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

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Maria Laura di Vacri¹, Isaac Arnquist¹, Silvia Scorza², Eric Hoppe¹, Jeter Hall²

¹ Pacific Northwest National Laboratory ² SNOLAB
Ultrasensitive detectors

- Ultralow background
  - Underground facilities
  - Detector shielding
  - Ultrapure 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
  - $\mu$Bq/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 $\mu$Bq $^{232}$Th/kg
1 ppt U = 12.4 $\mu$Bq $^{238}$U/kg
1 ppb $^{40}$K = 30.5 $\mu$Bq $^{40}$K/kg

https://pubs.usgs.gov/of/2005/1413/maps.htm
A dedicated facility at PNNL

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

| Material                                    | Th/U Detection Limits |
|---------------------------------------------|-----------------------|
|                                             | µBq/kg | ppt       |
| Copper†                                      | <0.1   | <0.01     |
| (Electroformed or commercial OFHC)          |        |           |
| Lead†                                       | <1     | <0.1      |
| Titanium                                    | <1     | <0.1      |
| Stainless Steel                             | <1     | <0.1      |
| Polymers‡                                    | <1     | <0.1      |
| (PTFE, PVDF, “Acrylic”, Kapton*, etc.)      |        |           |
| Linear Alkyl Benzene (LAB)                  | <0.1   | <0.01     |
| Quartz, Fused Silica                        | <1     | <0.1      |
| Electronic Components                       | <0.1 pg/piece | <1 nanoBq/piece |
| (FETs, resistors, thermocouples, etc.)      |        |           |
| Solutions                                   | <0.01  | <0.001    |

†I.J. Arnquist, M.L. di Vacri, E.W. Hoppe. NIM A (2020)
‡I.J. Arnquist, J.W. Grate, M. Bliss, E.W. Hoppe. Analytical Chemistry (2017)
*I.J. Arnquist, C. Beck, M.L. di Vacri et al., Nuclear Inst. And Methods in Physics Research, A 959 (2020)
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 ← 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

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

Exposure: ca. 30 days

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

* https://pubs.usgs.gov/of/2005/1413/maps.htm
In terms of radioactivity

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

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

* A. Alessandrello et al./Nucl. Instr. And Meth. In Phys. Res. B 142 (1998) 163-172
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

| Isotope | Office space [ng·day⁻¹·cm⁻²] | Cleanroom [ng·day⁻¹·cm⁻²] |
|---------|-----------------------------|---------------------------|
| Th-232  | (1.03 ± 0.06)x10⁻⁵          | (7.5 ± 1.5)x10⁻⁸          |
| U-238   | (2.7 ± 0.4)x10⁻⁵           | (2.1 ± 0.7)x10⁻⁷          |
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.

Map of the underground facility: investigated locations tagged with a letter and a star
Results
(reduced lab activities and mine shutdown)

| Isotope | 2000-class cleanroom UG | non cleanroom UG | non cleanroom Surface |
|---------|-------------------------|------------------|-----------------------|
| K-40    | (5.7 ± 5.9)x10^{-5}     |                  |                       |
| Pb-210  | (5.7 ± 13)x10^{-8}      |                  |                       |
| Th-232  | (6.3 ± 7.8)x10^{-7}     |                  |                       |
| U-238   | (6.5 ± 4.4)x10^{-7}     |                  |                       |

* 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

| Element | Rock (avg) [ppm] | Concrete [ppm] |
|---------|-----------------|----------------|
| K       | 1.0x10^4        | 1.6x10^4       |
| Ca      | 3.6x10^4        | 1.0x10^5       |
| Fe      | 6.5x10^4        | 2.6x10^4       |
| Pb      | 10.4            | 13.9           |
| Th      | 5.4             | 13.1           |
| U       | 1.2             | 2.4            |

Composition of rock and concrete at the SNOLAB site*

* From: I.T Lawson, “Analysis of Rock Samples from the New Laboratory”, STR-2007-003 SNOLAB Technical Report 2007
Comparison: ICP-MS and XRF results

- Same locations
- Same exposure time

Iron

Calcium

*Upper limits
Estimated/Measured ratios of fallout rates

Potassium

Lead

Thorium

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
| Analyte | DL     |
|---------|--------|
| K [fg·g⁻¹] | 30.0  |
| Ca [pg·g⁻¹] | 0.82  |
| Fe [pg·g⁻¹] | 0.66  |
| Pb [fg·g⁻¹] | 70.8  |
| Th [fg·g⁻¹] | 0.61  |
| U [fg·g⁻¹] | 0.79  |
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