMS applications

DOMAIN/RESEARCH AREA	RADIONUCLIDE[a]
Atmosphere	
Production of radionuclides by cosmic-ray interaction	^{10}Be, ^{14}C, ^{26}Al, ^{32}Si ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{129}I
Chemistry and dynamics of CO, CO_2, CH_4	^{14}C, ^{14}C
Mixing of stratospheric and tropospheric air	^{14}C, ^{10}Be
Releases from nuclear industry	^{14}C, ^{99}Tc ^{129}I
Fossil fuel effect, 'dead' CO_2	^{14}C
Bomb peak from nuclear weapons testing	^{14}C
Biosphere	
^{14}C dating in archaeology and other fields	^{14}C
^{14}C calibration (tree rings, corals, sediments)	^{14}C
Development of radiocalcium dating of bone	^{41}Ca
Bomb-peak dating (forensic medicine, DNA)	^{14}C
Microdosing for drug development	^{14}C
In vivo studies in plants, animals, humans	^{14}C, ^{26}Al, ^{41}Ca
Hydrosphere	
Dating of groundwater	^{14}C, [^{39}Ar], ^{36}Cl, ^{81}Kr, ^{129}I
Global ocean currents	^{14}C, ^{14}C, ^{39}Ar, ^{99}Tc, ^{129}I, ^{231}Pa, ^{236}U
Paleoclimatic studies in lake and ocean sed.	^{14}C
Cryosphere	
Paleoclimate studies in polar ice	^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, [^{81}Kr]
Paleoclimate studies in glaciers	^{14}C, ^{32}Si
Tracing solar variability (Greenland ice cores)	^{10}Be, ^{14}C, ^{36}Cl
Bomb-peak record in recent ice	^{36}Cl, ^{41}Ca, ^{129}I
Lithosphere	
Exposure dating of rocks	^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{53}Mn
Paleoclimatic studies in loess	^{10}Be, ^{14}C
Tectonic plate subduction studies	^{10}Be
Neutron flux monitor in uranium minerals	^{236}U
Cosmosphere	
Meteorites, moon	^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{53}Mn, ^{59}Ni, ^{60}Fe
SN remnants on earth	^{26}Al, ^{60}Fe, ^{244}P, [^{146}Sm, ^{182}Hf]
Stable trace isotopes in presolar grains	^{194,195,196,198}Pt
Geochemical solar neutrino detection	[^{97,98}Tc, ^{205}Pb]
Search for superheavy elements in nature	Eka-Th, Ds, Rg, Fl, Eka-Bi (A~300, Z~114)
Search for exotic particles in nature	free quarks, heavy isotopes, strange matter
Technosphere	
Half-life measurements	^{32}Si, ^{41}Ca, ^{44}Ti, ^{60}Fe, ^{79}Se, ^{126}Sn, ^{146}Sm
Depth profiling in fusion walls	^{3}H
Possible fusion plasma thermometer	^{27}Al(n,2n)^{26}Al
Reaction studies for nuclear astrophysics	^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{44}Ti, ^{55}Fe, ^{59}Ni , ^{63}Ni, ^{68}Ge, ^{146}Sm, ^{210m}Bi
Neutron dosimetry of the Hiroshima bomb	^{36}Cl, ^{41}Ca, ^{63}Ni
Nuclear safeguards and nuclear technology	$^{146, 149, 151}$Sm, ^{202}Pb, $^{229, 233}$Th, ^{231}Pa ^{233}U, ^{236}U, ^{237}Np, $^{239, 240, 241, 242, 244}$Pu

[a]Radionuclides measured with AMS in the respective area of application [2]. Man-made radionuclides are underlined, and square brackets indicate a potential use in the future.

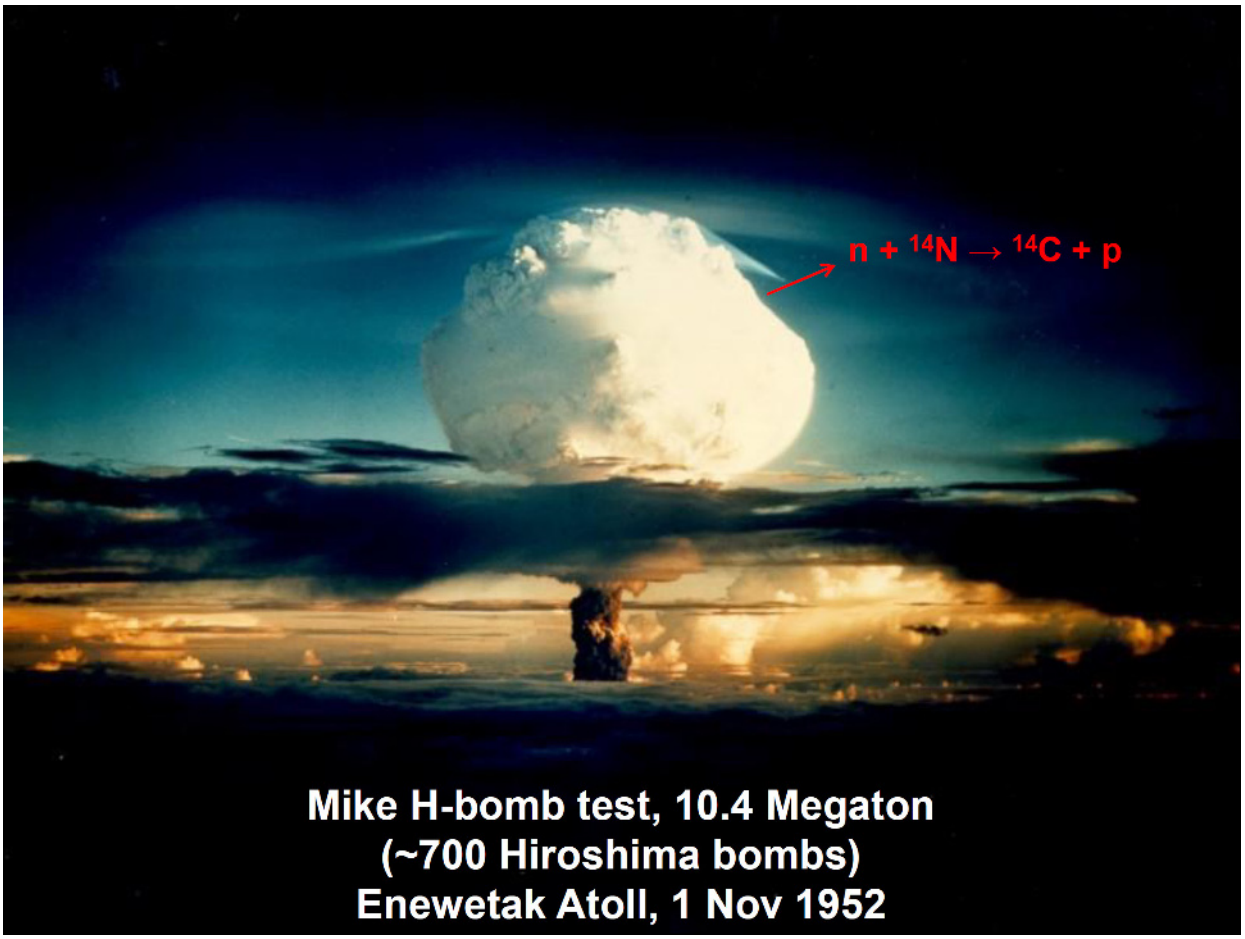

Figure 3. Picture of the first US test of a hydrogen bomb at the Marshall islands in 1952. This signaled the beginning of large above-ground nuclear weapon testing which lasted until 1963, when the nuclear test ban treaty (NTBT) stopped these activities. The neutron flux from the nuclear explosions produced the excess of ^{14}C depicted in Figure 4.

3.1 Dating human DNA with the ^{14}C bomb peak

A few years ago, an exciting possibility to determine retrospectively the birth of cells in humans was developed at the Karolinska Institute in Stockholm [18]. The above-ground nuclear weapons testing after the Second World War (Fig. 3) added significant amounts of ^{14}C to the atmosphere, which led to a doubling of the ^{14}C content by 1963 (Fig. 4). At this time the limited nuclear test ban treaty put a halt to the testing, and in the ensuing 50 years the atmospheric ^{14}C dropped back to almost normal through the exchange of $^{14}CO_2$ with the biosphere and the ocean [20]. This rapidly varying ^{14}C excess effectively labeled every living species on Earth including humans, and allows dating with a resolution of one to two years. (It should be noted that for ^{14}C bomb peak dating the radioactive decay of ^{14}C is negligible, see Fig. 4b).

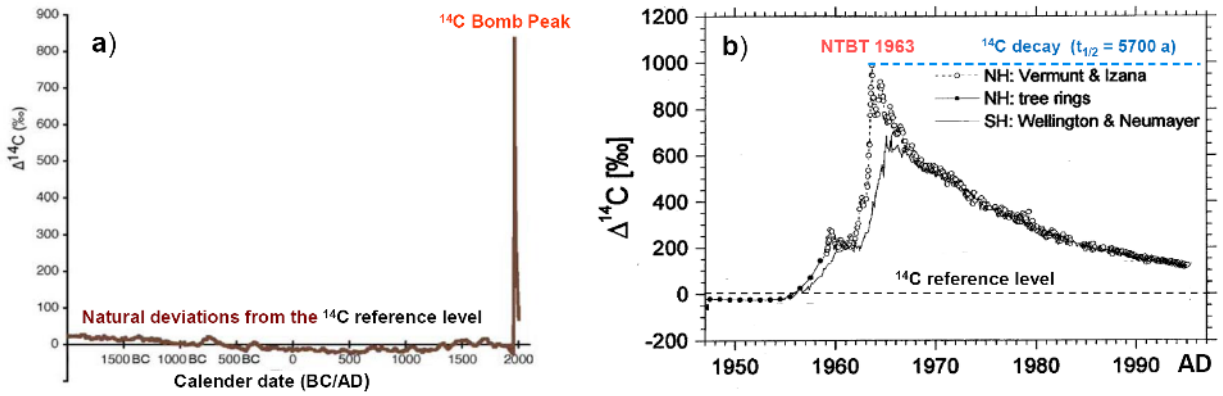

Figure 4. Plot of the atmospheric ^{14}C deviations ($\Delta^{14}C$) from a constant reference level ($^{14}C/^{12}C = 1.2 \times 10^{-12}$) as a function of time. (a) The long-term trend clearly indicates the large ^{14}C excess produced by nuclear weapons testing [18]. (b) Details of the ^{14}C bomb peak during the second half of the 20th century from atmospheric $^{14}CO_2$ measurements in the northern and southern hemisphere [19].

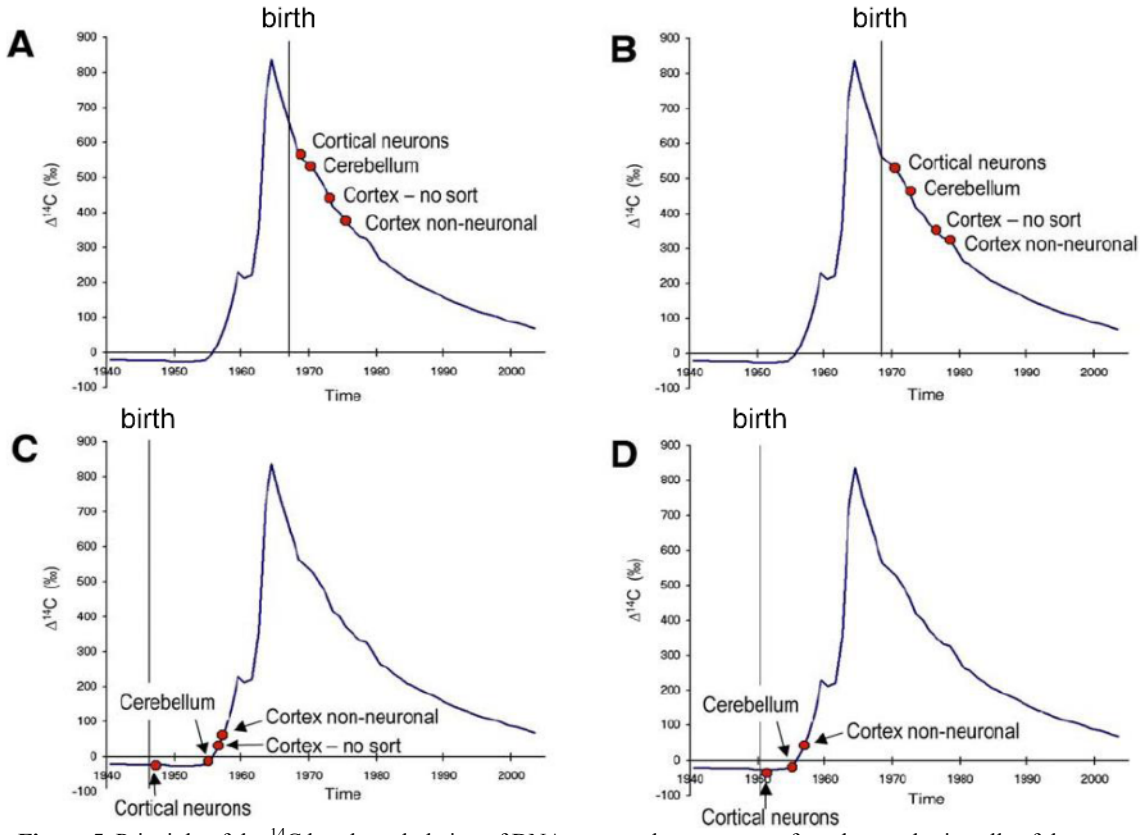

Figure 5. Principle of the ^{14}C bomb peak dating of DNA extracted *post mortem* from human brain cells of the cortex and the cerebellum [18]. The ^{14}C content measured in different cells (red dots) can be interpreted as the average time elapsed after birth (vertical line) to form the particular cell population. The results are shown for individuals born after the NTBT (A, B) and before it (C, D).

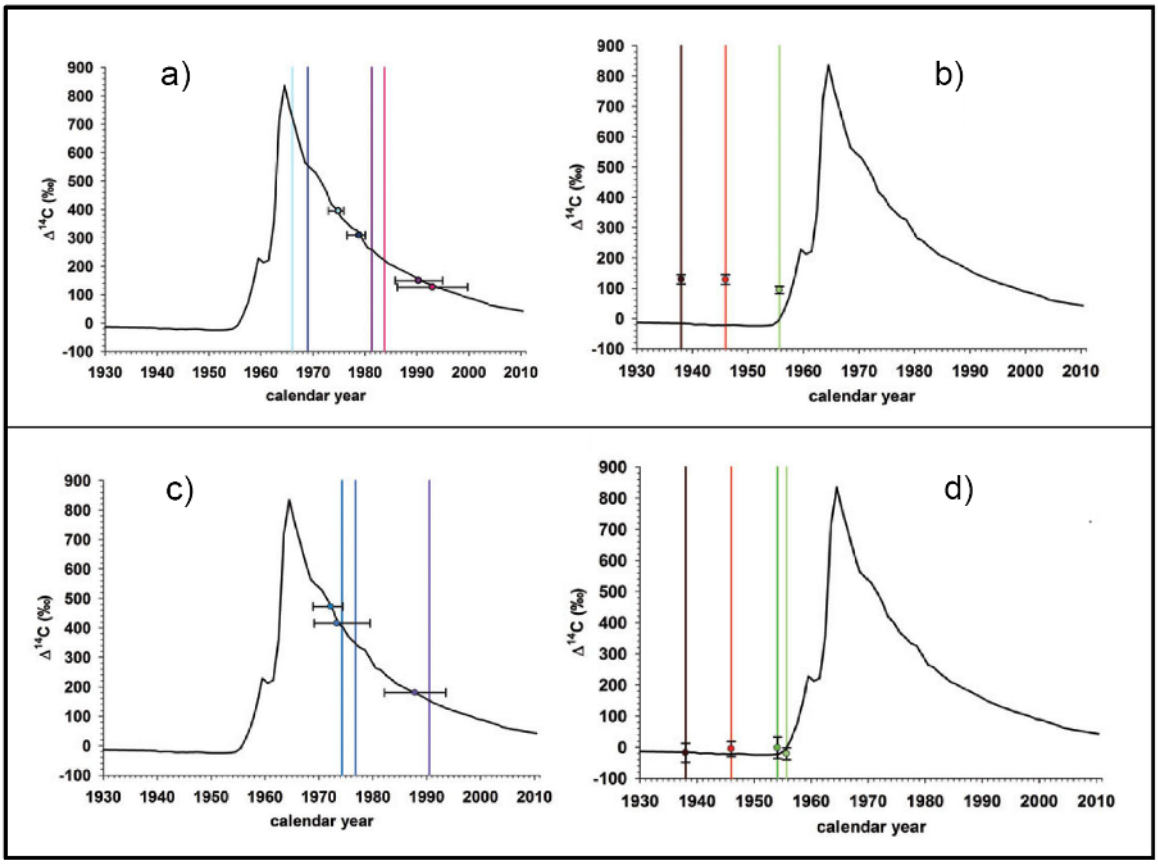

Figure 6. Results of ^{14}C bomb peak dating of the human olfactory bulb [21]. Vertical lines indicate the birth date of individuals investigated. The upper panel shows the results for non-neuronal cells (a, b), and the lower one for neurons (c, d). The latter one indicate very little if any neurogenesis after birth.

The basic principle to deduce the birth date of cells in humans is quoted here from Spalding et al. [18]: "Most molecules in a cell are in constant flux, with the unique exception of genomic DNA, which is not exchanged after a cell has gone through its last division. The level of ^{14}C integrated into genomic DNA should thus reflect the level in the atmosphere at any given point, and we hypothesized that determination of ^{14}C levels in genomic DNA could be used to retrospectively establish the birth date of cells in the human body." Figure 5 is reproduced from this seminal work and demonstrates the power of the method.

In a collaboration between the VERA lab in Vienna and the Karolinska Institute in Stockholm the olfactory bulb system in humans was investigated [21]. Here only a few microgram of DNA were available for the ^{14}C AMS measurements, demanding a particular careful sample preparation in every step from the human tissue to the final carbon sample [3]. Figure 6 shows that very little if any new neurons were formed in the human olfactory bulb after birth [21], in stark contrast to rodents where 50% turnover in adulthood has been observed.

However, in the most recent work of Spalding et al. [22] neurogenesis throughout adulthood was found in the human hippocampus, adding to the plasticity of this important part of the brain (Fig. 7).

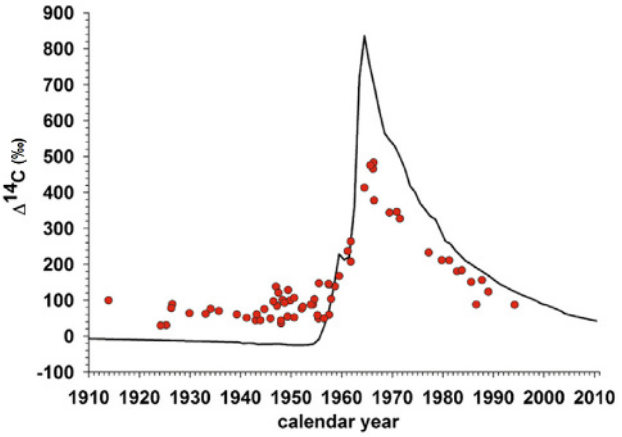

Figure 7. Results of ^{14}C bomb peak dating of the human hippocampus [22]. The $\Delta^{14}C$ values (red dots) for 64 individuals are plotted at their respective birth dates. Values above the curve before the rise of the ^{14}C excess, and below the curve after the peak therefore indicate neurogenesis after birth.

3.2. Search for superheavy elements in nature

In 1969 Glenn Seaborg schematically extended the periodic table beyond element 103 (Lawrencium), which was the heaviest known element at that time [23]. In the ensuing 40 years, elements 104 to 118 were synthesized

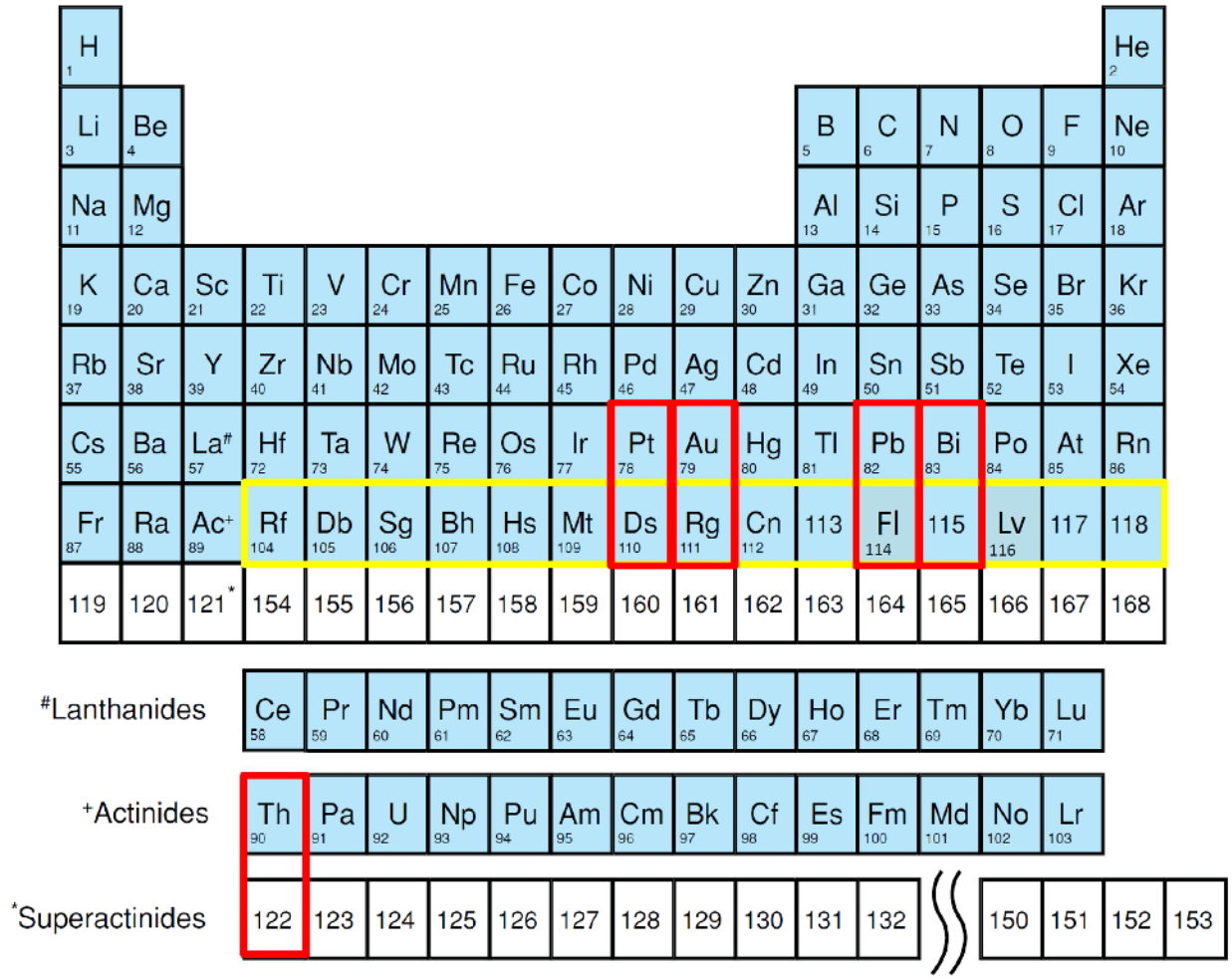

Figure 8. The periodic table of the elements based on the schematic extension beyond element 103 proposed in 1969 [23]. The yellow-framed elements (104 through 118) have since been produced in heavy ion reactions. None of the uncolored elements have yet been observed. The search for superheavy nuclides was performed with natural materials assuming that the SHE homologues, indicated by the red frames, will follow their respective host material (see text).

in heavy ion reactions at Berkeley, Dubna, GSI, and Riken (Fig. 8). These elements are commonly called superheavy elements (SHE).

Already around the time of Seaborg's extension of the periodic table, nuclear shell model calculations revealed [24] that an area of increased stability may exist for nuclides around A~ 300, quickly named "island of stability". However, due to the large neutron excess of this island, it cannot be reached with present-day heavy ion reactions. This may have to await neutron-rich projectiles from future radioactive-beam facilities. So far, the only way to search for a possible existence of these superheavy nuclides were searches in nature [25, 26]. An early AMS experiment to search for the platinum homologue ^{294}110 (now ^{294}Ds) in a Pt placer nugget was performed at the tandem accelerator of the University of Pennsylvania [27]. An abundance limit of 10^{-11} was found for ^{294}110 in Pt by assuming that the half-life was about 100 million years and the abundance in r-processed stellar material at the time of the formation of the solar system was 0.03 [28]. In none of the experiments [25-27] evidence for SHEs in nature was found.

However, the group of A. Marinov et al. from Jerusalem recently reported positive evidence from high-resolution inductively coupled plasma – sector field mass spectrometry (ICP-SF-MS) for long-lived neutron-deficient Th isotopes [29], neutron deficient Rg isotopes in gold [30], and a SHE nuclide with A = 292 and Z ~ 122 in thorium [31]. This triggered extensive AMS experiments at the 14-MV tandem accelerator in Munich [32, 33], and at the 3-MV VERA facility in Vienna [34-36]. None of the Marinov results were confirmed with AMS, with abundance limits several orders of magnitude below the one of the ICP-SF-MS experiments. In addition, abundance limits were set for 30 nuclides within the 'island of stability' with respect to their host materials, Pt, Au, Pb, and Bi [35, 36]. The results are summarized in Figure 9. Although an explanation for the positive results of the late Amnon Marinov and his group has not been found, it is possible that unidentified background caused the observed events. AMS is far superior over ICP-SF-MS to remove spurious background events.

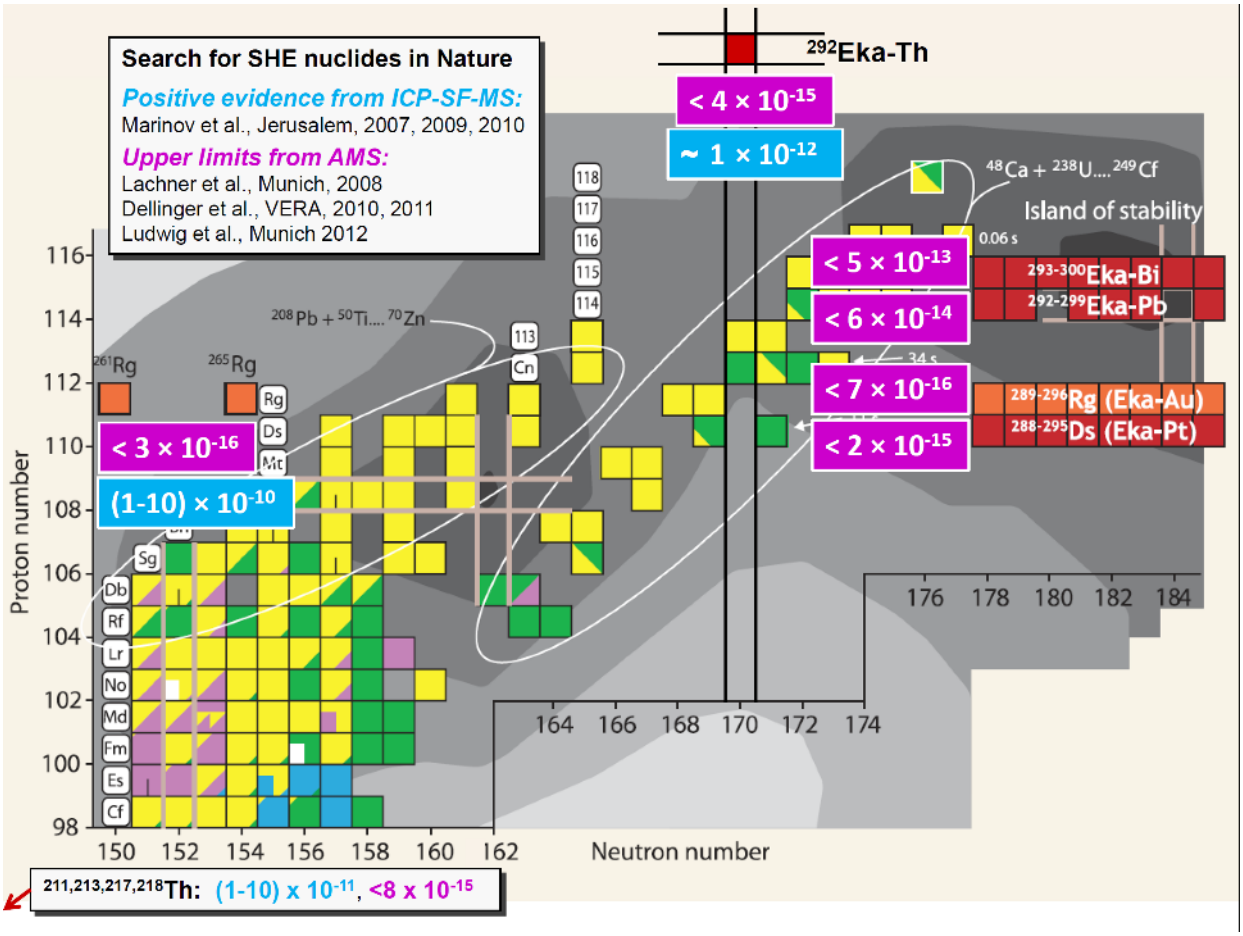

Figure 9. Summary of the results for searches of superheavy nuclides in nature. The basic layout of the figure is from [37], depicting the upper end of the chart of nuclides. The shades of gray in the background indicate the relative stability of nuclides due to shell model corrections (darker means more stable). Nuclides marked in orange and red have been measured with AMS. Abundance limits measured with respect to the corresponding host material (see Fig. 8) are given in the violet boxes. The positive evidence of the Marinov experiments are shown in the blue boxes.

An alternative to AMS experiments is the search for high neutron multiplicity (>3) from spontaneous fission events of SHEs. Such an experiment is performed by the Dubna-Orsay collaboration on 550g Osmium in the Frejus tunnel underground laboratory [38]. An upper limit for the abundance of Eka-Osmium (Hs) in Osmium at the level of 10^{-14} has been reported.

4 Conclusion

Accelerator mass spectrometry (AMS) is arguably the most powerful analytical tool to measure minute traces of long-lived radionuclides in almost any domain of the environment at large. In this paper the enormous breadth of applications could only be touched upon briefly. A more complete description of AMS applications can be found in Ref. [2]. Technical developments of AMS are described in Ref. [1].

The ^{14}C bomb peak dating shows great promise to study neurogenesis in various sections of the human brain. Other parts of the body can be studied as well, e.g. heart muscles [39] and fat cells [40]. It is indeed rewarding that something so useful comes out from the nuclear weapons testing period.

The basic problem for searches of SHEs in nature can perhaps be described by asking: "Is the absence of evidence, evidence of absence?" An answer to this question can only be approached by improved measurements and improved (theoretical) predictions about stellar production, physical and chemical properties, and half-lives. This will gradually reduce potential loopholes, eventually leading to harder evidence for a possible existence of SHEs in nature..

The newest developments of suppressing interfering isobars at the injection side of AMS tandem facilities, will likely make the use of additional radionuclides feasible and expand the use of small AMS facilities. This will enlarge applications of AMS into areas not yet available.

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