Direct method for the quantitative analysis of surface contamination on ultra-low background materials from exposure to dust

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PNNL is operated by Battelle for the U.S. Department of Energy
Ultrasensitive detectors

- Ultralow background
  - Underground facilities
  - Detector shielding
  - Ultraipure materials
Major contributors to material radioactive background

Uranium-238

Thorium-232

Potassium-40

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Material selection: a critical challenging task

- Radiopurity requirements
  - μBq/kg range or lower

- Extensive assay campaigns
  - Selection of the most-radiopure materials

- Ultrasensitive analytical techniques

- Ultraclean analytical procedures and material handling

For reference:
1 ppt Th = 4.1 μBq $^{232}$Th/kg
1 ppt U = 12.4 μBq $^{238}$U/kg
1 ppb $^{nat}$K = 30.5 μBq $^{40}$K/kg
A dedicated facility at PNNL

- Class 10,000 cleanroom
- 2 dedicated mass spectrometers (ICP-QQQ-MS)
- Cutting edge sample digestion techniques
- Only Optima grade reagents
- Low background PFA labware
- Automated system for analyte separation via extraction chromatography
# Current detection limits for different materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Th/U Detection Limits</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>µBq/kg</td>
</tr>
<tr>
<td>Copper†</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>(Electroformed or commercial OFHC)</td>
<td></td>
</tr>
<tr>
<td>Lead†</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Titanium</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Polymers‡</td>
<td>&lt;1</td>
</tr>
<tr>
<td>(PTFE, PVDF, “Acrylic”, Kapton*, etc.)</td>
<td></td>
</tr>
<tr>
<td>Linear Alkyl Benzene (LAB)</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>Quartz, Fused Silica</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Electronic Components (FETs, resistors, thermocouples, etc.)</td>
<td>&lt;0.1 pg/piece</td>
</tr>
<tr>
<td>Solutions</td>
<td>&lt;0.01</td>
</tr>
</tbody>
</table>

Dust particulate: a significant contribution to material surface contamination

- High purity materials
  - Concerning (even in cleanrooms!)

- Ongoing efforts to estimate backgrounds from dust, mainly from
  - Fallout *models*
  - Assumed dust composition

- Dust in cleanrooms = local soil \(\leftarrow\) *Not necessarily!*
  - Generated by handled materials and ongoing activities
Direct method for quantitative analysis

- Exposure of a dust collection media
- Dissolution of deposited contamination
- Analysis via Inductively Coupled Plasma Mass Spectrometry (ICP-MS, long-lived radionuclides and stable elements)

M.L. di Vacri, I.J. Arnquist, S. Scorza et al., Direct method for the quantitative analysis of surface contamination on ultra-low background materials from exposure to dust, Nuclear Inst. And Methods in Physics Research, A 994 (2021) 165051
Exposure in a class 10,000 cleanroom at PNNL

<table>
<thead>
<tr>
<th></th>
<th>PFA vial [ng·day⁻¹·cm⁻²]</th>
<th>Si coupon [ng·day⁻¹·cm⁻²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>K-nat</td>
<td>(1.1 ± 0.2)x10⁻³</td>
<td>(5.1 ± 1.6)x10⁻⁴</td>
</tr>
<tr>
<td>Pb-stable</td>
<td>(1.2 ± 0.2)x10⁻⁵</td>
<td>(1.1 ± 0.5)x10⁻⁵</td>
</tr>
<tr>
<td>Th-232</td>
<td>(1.8 ± 0.4)x10⁻⁸</td>
<td>(5 ± 4)x10⁻⁸</td>
</tr>
<tr>
<td>U-238</td>
<td>(1.7 ± 0.6)x10⁻⁸</td>
<td>(3 ± 2)x10⁻⁸</td>
</tr>
</tbody>
</table>

Exposure: ca. 30 days

K ~ 1.4 E+4 ppm
Th ~ 7.2 ppm
U ~ 1.2 ppm

In terms of radioactivity

\[
\begin{align*}
\text{n}^\text{at}K &= 30.5 \ \text{Bq} \ 40^\text{K} \cdot \text{g}^{-1} \\
\text{n}^\text{at}Pb &= 0.2 \ \text{Bq} \ 210^\text{Pb} \cdot \text{g}^{-1} \\
232^{\text{Th}} &= 4.1 \ \text{kBq} \cdot \text{g}^{-1} \\
238^{\text{U}} &= 12.2 \ \text{kBq} \cdot \text{g}^{-1}
\end{align*}
\]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>PFA vial [(\mu\text{Bq} \cdot \text{day}^{-1} \cdot \text{cm}^{-2})]</th>
<th>Si coupon [(\mu\text{Bq} \cdot \text{day}^{-1} \cdot \text{cm}^{-2})]</th>
</tr>
</thead>
<tbody>
<tr>
<td>K-40</td>
<td>((3.3 \pm 0.5) \times 10^{-5})</td>
<td>((1.5 \pm 0.5) \times 10^{-5})</td>
</tr>
<tr>
<td>Pb-210</td>
<td>((2.4 \pm 0.4) \times 10^{-9})</td>
<td>((2.1 \pm 0.9) \times 10^{-9})</td>
</tr>
<tr>
<td>Th-232</td>
<td>((7.6 \pm 1.5) \times 10^{-8})</td>
<td>((2.0 \pm 1.8) \times 10^{-7})</td>
</tr>
<tr>
<td>U-238</td>
<td>((2.1 \pm 0.7) \times 10^{-7})</td>
<td>((3.1 \pm 2.8) \times 10^{-7})</td>
</tr>
</tbody>
</table>

Cleanroom vs non-cleanroom

Exposure of 8 PFA vial sets for 30 days

- Exposure also performed in a Class 10 laminar flow hood at PNNL

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Office space [ng·day⁻¹·cm⁻²]</th>
<th>Cleanroom [ng·day⁻¹·cm⁻²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th-232</td>
<td>(1.03 ± 0.06)x10⁻⁵</td>
<td>(7.5 ± 1.5)x10⁻⁸</td>
</tr>
<tr>
<td>U-238</td>
<td>(2.7 ± 0.4)x10⁻⁵</td>
<td>(2.1 ± 0.7)x10⁻⁷</td>
</tr>
</tbody>
</table>
Campaign of measurements at SNOLAB

- Excavated 6,800 ft underground
- In the working Creighton nickel mine, operated by Vale Ltd.
- A ca. 54,000 sq ft class-2,000 cleanroom hosting neutrino and dark matter experiments
Selected locations

- 9 underground cleanroom locations
- 1 underground non-cleanroom location
- 1 non-cleanroom location in the surface building

- A: South Drift LBL.
- B: Rm 127.
- C: Drift F.
- D: Rm 141.
- E: SNO+ control room.
- F: Rm 104.
- G: Rm 123.
- I: Drift F/J.
- J: Rm 132.
- K: Rm 137.
Results
(reduced lab activities and mine shutdown)

<table>
<thead>
<tr>
<th>Cleanrooms*</th>
<th>[µBq·day⁻¹·cm⁻²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>AVG ± SD</td>
<td></td>
</tr>
<tr>
<td>K-40</td>
<td>(5.7 ± 5.9)x10⁻⁵</td>
</tr>
<tr>
<td>Pb-210</td>
<td>(5.7 ± 13)x10⁻⁸</td>
</tr>
<tr>
<td>Th-232</td>
<td>(6.3 ± 7.8)x10⁻⁷</td>
</tr>
<tr>
<td>U-238</td>
<td>(6.5 ± 4.4)x10⁻⁷</td>
</tr>
</tbody>
</table>

* Excluding 3 cleanroom locations where activities may have triggered higher accumulation rates (empty circle markers)

• Results reflected local activities
XRF-based method for dust fallout monitoring

- System of witness plates at SNOLAB
- X-Ray Fluorescence (XRF) analysis
  - Surrogate elements (i.e., Fe and Ca)
- Assumption: rock/concrete sole dust source
- Deposition rates for other elements (e.g., Th and U) estimated based on relative content in rock/concrete

<table>
<thead>
<tr>
<th>Element</th>
<th>Rock (avg) [ppm]</th>
<th>Concrete [ppm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>$1.0 \times 10^4$</td>
<td>$1.6 \times 10^4$</td>
</tr>
<tr>
<td>Ca</td>
<td>$3.6 \times 10^4$</td>
<td>$1.0 \times 10^5$</td>
</tr>
<tr>
<td>Fe</td>
<td>$6.5 \times 10^4$</td>
<td>$2.6 \times 10^4$</td>
</tr>
<tr>
<td>Pb</td>
<td>10.4</td>
<td>13.9</td>
</tr>
<tr>
<td>Th</td>
<td>5.4</td>
<td>13.1</td>
</tr>
<tr>
<td>U</td>
<td>1.2</td>
<td>2.4</td>
</tr>
</tbody>
</table>

Composition of rock and concrete at the SNOLAB site*

Comparison: ICP-MS and XRF results

- Same locations
- Same exposure time

Iron

Calcium

*Upper limits
Estimated/Measured ratios of fallout rates

Potassium

Thorium

Lead

Uranium
Conclusions and perspectives

- Effective method for the **direct measurement** of dust contribution to material radioactive background
  - Discrepancy directly measured/inferred fallout rates in cleanrooms

- Relatively inexpensive, practical collection media

- ICP-MS technique:
  - **Multi-elemental**
    - Potentially all the elements in the periodic table, elemental fingerprint to dust sources
  - **Ultrasensitive**: relatively short exposure

- New campaigns ongoing/planned
  - Different lab activity and lab conditions
  - Targeting a larger number of elements (local major contributor to dust)
  - Interest within the ultralow background community

- Quantitatively assess the efficiency of material cleaning procedures
Thank you

- DOE, USA Office of High Energy Physics Advanced Technology R&D subprogram (KA-25)
- Canada Foundation for Innovation
- Province of Ontario Ministry of Research and Innovation
Back up slides
Table 1
ICP-MS detection limits for K, Ca, Fe, Pb, Th and U, measured as 3-StdDev of n = 3 PBs, for each analyte.

<table>
<thead>
<tr>
<th>Analyte</th>
<th>DL</th>
</tr>
</thead>
<tbody>
<tr>
<td>K [fg·g⁻¹]</td>
<td>30.0</td>
</tr>
<tr>
<td>Ca [pg·g⁻¹]</td>
<td>0.82</td>
</tr>
<tr>
<td>Fe [pg·g⁻¹]</td>
<td>0.66</td>
</tr>
<tr>
<td>Pb [fg·g⁻¹]</td>
<td>70.8</td>
</tr>
<tr>
<td>Th [fg·g⁻¹]</td>
<td>0.61</td>
</tr>
<tr>
<td>U [fg·g⁻¹]</td>
<td>0.79</td>
</tr>
</tbody>
</table>
Material assay: analytical techniques

- High Purity Germanium (HPGe) gamma spectroscopy
  - \( \approx 100 \ \mu\text{Bq} \cdot \text{kg}^{-1} \) - \( \text{mBq} \cdot \text{kg}^{-1} \) sensitivity

- Neutron Activation Analysis (NAA)
  - \( \approx \mu\text{Bq} \cdot \text{kg}^{-1} \) sensitivity

- Inductively Coupled Plasma Mass Spectrometry (ICP-MS)
  - \( \approx \mu\text{Bq} \cdot \text{kg}^{-1} \) sensitivity
Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

- Fast, quantitative
- Surface and bulk
ICP-QQQ-MS