PHYSICS BEYOND THE STANDARD MODELS OF PARTICLES, COSMOLOGY AND ASTROPHYSICS

Proceedings of the Fifth International Conference - Beyond 2010

Cape Town, South Africa, 1 – 6 February 2010

H V Klapdor-Kleingrothaus
Heidelberg, Germany

I V Krivosheina
Heidelberg, Germany and Nishnij Novgorod, Russia

R Viollier
University of Cape Town, South Africa

editors

World Scientific

NEW JERSEY · LONDON · SINGAPORE · BEIJING · SHANGHAI · HONG KONG · TAIPEI · CHENNAI

Published by

World Scientific Publishing Co. Pte. Ltd.

5 Toh Tuck Link, Singapore 596224

USA office: 27 Warren Street, Suite 401-402, Hackensack, NJ 07601

UK office: 57 Shelton Street, Covent Garden, London WC2H 9HE

British Library Cataloguing-in-Publication Data
A catalogue record for this book is available from the British Library.

PHYSICS BEYOND THE STANDARD MODEL OF PARTICLES, COSMOLOGY AND ASTROPHYSICS
Proceedings of the Fifth International Conference — Beyond 2010

ISBN-13 978-981-4340-85-4
ISBN-10 981-4340-85-5

Printed in Singapore by Mainland Press Pte Ltd.

EXOTIC ARCHAEOLOGY: SEARCHING FOR SUPERHEAVY ELEMENTS IN NATURE AND DATING HUMAN DNA WITH THE ^{14}C BOMB PEAK

WALTER KUTSCHERA, FRANZ DELLINGER, JAKOB LIEBL and PETER STEIER

Vienna Environmental Research Accelerator (VERA)
Faculty of Physics — Isotope Research, University of Vienna, Austria
walter.kutschera@univie.ac.at

This contribution conveys the power of accelerator mass spectrometry (AMS) to measure ultra-low traces of long-lived radionuclides in two highly divers fields: Astrophysics and molecular biology. Our search for nuclides of superheavy elements (SHE) in several natural materials did not confirm the claims of positive evidence for SHEs reported by the group of Amnon Marinov from Jerusalem, even though the sensitivity of our AMS measurements were several orders of magnitude higher. We also report on the investigation by the group of Kirsty Spalding from Stockholm to date human DNA with the ^{14}C bomb peak. This allows one to determine retrospectively the birth date of cells in sections of the human body. Ongoing efforts to miniaturize carbon samples down to the level of 10 μg C for AMS measurements will allow one to venture into ever smaller subsections of the human brain.

1. Introduction

The advancement of accelerator mass spectrometry (AMS) some 30 years ago opened the possibility to detect both natural and man-made, long-lived radionuclides down to isotopic abundance levels of 10^{-16}. It literally became possible to explore our world atom by atom in almost every section of the environment.[1] A well-known application of AMS is radiocarbon (^{14}C) dating in archaeology and other fields, which can be performed with samples of only 1 mg of carbon. This is at least thousand times less than what is required for the classical beta-counting method.

A basic feature of any AMS facility is the ability to achieve good overall efficiency (the fraction of atoms in the sample actually detected), and at the same time providing utmost selectivity for the ultra-rare nuclide, in order to separate it from a usually overwhelming background. If the analyzing magnets are strong enough, an AMS facility can be tuned to any nuclide of the nuclear chart, even allowing one to venture into unexplored areas.

A universal AMS facility such as the Vienna Environmental Research Accelerator (VERA) in Vienna can therefore be used to explore 'white' areas of the nuclear landscape, such as the predicted 'island of stability' where long-lived Super Heavy Elements (SHEs) may exist (see e.g. the overview of Flerov and Ter-Akopian[2]). It

may therefore not be unreasonable to look with a very sensitive method such as AMS for minute traces of long-lived SHEs in natural materials. Such measurements were recently performed at the AMS facilities of Munich[3] and of Vienna,[4] after the startling reports about positive findings of SHEs by the group of Marinov.[5–7] In Section 2 of this contribution, these experiments will be described.

The above example attempts to find nuclides, which may have been synthesized in stars before our solar system formed some 4.6 billion years ago. In Section 3 ^{14}C AMS measurements are discussed which trace events of the last 50 years. It is well known that atmospheric nuclear weapons testing during the late 1950s and early 1960s doubled the ^{14}C content in atmospheric CO_2.[8] After the Nuclear Test Ban Treaty in 1963, the exchange of CO_2 with the biosphere and the ocean led to a rapid decrease of this extra ^{14}C transferring it to the respective reservoirs. It is interesting to note that the instantaneous labeling of atmospheric CO_2 with 'bomb' ^{14}C has wide-reaching implications for studying the dynamics of the CO_2 exchange of the atmosphere with the biosphere and the hydrosphere, respectively.

The latter is important in studying the uptake of CO_2 in the ocean, which is an essential ingredient in understanding the global CO_2 cycle and the fate of the anthropogenic CO_2 increase in the atmosphere. And this extra ^{14}C also allows one to study retrospectively the time of formation of new cells in the human body.[9]

2. Search for Superheavy Elements in Nature

Calculating the stability of heavy nuclei has always been a challenge to nuclear theorists. In the 1960s an exciting possibility emerged, when shell-model corrections to the liquid-drop model indicated that there may be a neutron-rich 'island of stability' beyond any known nuclide.[10–13] Nucleosynthesis calculation indicated that such nuclei may be produced under stellar r-process conditions.[14] For the superheavy nucleus with $Z = 110$ and $N = 184$ a half-life of 2.5 billion years was calculated.[15] A specific AMS search for this isotope was therefore performed in 1980 on a platinum nugget.[16] It was assumed that element $Z = 110$ has similar chemical properties as platinum. No events of a $^{294}110$ isotope were observed with an abundance limit of $^{294}110/Pt = 1 \times 10^{-11}$. Assuming a supernova-produced $^{294}110/Pt$ ratio of 0.02 to 0.06[14] one can conclude from the observed abundance limit that the half-life must be less than about 200 million years, provided, that element 110 follows platinum throughout the geochemical and geophysical history. Many other searches for SHEs in natural materials were performed.[2,17] To this day, no confirmed evidence for long-lived SHEs in nature exist, even though some evidence has been reported for the occurrence of long-lived neutron-deficient isotopes of thorium[5] and of Roentgenium ($Z = 111$) in natural gold,[6] and of a SHE ($Z \sim 122$) nuclide of mass 294 in thorium.[7] These measurements were performed with high-resolution ICP-SFMS (Inductively Coupled Sector Field Mass Spectrometry). Since it seems difficult to measure the reported abundance levels in the 10^{-10} to 10^{-12} range with ICP-SFMS, these extraordi-

nary claims certainly need independent verification by other experimental techniques.

So far two searches with AMS were performed. One at the Maier Leibniz Laboratory of the LMU and TU Munich,[3] and another one and at the VERA Lab of the University of Vienna.[4] Figure 1. shows a schematic presentation of the AMS facility in Vienna, indicating the setup for measuring ultra-low abundances of long-lived nuclides in thorium materials.[4] Compared to ICP-SFMS, which essentially identifies the claimed SHEs only through high-resolution mass measurements, the AMS set-up at VERA utilizes a system which eliminates background by a highly redundant filtering process. This clearly is an advantage for proving the existence of such a rare species once a positive signal is observed. However, it must be pointed out that the complexity of the AMS system requires a very good calibration with known pilot beams (see Fig. 2), in order to be sure that the set-up is sensitive for a possible detection of SHE nuclides. Simply speaking it is easier to miss rare species

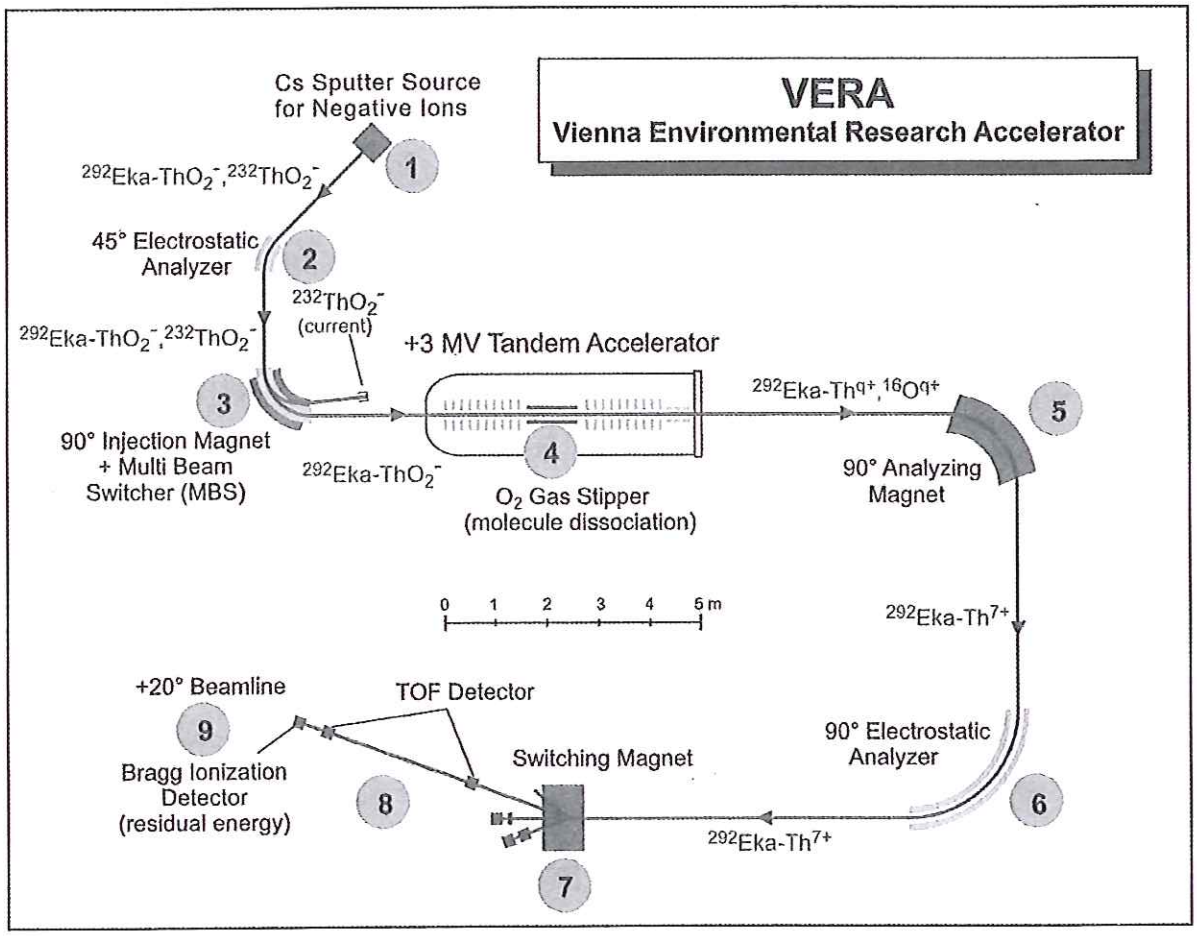

Fig. 1. Schematic presentation of the setup to search for ^{292}Eka-Th with VERA. The high selectivity to find rare events is indicated by the nine-fold filtering process of the ion beams generated from samples in the Cs-beam sputter source: (1) negative ion production, (2) energy/charge selection, (3) momentum/charge selection (mass sensitive), (4) break-up of molecules in the gas-stripping process, (5) momentum/charge selection, (6) energy/charge selection, (7) momentum/charge selection, (8) time of flight (velocity) measurement, (9) residual energy measurement.

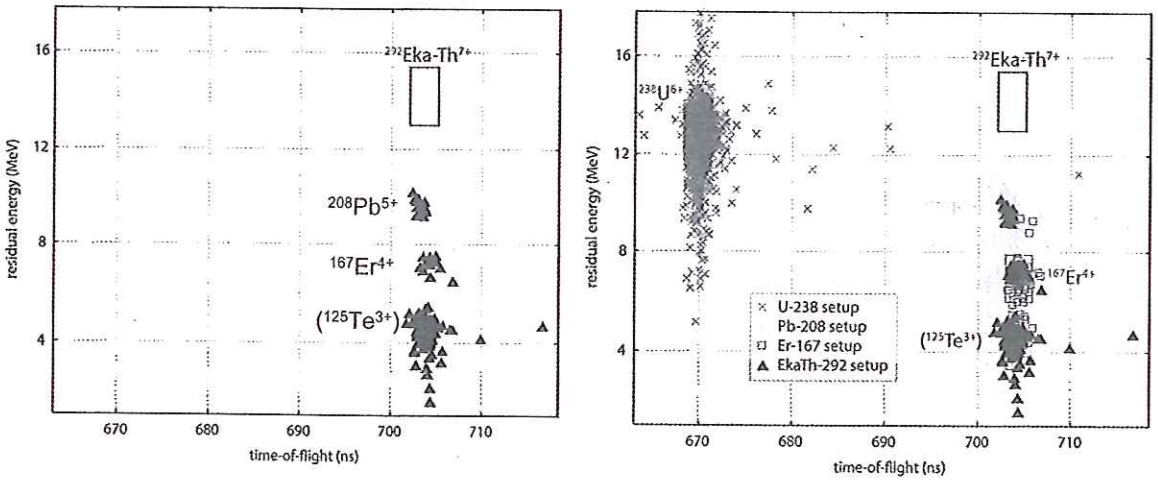

Fig. 2. Residual energy versus time-of-flight spectra measured in searching for the superheavy nuclide EkaTh-292. The left spectrum is recorded when the AMS system is set up for this nuclide only. The right spectrum shows an overlay of spectra, when the system was tuned to different pilot beams for calibration. No events for EkaTh-292 were recorded in the expected window.

with AMS than with ICP-SFMS. On the other hand, it is very difficult to be sure that events observed with ICP-MS are not caused by an unidentified background. Figure 2 shows the events observed in the search for ^{292}Eka-Th at VERA.[4] No events were observed in the region where one should have seen at least 1000 events if SHEs at the abundance level claimed by the Marinov group[7] were present.

Table 1. Summary of rare isotope measurements in thorium

Rare isotope	ICP-SFMS[a] (^{A}Th/^{232}Th)	AMS Munich[b] (^{A}Th/^{232}Th)	AMS Vienna[c] (^{A}Th/^{232}Th)
^{292}Eka-Th	$(1-10)\times10^{-12}$	not measured	$<4\times10^{-15}$
^{211}Th	$(1-10)\times10^{-11}$	$<9.6\times10^{-13}$	$<5\times10^{-15}$
^{213}Th	$(1-10)\times10^{-11}$	$<1.2\times10^{-12}$	$(7\times10^{-16}-8\times10^{-15})$[d]
^{217}Th	$(1-10)\times10^{-11}$	$<6.6\times10^{-13}$	$(1\times10^{-16}-6\times10^{-15})$[d]
^{218}Th	$(1-10)\times10^{-11}$	$<2.4\times10^{-12}$	$<5\times10^{-15}$

[a] $^{211-118}$Th,[5] ^{292}Eka-Th[7]
[b] Ref. 3
[c] Ref. 4
[d] For ^{213}Th two events, and for ^{217}Th one event was observed. These are, however, uncertain, see Ref. 4.

In Table 1 a comparison of the results for ICP-SFMS and AMS measurements performed so far in thorium materials is presented. It is clear that the limits of both AMS measurements are orders of magnitude lower than the observations of the Marinov group. The AMS results therefore exclude the existence of long-lived neutron-deficient thorium isotopes and of a ^{292}Eka-Th nuclide at the level reported by the ICP-SFMS measurements.[5,7]

At VERA the search for SHEs is continued with natural materials including platinum and gold nuggets, galenite (PbS), and bismuth ochre (Bi_2O_3). Here the

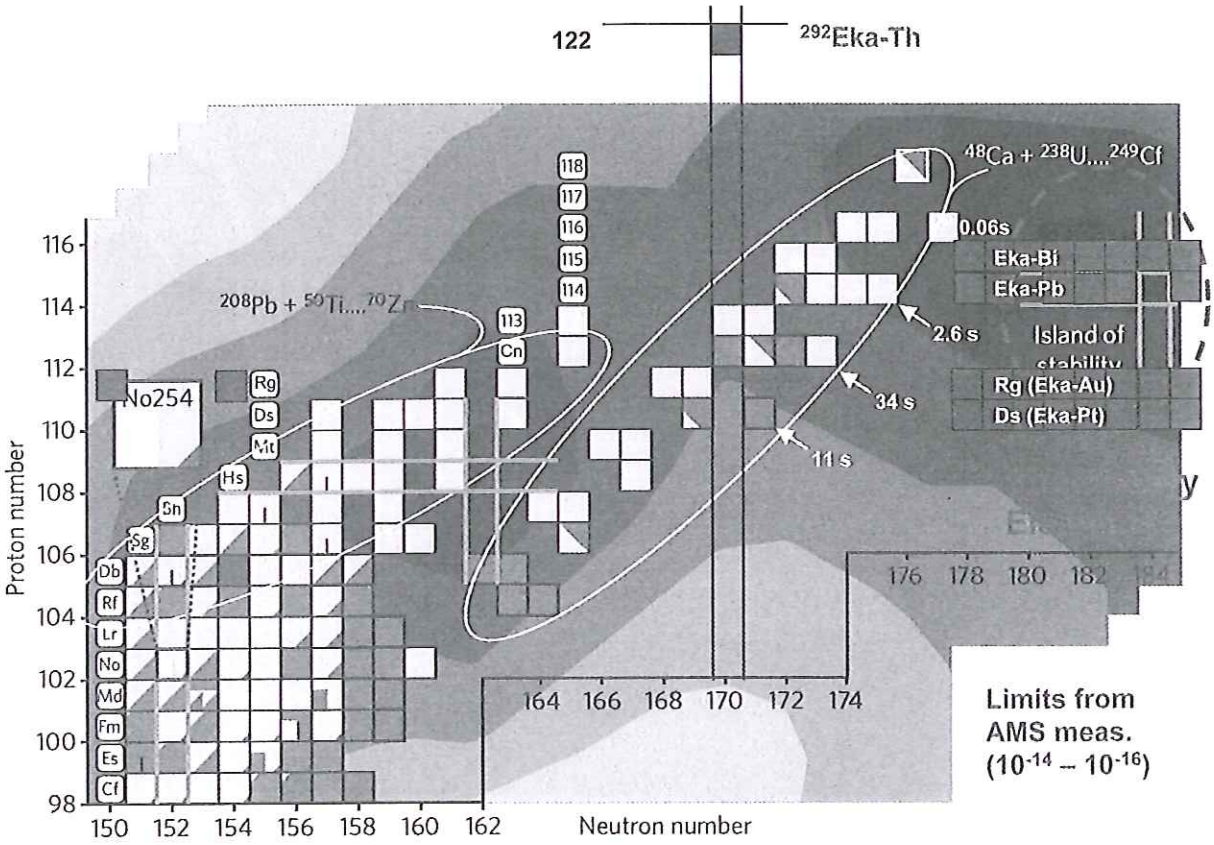

Fig. 3. Upper end of the chart of nuclides indicating the region of increased stability (dark shaded background) where superheavy nuclides are expected to be particularly stable.[21] The basic figure is adopted from Stoyer,[22] with the nuclides studied at VERA indicated by red squares.

assumption is that the eka-elements, Eka-Pt (Ds, Z = 110), Eka-Au (Rg, Z = 111), Eka-Pb (Z = 114), and Eka-Bi (Z = 115) follow the corresponding elements, although due to relativistic effects this may not be necessarily the case.[18,19] Of particular interest is gold, since positive results for the observation of long-lived isotopes of roentgenium (^{261}, ^{265}Rg) in gold have been reported by ICP-SFMS measurements.[6] So far, we have not found any evidence for these nuclides with three to four orders of magnitude higher sensitivities than the reported level of $(1 - 10) \times 10^{-10}$.[6] In addition, extensive searches for SHE nuclides in the vicinity of A $\sim$ 300 have not yet resulted in any positive evidence for the existence of such nuclides. Figure 4 summarizes the preliminary results of these searches, with abundance limits ranging from 10^{-14} to 10^{-16}.[20]

3. Dating Human DNA with the ^{14}C Bomb Peak

It is well known that after the Second World War, the superpowers (USA and USSR) started to test ever bigger nuclear bombs (fission and hydrogen fusion) in the atmosphere. This produced a number of radioisotopes, among them also ^{14}C. This happens because nuclear bomb explosions are accompanied by large neutron fluxes,

Fig. 4. Historical photo from the White House Treaty Room, where President John F. Kennedy (centre) signed the Nuclear Test Ban Treaty on October 3rd, 1963. On the far right is Vice President Lyndon B. Johnson (Photograph by Robert Knudsen, White House, in the John. F. Kennedy Presidential Library and Museum, Boston).

which convert nitrogen into ^{14}C through the $^{14}N(n,p)^{14}C$ reaction. Also natural ^{14}C is produced by the same reaction, with the neutrons coming from spallation reactions of cosmic ray protons on nuclei of the atmosphere.[23,24] Atmopsheric nuclear testing continued until 1963, when the Nuclear Test Ban Treaty (NTBT) put an end to atmospheric nuclear weapons testing (Fig. 4). By that time the ^{14}C content in atmospheric CO_2 had increased by 100%, but rapidly decreased thereafter due to the exchange of atmospheric CO_2 with the biosphere and the ocean (see Fig. 5).

Due to the exchange of atmospheric $^{14}CO_2$ with the ocean, the dynamics of this important process can be studied. On the other hand, it is remarkable that every species living in the second half of the 20th century got labeled with some bomb ^{14}C, since $^{14}CO_2$ enters the biosphere through the photosynthesis of plants. In 2005, an interesting paper was published in the journal Cell[9] reporting on the use of the ^{14}C bomb peak to retrospectively determine the birth date of cells in humans.

The basic idea is that ^{14}C in genomic DNA reflects the birth date of cells. The authors state in their paper[9]:

"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 time point, and we hypothesized that determination of ^{14}C

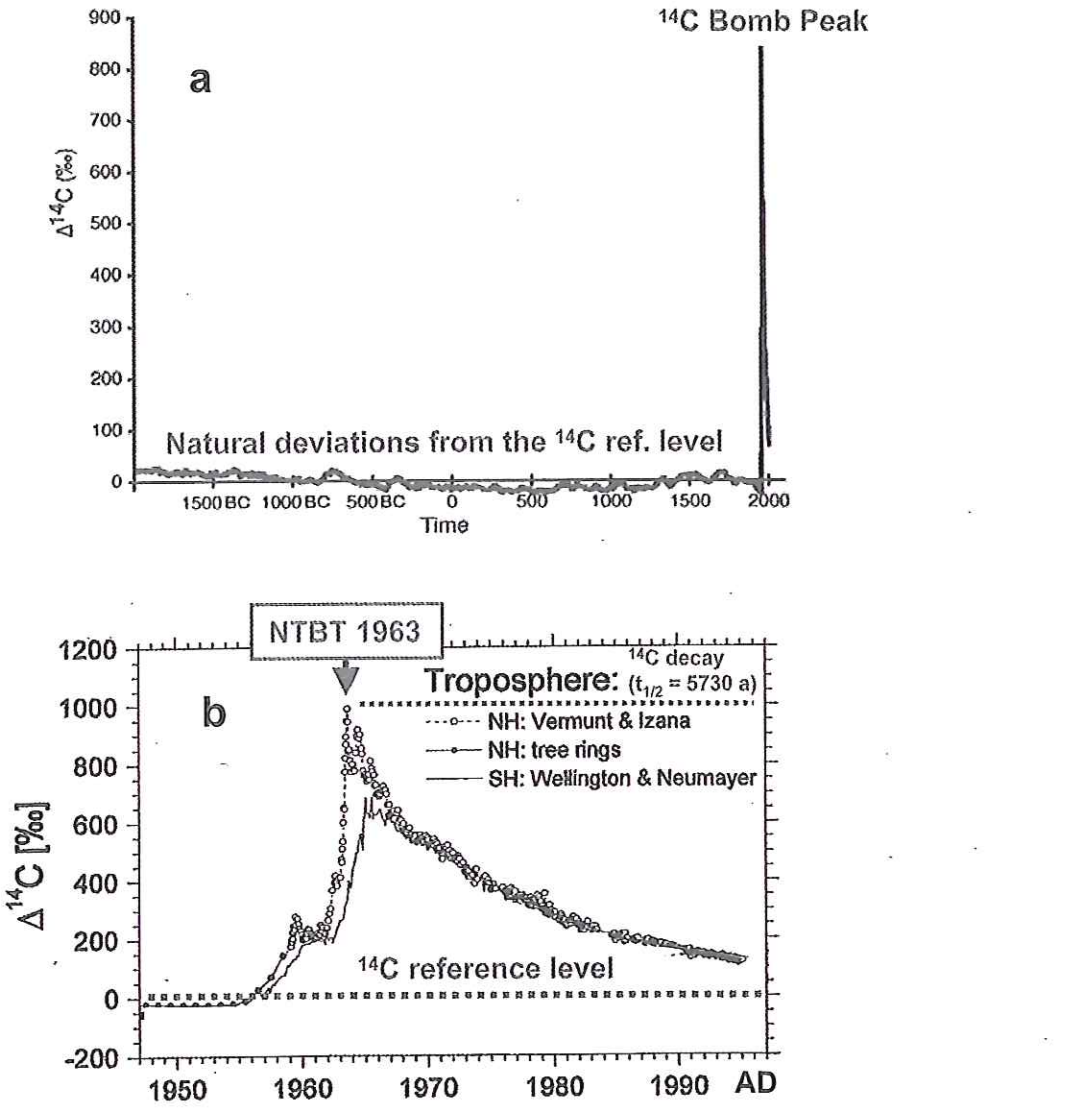

Fig. 5. (a) Variation of the ^{14}C content in atmospheric CO_2 during the last 4000 years including the ^{14}C bomb peak.[9] Deviations from the ^{14}C reference level ($^{14}C/^{12}C = 1.2 \times 10^{-12}$) are given. (b) Detail of the bomb peak during the last 50 years from measurements of ^{14}C in atmospheric CO_2 in the northern and in the southern hemisphere.[8] The decrease of ^{14}C due to radioactive decay is negligible (red dashed line) compared to the effect of CO_2 exchange with the biosphere and the ocean.

levels in genomic DNA could be used to retrospectively establish the birth date of cells in the human body."

The task of actually measuring $^{14}C/^{12}C$ ratios in human DNA can be assessed from Table 2 which summarizes its basic constituents. Even though the DNA molecule is very large containing about 10^{11} carbon atoms, one needs DNA from 10 cells to obtain one ^{14}C atom. With DNA extracted from 15 million cells, one obtains 1.5 million ^{14}C atoms and 36 μg of carbon. Since about 2% of the ^{14}C atoms can be counted with AMS, there is enough signal to determine the $^{14}C/^{12}C$ ratio. However the small amount of total carbon available for these measurements is still a challenge to the AMS technique. In recent years, progress in sample preparation

Table 2. Composition of the DNA molecule and its ^{14}C content (a physicist's view)

Basic composition of DNA:	Macromolecule with 3×10^9 base pairs (bp)
Chemical sum formula per bp:	$C_{20}H_{23}N_7O_{13}P_2$ and $C_{19}H_{22}N_8O_{13}P_2$
Molecular weight:.	~630 Daltons (Da) per base pair, total $\sim 1.9 \times 10^{12}$ Da
Mass of DNA per cell:	2 DNA per cell $= 2 \times 3$ pg $= 6$ pg
Mass of carbon (40 wt% C):	2.4 pg
Total length of DNA per cell:	$2 \times 3 \times 10^9 \times$ (0.34 nm, distance between bp) $= 2$ m
C atoms of DNA per cell:	$2 \times 3 \times 10^9 \times$ (20 C) $= 1.2 \times 10^{11}$ C atoms
$^{14}C/^{12}C$:	1.2×10^{-12}
DNA of 10 cells:	~ 1 ^{14}C atom
15 million cells:	1.5 million ^{14}C atoms
C from DNA of 15 million cells:	~ 36 μg C
Tote ^{14}C AMS detection efficiency:	$\sim 2\% \rightarrow \sim 30,000$ ^{14}C atoms detected

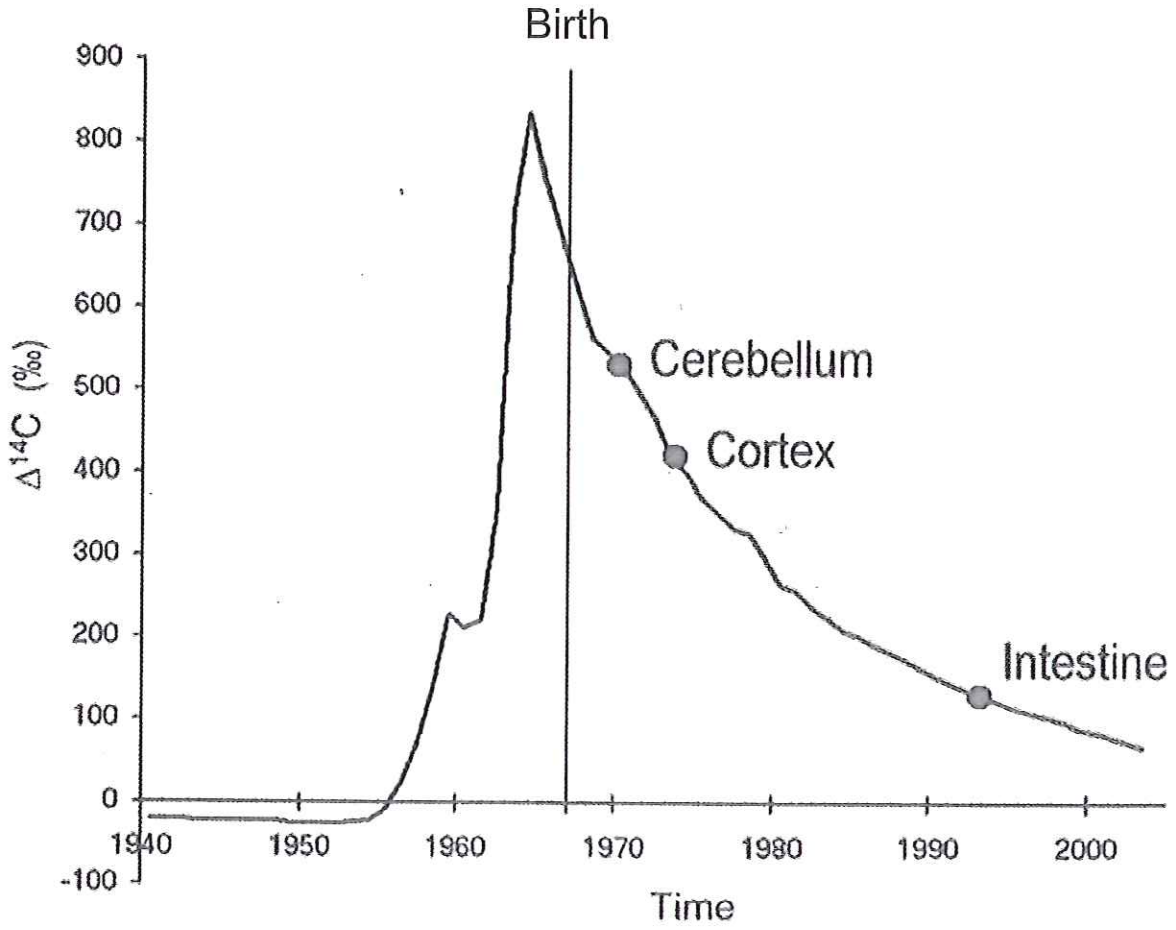

Fig. 6. The ^{14}C content of different human cells indicates the time after birth when the respective cells were formed.[9] The vertical line shows the birth date of the individual.

and background reduction allows one to reliably measure ^{14}C in samples down to 10 μg carbon.[25]

An example of the different times when human cells are formed after a person's birth is shown in Fig. 6. It is obvious from the figure that the ^{14}C content of the DNA from brain cells point to a time much closer to the birthdate than cells from the intestine, which are known to be frequently renewed. As for the brain itself,

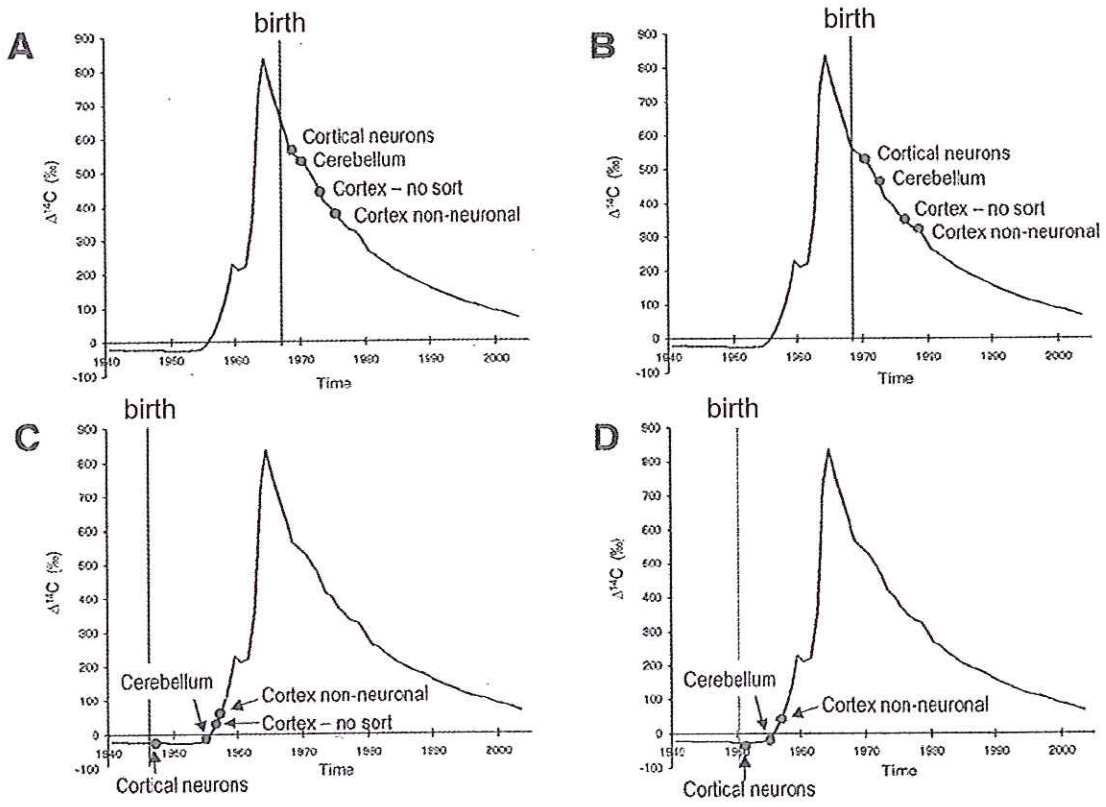

Fig. 7. Separation of neurons from non-neuronal cells show the different times after birth where they were formed.[9] Two individuals (A, B) were born after the bomb peak, and two (C, D) were born before the bomb peak.

one can see that cells of the cerebellum are generated closer to the person's birth date than the ones of the cortex. In Fig. 7 the brain of four different individuals were investigated, two of them born after the bomb peak (A and B), and two before the bomb peak. Differentiation into neuronal and non-neuronal cells shows that the cortical neurons are closest to the birth, whereas non-neuronal cells of the cortex point to a substantial production at a later stage in life. The method has been applied by the Stockholm group also for other questions such as the neurogenesis of the human neocortex,[26] turnover of fat cells in humans,[27] and the renewal of heart cells (cardiomyocites) in humans.[28]

4. Conclusion

AMS is capable of tracing long-lived radionuclides at ultra-low levels in almost any domain on Earth. This allows one to study natural and anthropogenic radionuclides at unprecedented low levels, literally 'atom by atom'. The two examples discussed here present two extreme applications of AMS. On the one hand, a search for long-lived superheavy elements in nature did not confirm the positive reports on their existence. On the other hand, the ^{14}C bomb peak — once considered only a menace

to man — turned out to be a valuable tool in studying one of the most interesting questions in molecular biology: When do cells in the human body form, and what is their renewal rate. AMS thus demonstrates its analytic power of studying questions about the beginning of our Solar System as well as of the intricacies of cell formation in the human body.

Acknowledgment

The continuing collaboration with the group of Kirsty Spalding at the Department of Molecular Biology of the Karolinska Institute in Stockholm on the ^{14}C bomb peak dating of human DNA is greatfully acknowledged. We also thank Robin Golser for discussions on the manuscript.

References

1. W. Kutschera, Progress in isotope analysis at ultra-trace level by AMS, Int. J. Mass Spectrom. 242 (2005) 145.
2. G. N. Flerov, G. M. Ter-Akopian, *Superheavy nuclei, Rep. Prog. Phys.* **46** (1983) 817.
3. L. Lachner, I. Dillmann, T. Faestermann, G. Korschinek, M. Poutivtsev, G. Rugel, Search for long-lived isomeric states in neutron-deficient thorium isotopes, *Phys. Rev. C* **78** (2008) 064313.
4. F. Dellinger, O. Forstner, R. Golser, W. Kutschera, A. Priller, P. Steier, A. Wallner, G. Winkler, Search for a superheavy nuclide with A = 292 and neutron-deficient thorium isotopes in natural thorianite, *Nucl. Instr. and Meth. B* **268** (2010) 1287.
5. A. Marinov, I. Rodushkin, Y. Kashiv, L. Halicz , I. Segal, A. Pape, R. V. Gentry, H. W. Miller, D. Kolb, R. Brandt, Existence of long-lived isomeric states in naturally-occurring neutron-deficient Th isotopes, *Phys. Rev. C* **76** (2007) 021303 (R).
6. A. Marinov, I. Rodushkin, A. Pape, Y. Kashiv, D. Kolb, R. Brandt, R. V. Gentry, H. W. Miller, L. Halicz, I. Segal, Existence of long-lived isotopes of a superheavy element in natural *Au, Int. J. Mod. Phys. E* **18** (2009) 621.
7. A. Marinov, I. Rodushkin, D. Kolb, A. Pape, Y. Kashiv, R. Brandt, R. V. Gentry, H. W. Miller, Evidence for the possible existence of a long-lived superheavy nucleus with atomic mass number A = 292 and atomic number Z × 122 in natural Th, *Int. J. Mod. Phys. E* **19** (2010) 131.
8. I. Levin, V. Hesshaimer, Radiocarbon – a unique tracer of global carbon cycle dynamics, Radiocarbon **42/1** (2000) 69.
9. K. L. Spalding, R. D. Bhardwaj, B. A. Buchholz, H. Druid, J. Frisń, Restrospective birth dating of cells in humans, *Cell* **122** (2005) 133.
10. W. D. Myers, W. J. Swiatecki, Nuclear masses and deformations, *Nucl. Phys.* **81** (1966) 1.
11. S. G. Nilsson et al., On the spontaneous fission of nuclei with Z near 114 and N near 184, *Nucl. Phys. A* **115** (1968) 545.
12. V. M. Strutinsky, Shells in deformed nuclei, *Nucl. Phys. A* **122** (1968) 1.
13. S. G. Nilsson et al., On the nuclear structure and stability of heavy and superheavy elements, *Nucl. Phys. A* **131** (1969) 1.
14. D. N. Schramm, W. A. Fowler, Synthesis of superheavy elements in the r-process, *Nature* **231** (1971) 103.
15. E. O. Fiset, J. R. Nix, Calculation of half-lives for superheavy nuclei, *Nucl. Phys. A* **193** (1972) 674.

16. W. Stephens, J. Klein, R. Zurmhle, Search for naturally occurring superheavy element Z = 110, A = 294, *Phys. Rev. C* **21** (1980) 1664.
17. G. Herrmann, Superheavy-element research, *Nature* **280** (1979) 543.
18. E. Eliav, U. Kaldor, P. Schwerdtfeger, B. A. Hess, Y. Ishakawa, Ground state configuration of element 111, *Phys. Rev. Lett.* **73** (1994) 3203.
19. R. Eichler et al., Chemical characterization of element 112, *Nature* **447** (2007) 72.
20. F. Dellinger, PhD thesis, University of Vienna, in preparation.
21. A. Sobiczewski, K. Pomorski, Description of structure and properties of superheavy nuclei, *Prog. Part. Nucl. Phys.* **58** (2007) 292.
22. M. A. Stoyer, Island ahoy!, *Nature* **442** (2006) 876.
23. W. F. Libby, Atmospheric helium three and radiocarbon from cosmic radiation, Phys. Rev. 68 (1946) 671.
24. E. C. Anderson, W. F. Libby, S. Weinhouse, A. F. Reid, A. D. Kirshenbaum, A. V. Grosse, Natural radiocarbon from cosmic radiation, *Phys. Rev.* **72** (1947) 931.
25. J. Liebl, P. Steier, R. Avalos Ortiz, R. Golser, F. Handle, W. Kutschera, P. Steier, E. M. Wild, Studies on the preparation of small ^{14}C samples with an RGA and ^{13}C enriched material, *Radiocarbon* 52(2-3) (2010), in print.
26. R. D. Bhardwaj et al., Neocortical neurogenesis in humans is restricted to development, *Proc. Nat. Acad. Sci.* **103** (2006) 12564.
27. K. L. Spalding et al., Dynamics of fat cell turnover in humans, *Nature* **453** (2008) 783.
28. O. Bergman et al., Evidence for cardiomyocyte renewal in humans, *Science* **324** (2009) 98.