Radionuclides in the Environment and the Arizona TAMS Facility

Dr. Dana L. Biddulph
Accelerator Mass Spectometry Lab
The NSF-Arizona Accelerator Mass Spectrometry (AMS) Laboratory has operated as an NSF research and service Facility since 1981. It is jointly operated by the Physics and Geosciences Departments at the University of Arizona, serving as an interdisciplinary hub for a broad range of research and educational activities. During the last five years, 76 students have utilized this facility for their research, leading to 32 Doctoral and 13 Master's degrees completed from various universities. The facility has two tandem accelerators with terminal potentials of up to 3 million volts that are used for measuring cosmogenic isotopes with ultra-low abundances, such as $^{14}$C, $^{10}$Be, $^{129}$I, and $^{26}$Al. These are used to investigate many research topics, including tracer studies, radiometric dating, carbon cycle dynamics, terrestrial magnetic field, solar wind, ocean sciences, cosmic ray physics, meteoritics, geology, paleoclimate, faunal extinctions, hydrologic balance, forest fire-frequency, archaeology, art history, forensic science, and instrument development.
Our Team

**Research Scientists**

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Why Study Radioisotopes?

- The study of radioisotopes can reveal information about:
  - Abundances of target nuclei
  - Pathways in nature
  - Changes in climate, ocean and atmospheric circulation patterns
  - Changes in Earth’s magnetic field
  - Changes in cosmic ray flux

- Certain isotopes can also be used in radiometric dating techniques
Production of Radioisotopes

- Fissiogenic
- Cosmogenic
- Anthropogenic
Radioisotopes Measured at Arizona

- $^{10}\text{Be}$, $t_{1/2} = 1.5 \times 10^6$ years
- $^{14}\text{C}$, $t_{1/2} = 5730$ years
- $^{26}\text{Al}$, $t_{1/2} = 7.3 \times 10^5$ years
- $^{129}\text{I}$, $t_{1/2} = 1.6 \times 10^7$ years
Radiocarbon Dating

- $^{14}$C dating can be used to date material that was living in the last few ten thousands of years and that received its carbon from the air.
- $^{14}$C is produced in the atmosphere.
Radiocarbon Dating

- The production rate of $^{14}$C in the atmosphere has been relatively constant for tens of thousands of years
  - $^{14}$C:$^{12}$C = 1:10$^{12}$
  - $^{14}$C decays via beta decay to $^{14}$N with $t_{1/2} = 5730$ years
  - When living, there is an equilibrium of $^{14}$C:$^{12}$C
  - When dead, the $^{14}$C decays away while the $^{12}$C doesn’t

- Measurement of the activity of $^{14}$C or the amount of $^{14}$C determines the object’s age
Figure 3. Increase of carbon-14 in the atmosphere due to atmospheric nuclear weapons testing in the period 1950-1963 AD. Subsequently, the level has declined to about 110% of the pre-bomb level, due to re-equilibration with the ocean.
$^{14}$C Dating Applications

- Archaeology
- Oceanography
- Forensics
- Meteorology
- Meteorites
- Paleoclimate
- Art History
$^{129}$I Applications

- Cosmogenically produced when thermal neutrons strike a Xenon target in the atmosphere
  - May be used to track changes in Earth’s magnetic field or cosmic ray flux
- Tracer studies from anthropogenic releases
- Solomon Islands
- Easter Island
Measurement of Radioisotopes

- Make an ion beam and separate the ions with magnetic and electric fields.
- First the sample under study must be cleaned and purified. This is done with a variety of chemical extraction techniques.
Sample Preparation for $^{14}$C Analysis

ABA Pretreatment
Combust sample in vacuum chamber with CuO
Collect liberated CO$_2$ gas
Reduce CO$_2$ with Zn in hot chamber
Attach C with Fe in vessel
Sample Preparation

- Carrier-free iodine extraction and AMS target preparation.
- Annual sample sizes roughly 30 g; iodine content 3 to 5 ppm.
- Clean samples with 10% acid dissolution and sonication.
- Rinse in distilled water, dissolve in 85% H₃PO₄ + H₂O.
- Oxidize I⁻ to I₂ with 2 drops 1 M NaNO₂, 5 to 10 ml CHCl₃ for iodine extraction.
- Drain CHCl₃ + I₂ into glass vial containing 10 to 15 mg 120 mesh Ag powder. Repeat extraction until fresh CHCl₃ remains clear.
- Allow to settle overnight, then evaporate CHCl₃.
- Rinse Ag + I₂ mixture three times with distilled water and dry.
- Press into Al cathode for AMS measurement.
Ion Source

Cesium sputter ion source – AMS

SNICS II Schematic Diagram
Ion Beam Analysis

- Rare isotope analysis not possible with just magnets
- For example, setting a magnet to pass $^{14}\text{C}^{-1}$ allows all atomic and molecular species with mass 14 and charge $-1$ through
  - This includes $^{14}\text{N}^{-1}$, $^{13}\text{CH}^{-1}$ and $^{12}\text{CH}_2^{-1}$
Solution: Break Molecules Apart

- Accelerate ions to high energy (a few MeV)
- Collide ions with low pressure gas (Ar)
- Electrons removed from ions to create positively charged ions
- Molecules with a net high positive charge are unstable and break up
Electrostatic Accelerator
Cockroft – Walton type accelerator shown here

High voltage is created by moving charge up a ladder of diodes.
Tandem Van de Graaf Accelerator
AMS Facility
\[ F = q(E + v \times B) = ma \]

For Magnetic Case Only, \( E = 0 \), and \( v \) and \( B \) are perpendicular

\[ F = qvB = ma = \frac{mv^2}{r} \text{ for a constant radius of curvature } r \]

Squaring and arranging terms,

\[ B^2r^2 = \frac{m^2v^2}{q^2} = 2\frac{mE}{q^2}, \text{ for kinetic energy } E \]

Therefore, magnetic analysis selects all ions with the same

\[ \frac{mE}{q^2} \]
\[ F = q(E + v \times B) = ma \]

For Electric Case Only, \( B = 0 \)

\[ F = qE = ma = \frac{mv^2}{r} \text{ for a constant radius of curvature } r \]

\[ E_r = \frac{mv^2}{q} = \frac{2E}{q}, \text{ for kinetic energy } E \]

Therefore, electrostatic analysis selects all ions with the same \( \frac{E}{q} \)

\[ E/q \]
Energy Calculations

Pre-acceleration energy
\[ E = qV \]
Nominally 60 keV

Post-acceleration energy
\[ E = qV = TV(1 + q) \]
Example: \( ^{14}C^{+3} \)
\[ E = 2.5\text{MeV}(1 + 3) = 10\text{MeV} \]
Scaling

Passing $^{13}\text{C}^+3$ through magnet, $B = 4190$ g
To pass $^{14}\text{C}^+3$, must change magnetic field

$$B^2)_{^{14}\text{C}+3} / B^2)_{^{13}\text{C}+3} = ME/q^2)_{^{14}\text{C}+3}/ME/q^2)_{^{13}\text{C}+3}$$

$$= 14E/q^2 / 13E/q^2$$

or $B)_{^{14}\text{C}+3} = \sqrt{14/13}B)_{^{13}\text{C}+3}$

$$= 1.038*4190 \text{ g} = 4348 \text{ g}$$
Now that the Molecules are Gone...

- Still not done!
- Interference from isobars and molecular fragments can still overwhelm signal if \( m/q = M/Q \)
  - Examples: \(^{10}\text{B}^+\text{2} \) and \(^{10}\text{Be}^+\text{2} \), \(^{7}\text{Li}^+\text{2} \) and \(^{14}\text{C}^+\text{4} \), \(^{97}\text{Mo}^+\text{3} \) and \(^{129}\text{I}^+\text{4} \)
- For these cases nuclear physics techniques can be employed
<table>
<thead>
<tr>
<th>Ion Charge</th>
<th>Transmission (%)</th>
<th>Interferences</th>
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<tbody>
<tr>
<td>+1</td>
<td>Not Measured</td>
<td>Molecular</td>
</tr>
<tr>
<td>+2</td>
<td>30</td>
<td>Molecular</td>
</tr>
<tr>
<td>+3</td>
<td>17</td>
<td>(^{43}\text{Ca}^{+1}, , ^{86}\text{Kr}^{+2}, , ^{86}\text{Sr}^{+2})</td>
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<tr>
<td>+4</td>
<td>7.8</td>
<td>(^{97}\text{Mo}^{+3})</td>
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<tr>
<td>+5</td>
<td>4.1</td>
<td>(^{103}\text{Rh}^{+4})</td>
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<tr>
<td>+6</td>
<td>2.1</td>
<td>(^{43}\text{Ca}^{+2}, , ^{86}\text{Kr}^{+4}, , ^{86}\text{Sr}^{+4})</td>
</tr>
<tr>
<td>+7</td>
<td>0.7</td>
<td>(^{37}\text{Cl}^{+2}, , ^{92}\text{Zr}^{+5}, , ^{92}\text{Mo}^{+5}, , ^{111}\text{Cd}^{+6})</td>
</tr>
</tbody>
</table>
surface barrier "dE/dx" detector

250 $\mu$g/cm$^2$
Mylar Foil

300 mm$^2$ area
100 micron thick
Silicon surface barrier detector
Gas ionization detector: $^{10}$Be detection
Questions?