Operation experience of the BINP Accelerator Mass Spectrometer

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The AMS is mainly dedicated for research in archaeology, geology, biomedical science and other fields by measurements of the ratio between carbon isotopes. The accelerator mass spectrometry is an ultra-sensitive method of isotopic analysis. The ratio between isotopes ^{14}C (Radiocarbon) and ^{12}C in samples can be less than 10^{-15}. So, the counting of the individual atoms is used for detection of such low radiocarbon concentration.

The ratio $^{14}C/^{12}C$ in modern carbon (living organics) is about $1.2*10^{-12}$
The radiocarbon concentration in "dead" (very old organics) sample is almost zero.

The half-life of a radiocarbon is 5730 years.

The main advantages of AMS method compared to conventional radiometric method are the use of three orders of magnitude smaller samples and the measurement time is an order of magnitude shorter. Only about 1 mg of carbon samples is needed for AMS analysis. This is very important because usually samples either represent a great historical value or large sizes sample are not available.

Atomic and molecular isobars of radiocarbon

- ^{14}N (The negative nitrogen ions are unstable.)

- ^{13}CH, $^{12}CH_2$ and other molecular ions of mass 14
  (The ratio between molecular isobars ions and radiocarbon ions more than 10^8 in the carbon sample)

  Such molecular ions are stable from 1- to 2+ charge state.

Background ions problem:

The scattering and charge exchange processes
allow backgrounds ion to pass through electrostatic and magnetic filters.
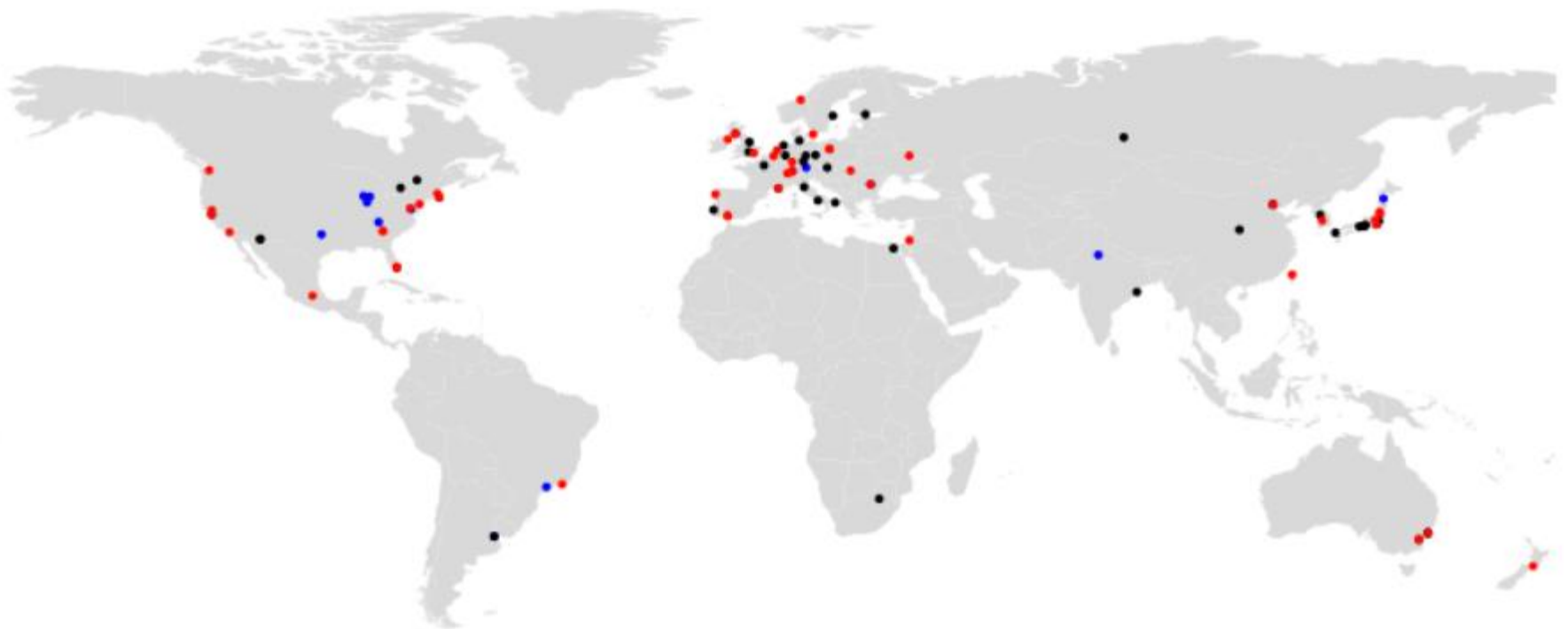(The ions can interact with molecules of residual gas and parts of vacuum chamber.)
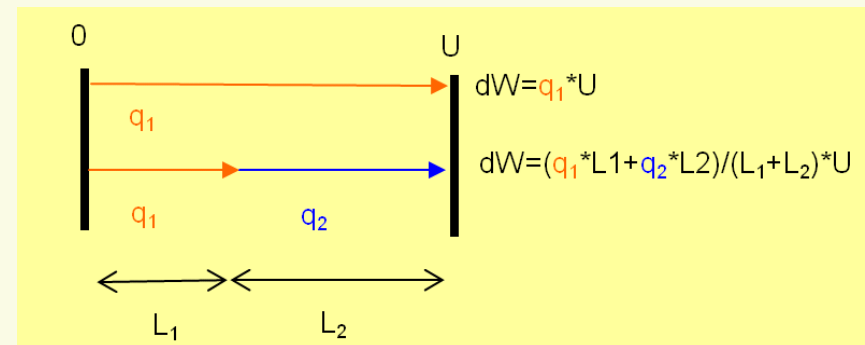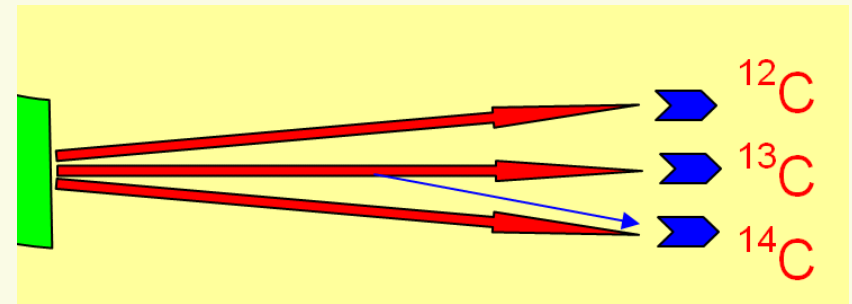
About 100 AMS facilities in the world

ETH
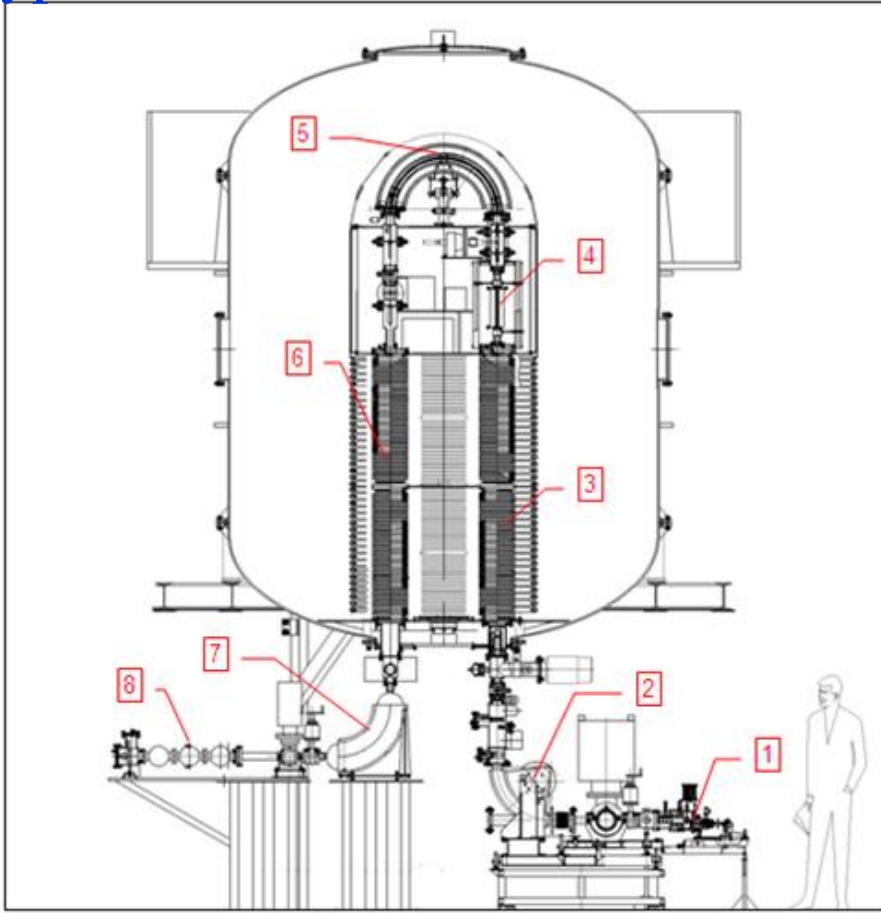Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

AMS facilities around the World

BINP AMS

The BINP AMS is based on a folded type electrostatic tandem accelerator

BINP AMS layout: 1 - ion source, 2 - low energy beam line magnet, 3 - first accelerating tube, 4 - magnesium vapors stripper, 5 - 180^0 electrostatic bend, 6 - second accelerating tube, 7 - high energy beam line magnet, 8 - TOF telescope

The ion source is necessary to formation of the negative ions beams from the atoms of the test sample.

The low energy beam line is used for initial isotope selection.

The tandem accelerator is necessary for rejection of the molecular ions.

The high energy beam line is intended for the further ions selection.

The final detector is used for accurate rare ion registration and counting.
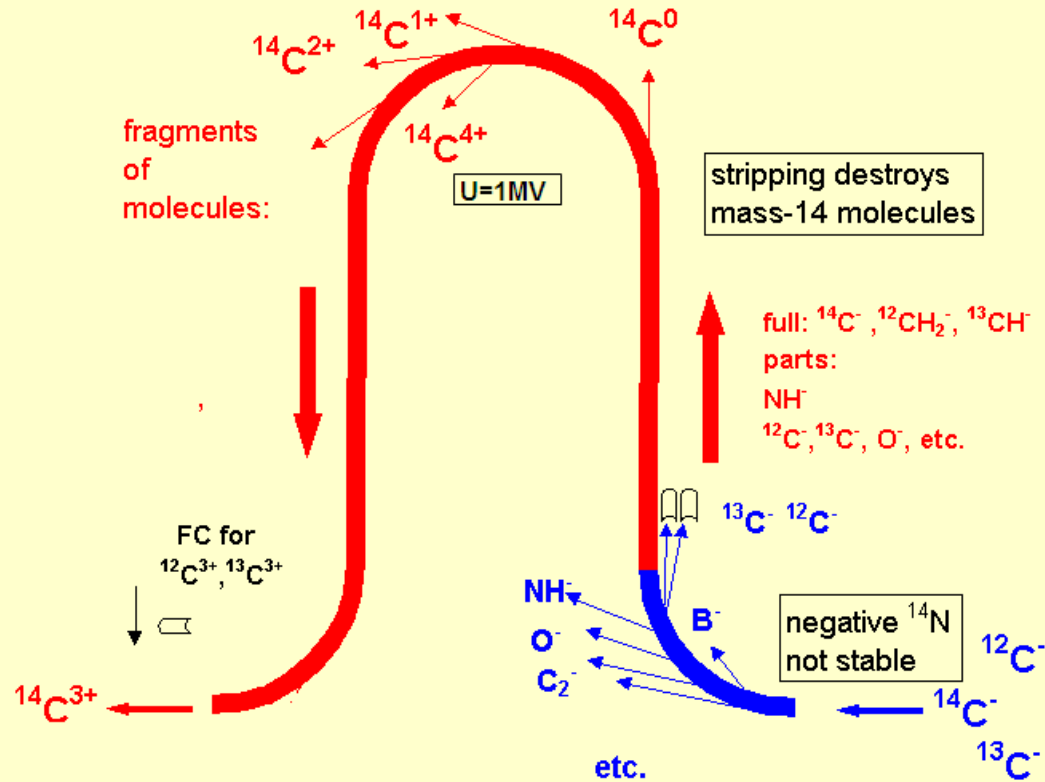
The multi-cathode (for 23 samples) sputter ion source is used for AMS analysis. The negative ions are produced by bombarding the graphite target with positive cesium ions and are horizontally extracted from the ion source. Then the beam is analysed at low energy with 90° magnet and vertically injected into the first accelerating tube through injection channel. The negative ions are accelerated to the 1 MV high voltage terminal and stripped to 3+ charge state in magnesium vapors stripper. Then they pass through the 180^0 electrostatic bend working as separator and then again vertically accelerated into the second accelerating tube to the ground potential. The ions after tandem accelerator are separated in 90° magnet and rare ions move horizontally to the final detector through high energy channel.

Most important distinguishing features of BINP AMS

- The ion energy selection just after molecular destruction (by **180^0 electrostatic bend into the high voltage terminal**) → **effective filtration of the molecular fragments**, because energy of fragments always less then ion energy (at this moment).

- The **magnesium vapor target** as a molecule destroyer → **localized molecular destruction**

- **2D time of flight detector** → **accurate recognition of each ion**

Magnesium vapors stripper

The magnesium vapors stripper is used in BINP AMS instead of the gas stripper usually applied in AMS. The use of magnesium vapors stripper allows obtaining the vacuum level being comparable with systems with solid targets, because the magnesium vapor is condensed into the special containers at the room temperature, which are placed at the entrance and exit of the stripper tube. Gaseous target requires the high-performance pumping system in high voltage terminal to reduce gas flux into accelerator tubes. The residual gas in accelerating tubes leads to big energy spread in the beam and scattering resulting in background growth thus limiting the sensitivity and accuracy of spectrometer. Solid targets on these energies have a short lifetime and therefore are not applied.
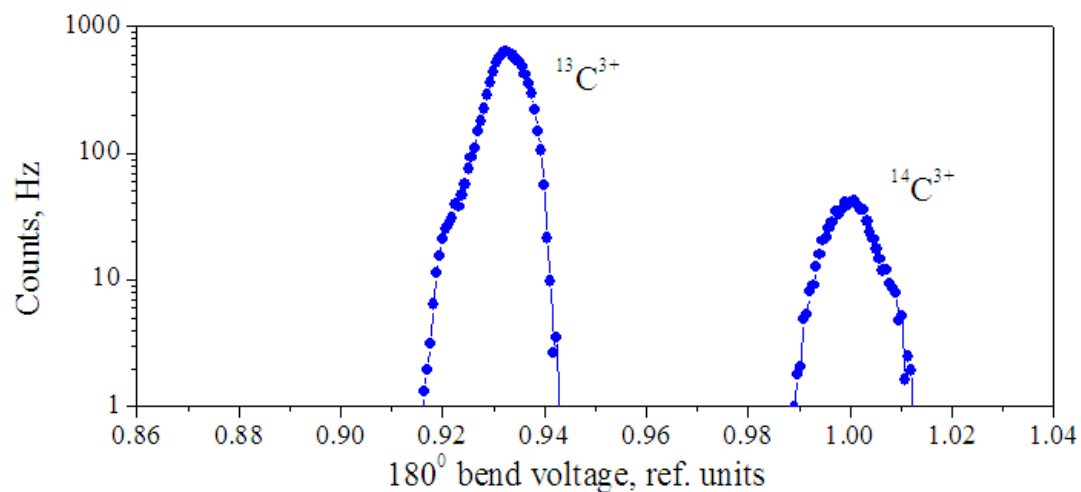
Time-of-flight telescope
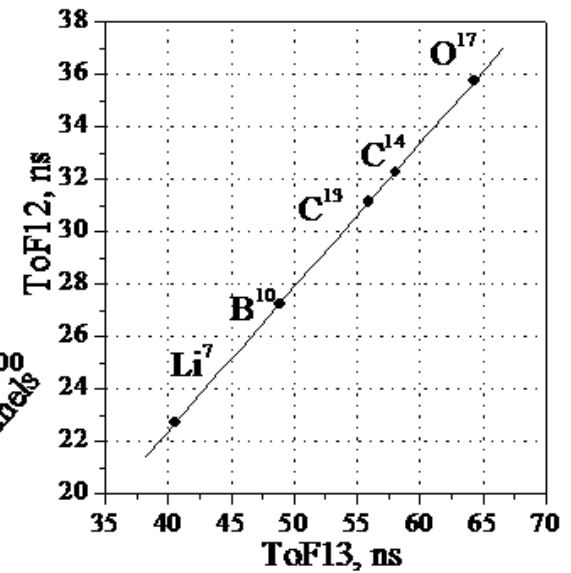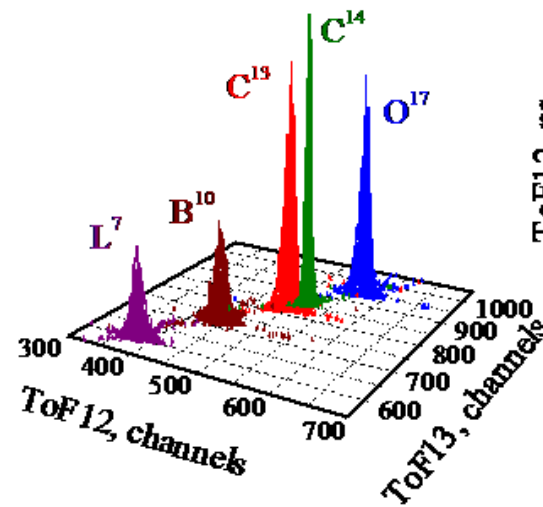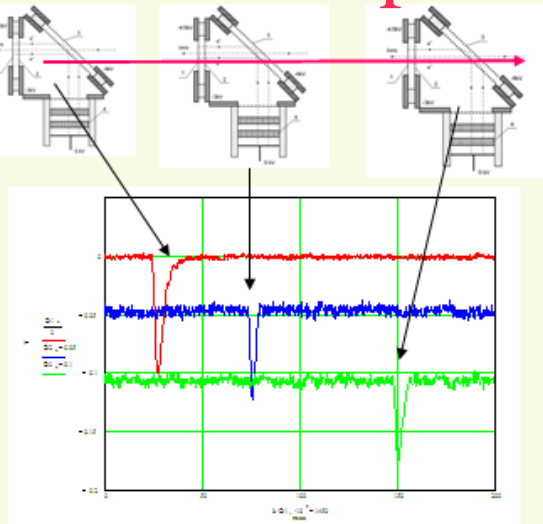
TOF telescope calibration by different atoms detection.
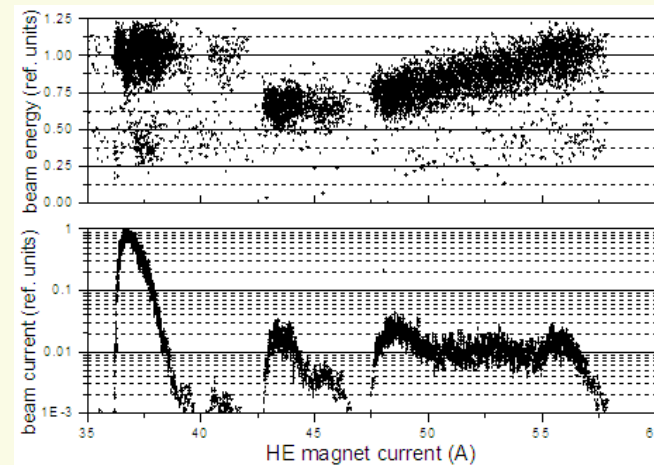
AMS settings were changed to pass each atom type.

The TOF channel width is about 70 ps. The atoms are well detected by the TOF telescope. But, when an electrostatic breakdown occurs in an ion source (or in other electrostatic elements), a large number of background ions can pass through the full system of selection and reach the TOF telescope. Tails from intense ion peaks can reach to the TOF detection area of other atoms. The moment of time for ion detection is registered with 16 us channel width in BINP AMS. This data is used for calculation of number of detected ions per unit time, that allows to numerically discard the background ions from electrical breakdowns.

180^0 electrostatic bend into the high voltage terminal

The most part of the background ions are removed by this filter. Interfering isobaric molecules are destroyed by collisions in the stripper into the terminal and selected immediately after stripping process. It is important to decrease the background from molecular fragments before the second acceleration stage because otherwise they can obtain large energy spread by recharging on residual gas into electric field


Ion background when scanning high energy magnet

The TOF telescope counts as a function of 180^0 bend voltage. The ^{13}C fragments of the molecules ^{13}CH are reliably separated from the radiocarbon ^{14}C.

The voltage value "1" corresponded to optimal $^{14}C^{3+}$ passing. The main ion peak $^{14}C^{3+}$ and additional peak are visible. The additional ion peak corresponded to voltage value about 13/14. It is ^{13}C ions from ^{13}CH ions, because the velocities of ions fragments and primary ions are equal in tandem terminal position. These fragments of the destructed molecular ions are filtered by tandem bend at operational bend value "1", and can not take part in charge exchanging process in the second accelerator tube.

The 180⁰ electrostatic bend is effective for the isobaric background filtration.

The negative nitrogen ions are unstable, but the negative molecular NH⁻ ions can rich high voltage terminal. Such ions can be produced in the ion source or from residual gas near the entrance of the accelerating tube. The NH⁻ ions produced inside the ion source can be significantly filtered in low energy channel, by mass difference. The NH⁻ ions from residual gas are accelerated at first tube without filtration . The positive nitrogen ions are produced from the breakup of NH ions after passing through the stripper. The energies for nitrogen from NH molecules are always less than the radiocarbon ions energy into tandem terminal, because nitrogen ion gets only a part of the molecules energy.
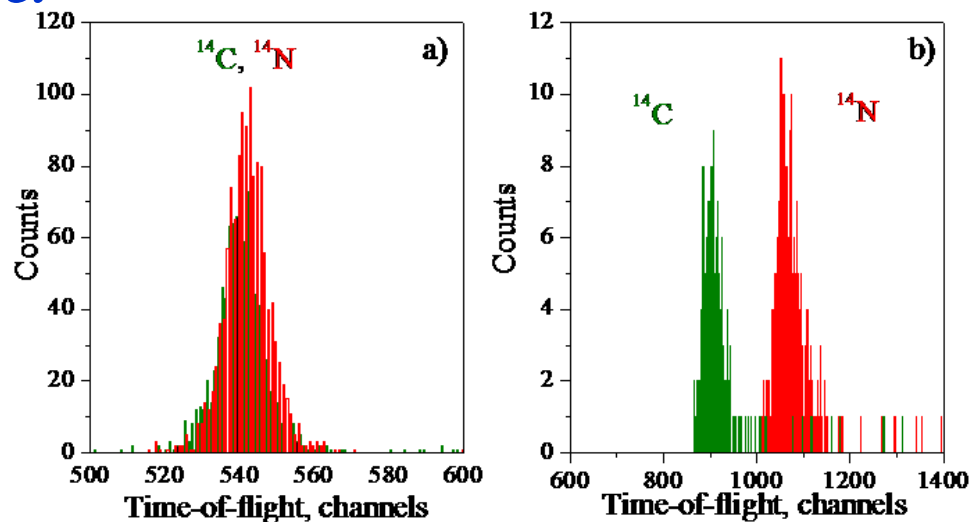
TOF spectrum for radiocarbon and nitrogen with (a) and without (b) passing through the 2.5 um Mylar film. For nitrogen observation at the exit of BINP AMS, the voltage at the 180⁰ electrostatic bend was reduced to 14/15 from the optimum for radiocarbon transit.
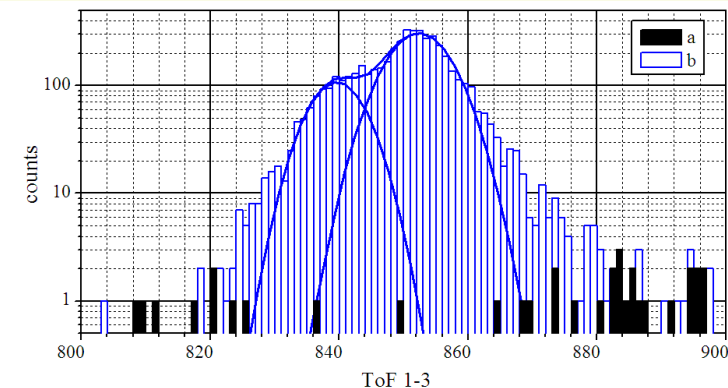
Nitrogen background with energy filter in high voltage terminal (black) and the emulation of absence of filter (blue).

Sample preparation

For AMS analysis, all natural samples must be converted to a universal form by sample preparation procedure. For these purposes, a sample is combusted in vacuum. Then the carbon from formed CO_2 gas catalytically deposited on iron powder. The Fe-C mixture is pressed in aluminum sample holder (cathode for ion source).

Sample preparation for the BINP AMS is carried out by chemists from LRMA NSU
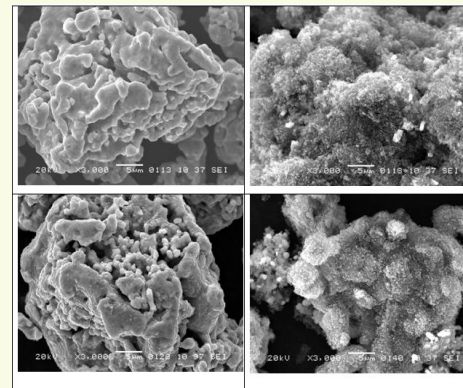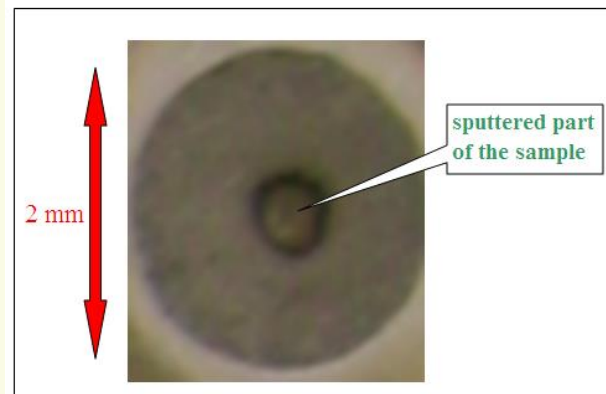

Carbon on the iron powder.

Graphitized samples - cathodes for ion source
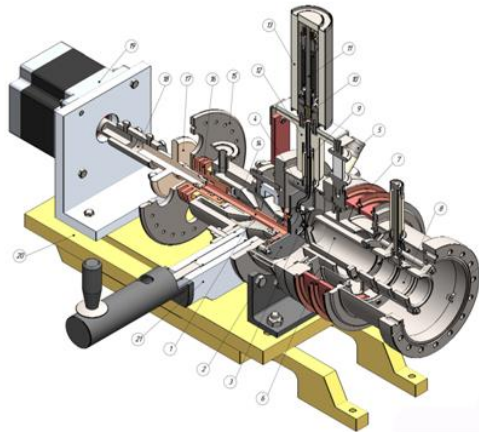
Ion source

Sample wheel for 23 samples
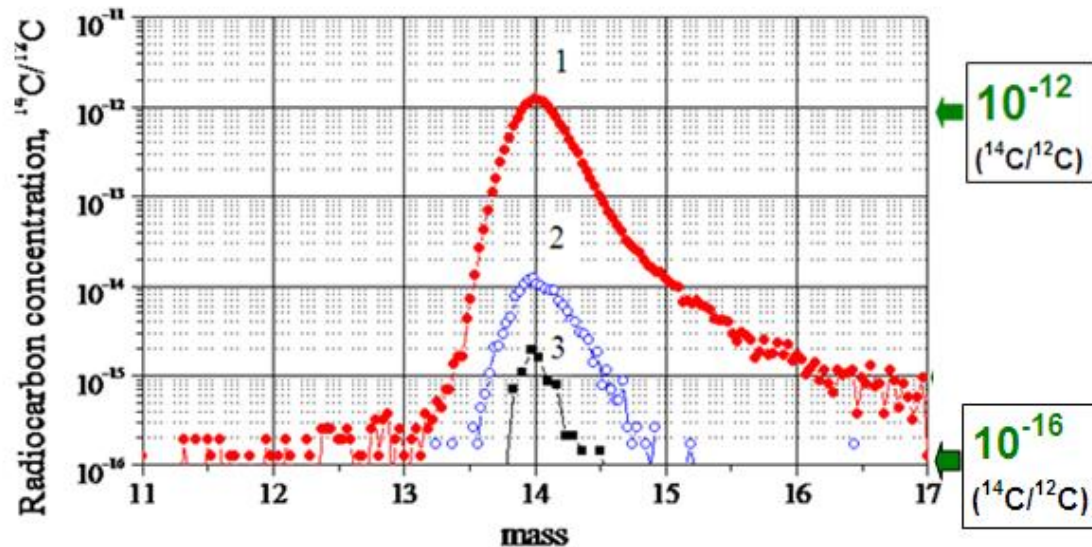

2 mm

sputtered part of the sample

The sputtering by Cs beam region of the sample is only about 0.5 mm in diameter.

Now at BINP AMS used two types of sample holders: with inner diameter of 2 mm (for about 3 mg of carbon sample) and with inner diameter of 1 mm (for 1 mg or less of carbon sample).

Atmospheric carbon is permeated into the samples during sample preparation. Samples used for radiocarbon dating must be handled carefully to avoid contamination. The contamination level in the samples during the sample preparation procedure is estimated by the radiocarbon content in graphite MPG ("dead" sample) with and without sample preparation.

The mass spectrums at the exit of BINP AMS for modern organic (1) and for "dead" samples with (2) and without (3) sample preparation procedure.

The content of radiocarbon atoms in graphite MPG without sample preparation is 15 orders of magnitude smaller than the content of carbon atoms. But the content of radiocarbon is increased by an order of magnitude after sample preparation. Typically, the quantity of pollution (radiocarbon concentration) composes approximately 1.5% of the concentration level in the modern samples.

To switch between the isotopes:

When measuring the concentration of radiocarbon in the samples, the switching algorithm is used. The isotope ^{14}C is detected by TOF telescope and ^{13}C currents are measured at the exit of AMS by shifted Faraday cup. For switching algorithm the high voltage of ion source is changed. The energy of the cesium ions remains constant. The electrostatic lens and correctors at the exit of the ion source are changed for each isotope. Thus, the passage of isotopes is carried out through a dipole magnet in low energy channel without changing the magnetic field. The magnetic field in high energy beam line magnet is not changed to, because the radial aperture is wide enough for passing radiocarbon ions to TOF detector and ^{13}C ions to shifted FC.
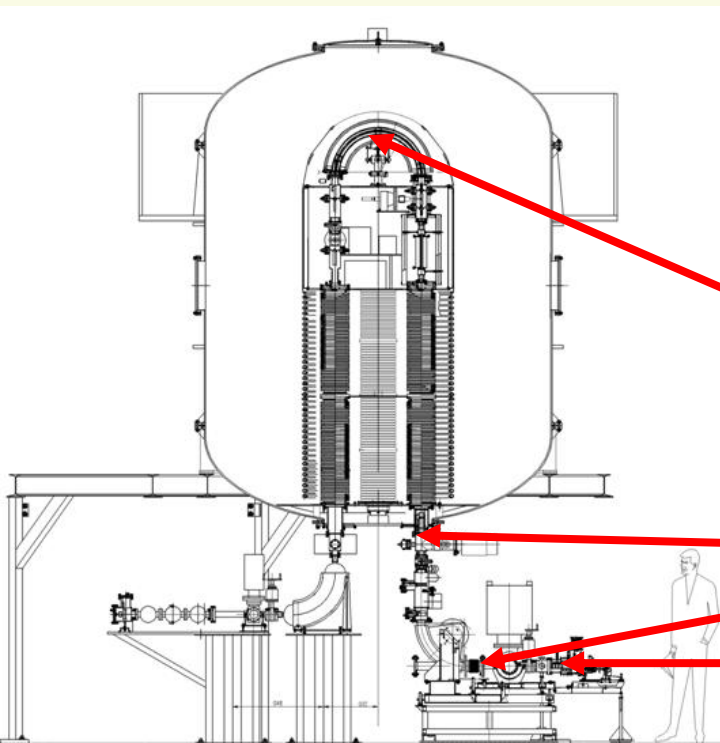
~ 80% of the time - ^{14}C counting

~ 15% of the time - isotope switching

+ ^{13}C current measuring

~ 5% of the time – sample wheel rotation

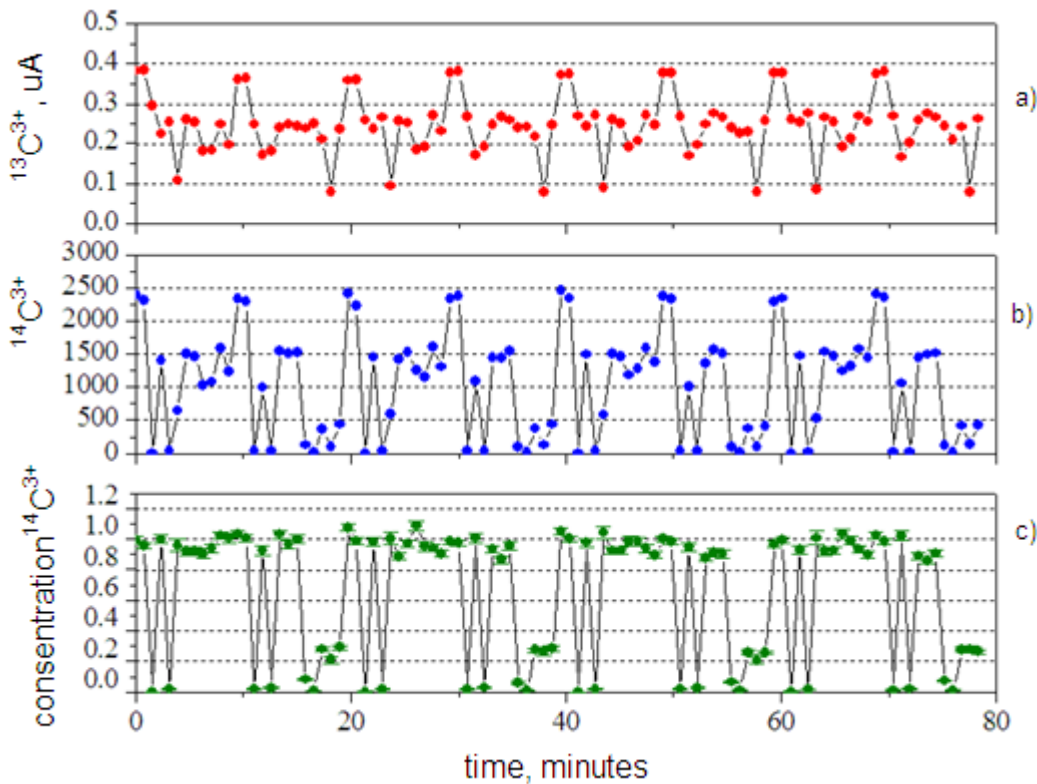| |
|---|
| The voltage of electrostatic bend in terminal is changed |
| The voltage of electrostatic lenses dipole correctors are changed |
| The high voltage of ion source is changed |

# Algorithm for measuring of the radiocarbon concentration on BINP AMS

$^{13}C^{3+}$ currents (a), radiocarbon counts $^{14}C^{3+}$ (b), and radiocorbon conctntration (c) depending from the time

For each sample, the ^{14}C ions are counted four times (10 seconds each) and twice the ^{13}C currents are measured for each 10 seconds counting. After that, the samples wheel is turned to the next sample for process repetition. Measuring of whole sample wheel (20 samples) takes about 15 minutes. For a set of statistics the wheel are moving to the second turn, third, etc. Typically, the measurement will take approximately 5 hours, with a statistical error of measurement for modern samples less than 1%.

An example of a radiocarbon analysis of samples

In order to increase the reliability of the samples dating, the samples are prepared and measured in pairs. The duplicating samples are circled. It is seen, that the measured values of the radiocarbon concentration in identical samples are statistically identical. The international standard reference materials OXI (SRM 4990 B) and OXII (SRM 4990 C) are used for calculation radiocarbon age of dated samples by normalization to samples with a known radiocarbon concentration.
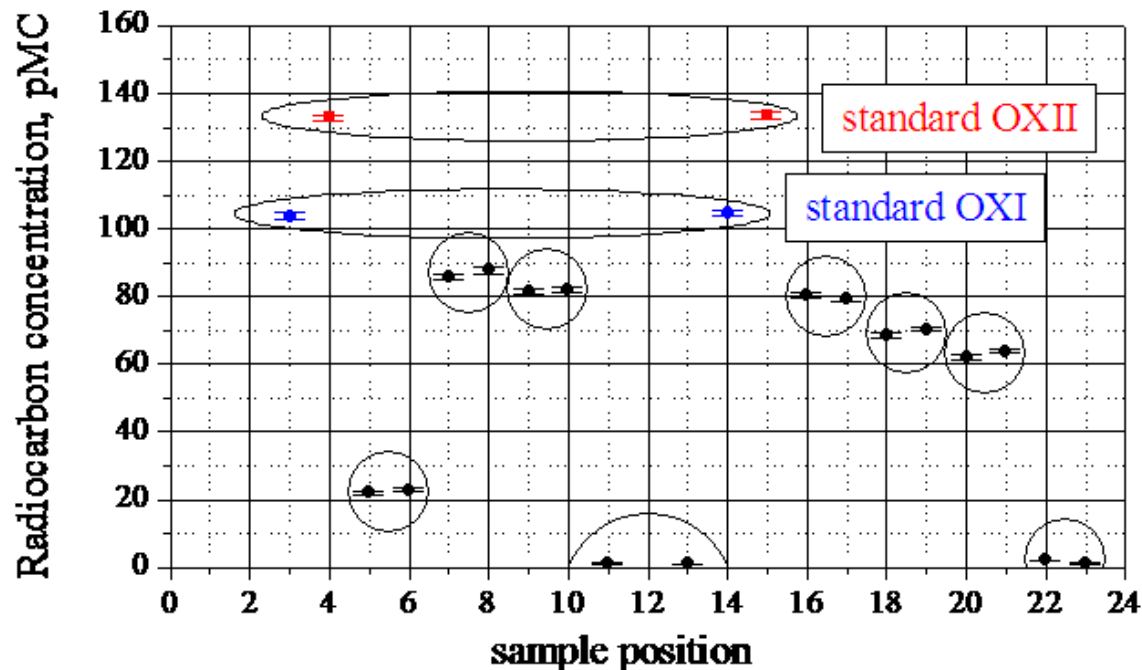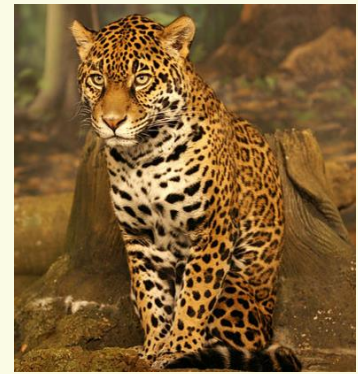
Old fauna of Russia dated by BINP AMS

15809 ± 138 years

19034 ± 165 years

781 ± 65 years

6865 ± 89 years

3813 ± 72 years

5281 ± 77 years

5190 ± 80 years

4775 ± 75 years

18141 ± 169 years

3161 ± 78 years

All the photos were taken from: ru.wikipedia.org

SUMMARY

• The BINP AMS is demonstrated the sustained performance on 1MV running, without electrical breakdowns of high voltage.

• The accelerator complex and algorithm of the radiocarbon analysis are described.

• The BINP AMS with additional ion selection properties is demonstrated the good radiocarbon ions identification.

• The statistical error of radiocarbon analysis for modern samples less than 1% when measuring 20 samples per 5 hours.

• The concentration of radiocarbon in the "dead" samples is approximately 10^{-14} ($^{14}C/^{12}C$) due to contamination by atmospheric carbon during sample preparation of natural samples.

• The BINB AMS is used for radiocarbon analysis of graphitized natural samples for users. About 1000 samples were measured last year for 25 user organizations.