New Hydrological Age-Dating techniques using cosmogenic radionuclides Beryllium-7 and Sodium-22

S. Frey, C. Kuells & C. Schlosser
Why new age-dating methods?

- Tritium levels close to natural background
- $^3$H/$^3$He, $^{85}$Kr, SF$_6$, CFC etc. only if no contact to atmosphere

[Happell et al. (2004), Tellus B]
Radionuclide Production

- Produced by spallation processes in upper atmosphere
- $^7$Be about four times higher than $^{22}$Na

Surfaces of a constant $^7$Be production rate (nuclei min$^{-1}$ m$^{-3}$, at standard temperature and pressure).

[Lal & Peters (1967), Handbuch der Physik]
Sorption of Sodium

[Birkholz 2007, unpublished]
Sorption of Beryllium

- About 3 – 10 % of beryllium input is not affected by sorption
- Recovery indicates mobilization mechanism

[Data from Hohwieler 2005, unpublished]
Sampling

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Chemical Separation

1. Eluting the resin
2. Evaporating to dryness → weighting
3. Dissolving in 0.1M HCl
4. Adjusting pH to 8 – 9 (NH₄OH)
5. Adding (NH₄)₂CO₃ + EtOH
6. Centrifuging
7. Evaporating to dryness (500°C)
8. Dissolving in 0.1M HCl
9. Adding NaTPhB (Sodium-Tetra-Phenyl-Borate)
10. Filtering and washing
11. Evaporate to dryness

γ-Spectroscopy

[Sakaguchi et al. 2003, J. Radioanal. Nucl. Chem.]
Why separating potassium?

Compton scattering masks exact measurements of the 1275 MeV-peak of $^{22}\text{Na}$
Case Study in the Black Forest

Dreisam Catchment
- Area: ~ 258 km²
- Precipitation: ~ 1500 mm/a
- Evaporation: ~ 600 mm/a
- Discharge: ~ 820 mm/a
- Groundwater Discharge: ~ 60 mm/a

Sampling locations
- Two Groundwater wells (334 m & 1284 m a.s.l.)
- Two Rivers (316 m & 434 m a.s.l.)
- One Precipitation Station (melted snow, 277 m a.s.l.)
Results

### Activities and resulting ages of Sodium-22 measurements

<table>
<thead>
<tr>
<th>Sample</th>
<th>Date</th>
<th>Error [%]</th>
<th>activity [mBq/l]</th>
<th>age [d] related to Literature*</th>
<th>Data obtained</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schauinsland</td>
<td>12/01/09</td>
<td>NA</td>
<td>&lt; 0.04</td>
<td>&gt; 2763</td>
<td>&gt; 37</td>
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<tr>
<td>Brugga</td>
<td>12/14/09</td>
<td>NA</td>
<td>&lt; 0.03</td>
<td>&gt; 3066</td>
<td>&gt; 340</td>
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<tr>
<td>Hungerbrunnen</td>
<td>01/14/10</td>
<td>NA</td>
<td>&lt; 0.04</td>
<td>&gt; 2847</td>
<td>&gt; 121</td>
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<tr>
<td>Snow</td>
<td>01/21/10</td>
<td>19.3</td>
<td>0.04</td>
<td>2726</td>
<td>0</td>
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<tr>
<td>Dreisam</td>
<td>02/04/10</td>
<td>NA</td>
<td>&lt; 0.07</td>
<td>&gt; 1974</td>
<td>0</td>
</tr>
</tbody>
</table>

### Activities and resulting ages of Beryllium-7 measurements

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<th>Sample</th>
<th>Date</th>
<th>Error [%]</th>
<th>activity [mBq/l]</th>
<th>age [d] related to Literature**</th>
<th>Data obtained</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schauinsland</td>
<td>12/01/09</td>
<td>NA</td>
<td>&lt; 0.12</td>
<td>&gt; 555</td>
<td>&gt; 518</td>
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<tr>
<td>Brugga</td>
<td>12/14/09</td>
<td>7.2</td>
<td>1.66</td>
<td>360</td>
<td>323</td>
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<tr>
<td>Hungerbrunnen</td>
<td>01/14/10</td>
<td>24.4</td>
<td>0.22</td>
<td>512</td>
<td>475</td>
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<tr>
<td>Snow</td>
<td>01/21/10</td>
<td>5.9</td>
<td>122.80</td>
<td>209</td>
<td>0</td>
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<tr>
<td>Dreisam</td>
<td>02/04/10</td>
<td>6.0</td>
<td>13.64</td>
<td>201</td>
<td>165</td>
</tr>
</tbody>
</table>

* 0.1 – 0.3 mBq/l (Tokuyama & Igarashi (1998), J. Environ. Radioactivity)
** 1 – 2 Bq/l (Knies et al. (1994), Nucl. Instr. and Meth. in Phys.)
Summary

• Sodium-22 only detected in snow
  Possible explanation:
  Exchange resin, chemical separation, detector (HPGe 30% Efficiency),
  Sodium-Sodium-Exchange

• Beryllium-7 in all samples but ‘Schauinsland’
  Possible explanation:
  Age, sorption processes

• Dated ages match previously observed ages (Uhlenbrook et al. (2002), WRR):
  • Shallow groundwater: 2 – 3 years
  • Deep groundwater: younger than 10 years
Discussion

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<th>Disadvantages</th>
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<tbody>
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<td>^{22}\text{Na} provides an alternative to Tritium, if combined with ^{7}\text{Be} even younger waters detectable</td>
<td>Long time for chemical analysis</td>
</tr>
<tr>
<td>No gaseous tracers</td>
<td>Detection limit (esp. ^{22}\text{Na})</td>
</tr>
<tr>
<td></td>
<td>Large water samples needed (but small compared to ^{85}\text{Kr})</td>
</tr>
</tbody>
</table>

Thank you for your attention


Sakaguchi et al. 2003: Low-level measurement of the cosmogenic $^{22}$Na radionuclide in fresh water by ultra low-background gamma-ray spectrometry after simple radiochemical separation *J. Radioanal. Nucl. Chem* 258, pp. 101-105.

Tokuyama & Igarashi 1998: Seasonal variation in the environmental background level of cosmic-ray-produced $^{22}$Na at Fukui City, Japan. *J. Environ. Radioactivity* 38, pp. 147 - 161.