Australian Facility For Noble-Gas Radio Isotope Measurements Using Atom Trap Trace Analysis

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A new facility for fast and accurate measurements of noble gas radio-nuclide ratios

$^{81}\text{Kr}/\text{Kr}$, $^{85}\text{Kr}/\text{Kr}$, $^{39}\text{Ar}/\text{Ar}$
Noble Gas Radio-Isotope Tracers

- Environmental markers to study ground and ocean water movement

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Half-Life (years)</th>
<th>Groundwater Age / Years</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{39}$Ar</td>
<td>$\tau_{1/2} = 269$</td>
<td>1000 - 1000000</td>
</tr>
<tr>
<td>$^{81}$Kr</td>
<td>$\tau_{1/2} = 229,000$</td>
<td>1000000 - 100000000</td>
</tr>
<tr>
<td>$^{14}$C</td>
<td>$\tau_{1/2} = 5730$</td>
<td>10000000 - 1000000000</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
**Noble Gas Radio-Isotope Tracers**

- Application to water dating:
  - $^{39}\text{Ar}$, $^{81}\text{Kr}$, $^{85}\text{Kr}^+$ are cosmogenic nuclides
  - Nearly ideal physical & chemical characteristics (e.g. unreactive)
  - Well defined input function at water/atmosphere interface

$^{85}\text{Kr}$ atmospheric concentration is primarily due to nuclear fuel reprocessing

*Picture credit: Axel Suckow, CSIRO*
Noble Gas Radio-Isotope Tracers

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  - Well defined input function at water/atmosphere interface

- Rarely used due measurement difficulty

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Picture credit: Axel Suckow, CSIRO
Noble Gas Radio-Isotope Tracers

- Environmental markers to study ground and ocean water movement

- Noble-gas mass spectrometry
- Gas chromatography
- Radiometry
- Accelerator mass spectrometry
- Atom trap trace analysis

<table>
<thead>
<tr>
<th>Noble Gas Tracer</th>
<th>Groundwater Age (Years)</th>
</tr>
</thead>
</table>
| $^3$He/T$^3$He | $^{81}$Kr: $\tau_{1/2} = 10.8$ years $^{39}$Ar: $\tau_{1/2} = 269$ years $^{85}$Kr: $\tau_{1/2} = 10.8$ years $^{14}$C: $\tau_{1/2} = 5730$ years $^{36}$Cl: $\tau_{1/2} = 100000$ years
| CFC/SF$_6$ | $^4$He (estimates) $^{40}$Ar

16th September 2016
AUSTRALIAN FACILITY FOR NOBLE-GAS RADIO ISOTOPE MEASUREMENTS – INPC 2016
Radio-Isotope Measurements

- Measurements difficult due to small atmospheric abundance:
  - $^{85}\text{Kr}/\text{Kr}: 2\times 10^{-11}$ $^{81}\text{Kr}/\text{Kr}: 5.2\times 10^{-13}$ $^{39}\text{Ar}/\text{Ar}: 8\times 10^{-16}$
  - 1L of surface water contains just $\sim 9000$ $^{39}\text{Ar}$ atoms & $\sim 1500$ $^{81}\text{Kr}$ atoms
  - Need to measure ratio to $\sim 1\%$ of atmospheric abundance in water dating applications

- Low-level decay counting (LLC) is traditional measurement technique
  - Large water samples required (2000-5000 litres for $^{39}\text{Ar}$)
  - Long measurement times (8-60 days for $^{39}\text{Ar}$)
  - Performed where a low background count is present
    - e.g. University of Bern, 35m below ground
Atom Trap Trace Analysis (ATTA)

- Laser-based technique for measuring noble-gas radio-isotope ratios
- Atom-counting rather than decay-counting ➞ smaller samples, faster measurement
- Lasers used to cool specific isotope and hold in a magneto-optical trap
- Shifts in energy levels between isotopes permit selection of a single isotope by tuning laser frequencies

Rohan Glover, PhD Thesis, Griffith University
Atom Trap Trace Analysis

- Based on established laser cooling and magneto-optical trapping

Atom Trap Trace Analysis

- Current ATTA systems worldwide:
  - Argonne National Laboratory, IL, USA
    - Predominantly focussed on Kr measurements
    - Only facility currently open for general samples
    - Throughput ~100 samples / year
  - University of Heidelberg, Germany
    - Initial $^{39}$Ar measurements demonstrated
  - University of Science and Technology of China, Hefei, China
    - Developing Kr capability
Performance

- Initial facility will offer ATTA measurements with performance comparable to existing labs

- Upgraded facility will offer reduced sample size and measurement time

<table>
<thead>
<tr>
<th></th>
<th>$^{39}$Ar</th>
<th>$^{81}$Kr</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ml-STP</td>
<td>Water / L</td>
<td>Time</td>
</tr>
<tr>
<td>AMS</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>LLC</td>
<td>~700</td>
<td>~3000</td>
<td>8-60 days</td>
</tr>
<tr>
<td>ATTA Facility - initial</td>
<td>1</td>
<td>5</td>
<td>~12 hours</td>
</tr>
<tr>
<td>ATTA Facility - upgraded</td>
<td>0.1</td>
<td>0.5</td>
<td>~5 hours</td>
</tr>
</tbody>
</table>
Sample Preparation

- ATTA measurement requires small gas-phase noble-gas sample
  - Mixed noble-gas samples are not a problem due to selectivity of atom-trapping

- CSIRO’s Environmental Tracer and Noble Gas Laboratory in Adelaide can
  - Extract noble gases from water samples
  - Extract gas from water in the field, and separate out noble-gas components

Picture credit: Arne Kersting, Axel Suckow, CSIRO
Timeline

- **Q3 2016**: Construction commenced at University of Adelaide and Griffith University.
- **Q4 2017**: Initial uncalibrated measurements available in Adelaide.
- **Q3 2018**: Fully calibrated measurements – agreement with low-level counting.
- **Q2 2019**: Laser excitation upgrade installation – smaller sample size.
- **Q4 2019**: Fully calibrated measurements – increased sample throughput.
Australian Facility For Noble-Gas Radio Isotope Measurements

- Fast, accurate measurements of $^{81}$Kr, $^{85}$Kr and $^{39}$Ar radio-nuclide ratios
- Small sample size requirements (isotope dependent)
- Expected to commence sample measurement late 2017
- Funded through per-sample analysis fee

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