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Studies of bulk ^{210}Po/^{210}Pb contamination in high purity copper for low background detectors

Presented by T. Mróz

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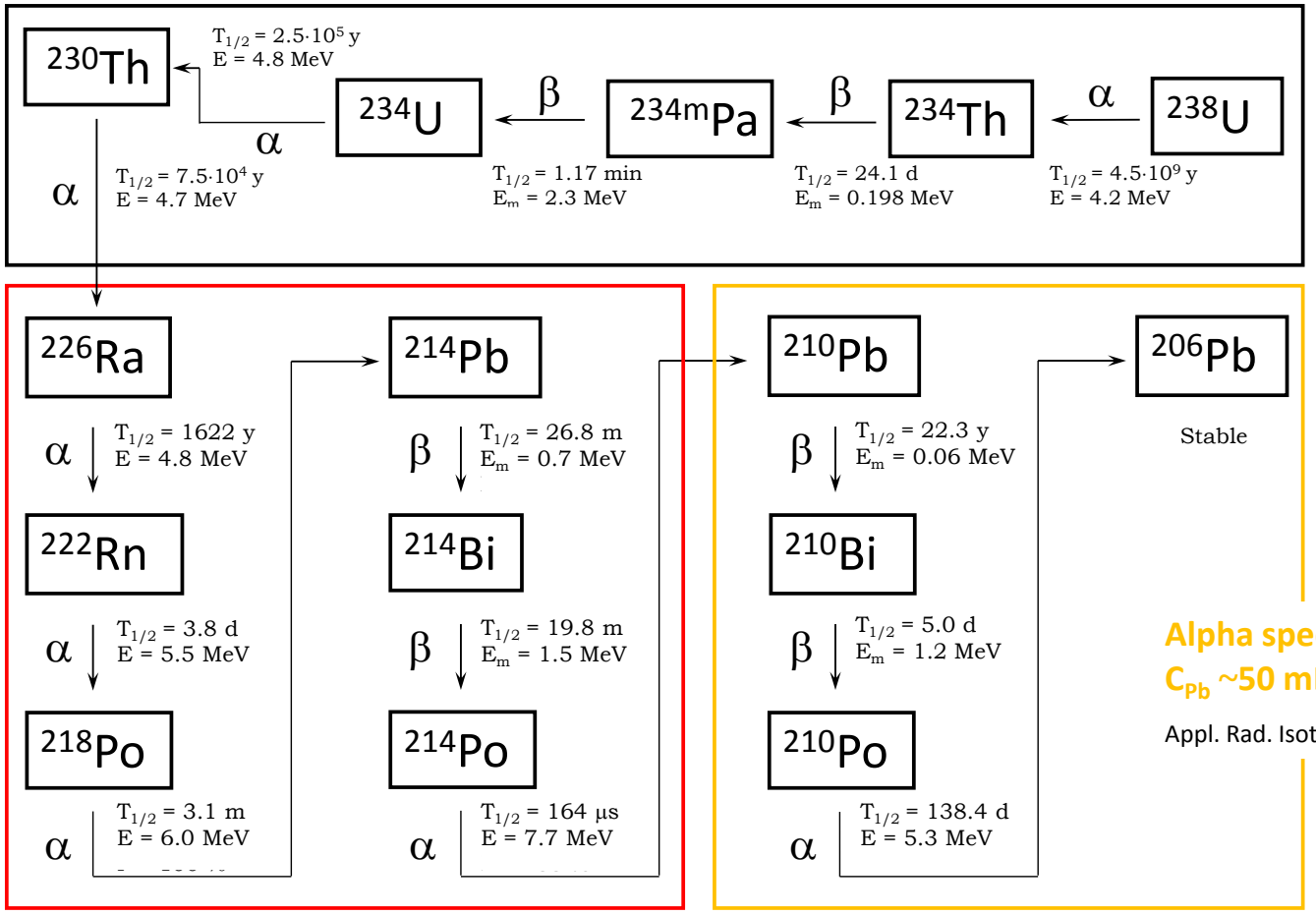
Potential disequilibrium in ^{238}U chain



ICP-MS:
$C_{U/Th}$ < 0.1 μBq/kg

NIM A 775 (2015) 93

HPGe:
C_{Ra} ~10 μBq/kg

Appl. Rad. Isot.
53 (2000) 191
Astrop. Phys.
18 (2002) 1

Alpha spectrometry
C_{Pb} ~50 mBq/kg

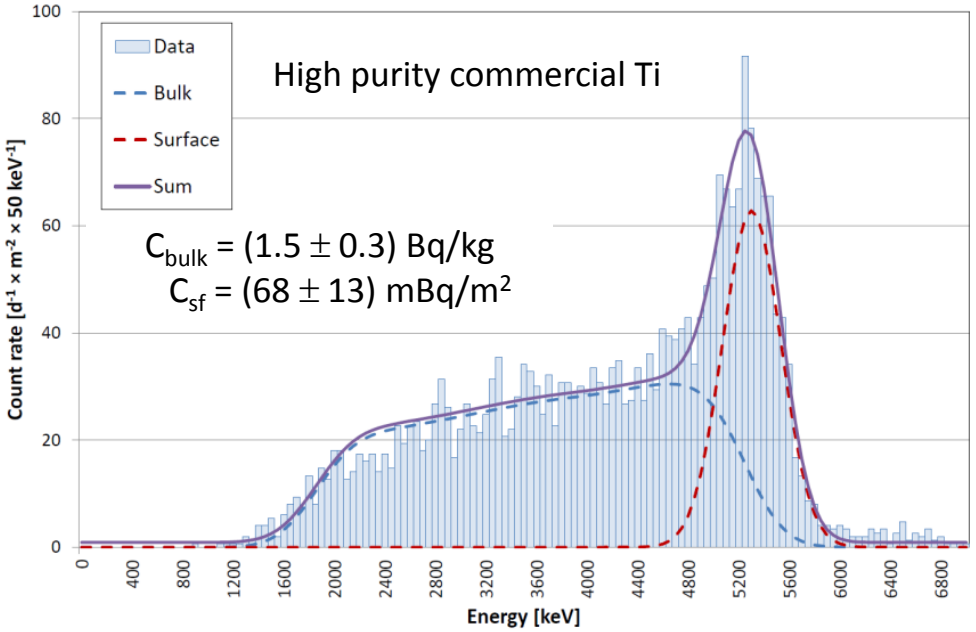
Appl. Rad. Isot. 126 (2017) 165

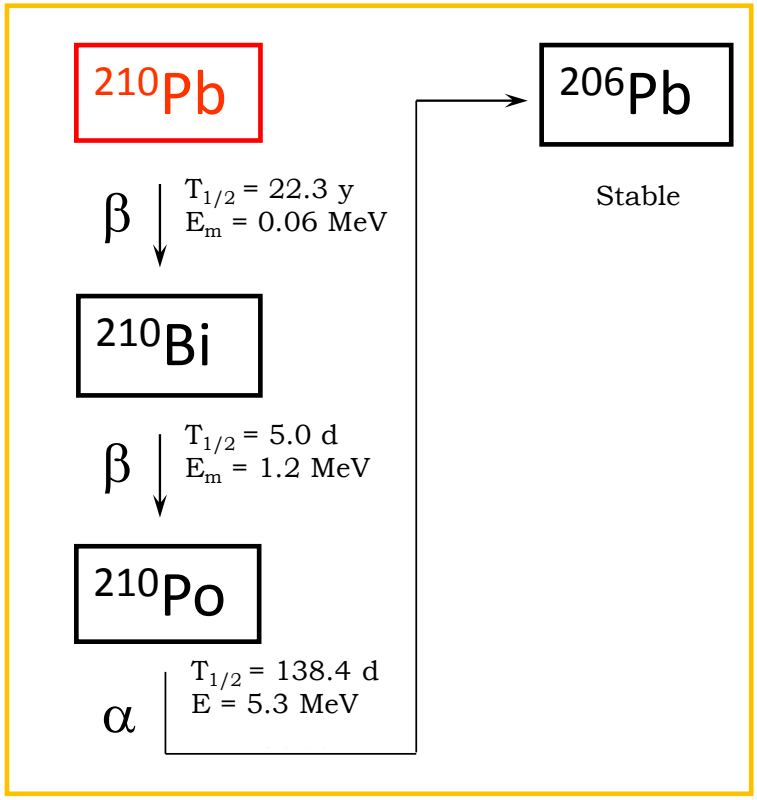
Sample	^{238}U (ICP-MS)	^{226}Ra (HPGe)	^{210}Pb
Arlon 55NT (DarkSide Collaboration)	(1.95 ± 0.05) mBq/kg	(53 ± 5) mBq/kg	(128 ± 26) mBq/kg

^{238}U decay chain – ^{210}Pb / ^{210}Po

Alpha spectrometry

- Large surface, low background alpha spectrometer
- Deconvolution of bulk / surface contribution from registered spectra
- Sensitivity: **~ 50 mBq/kg (bulk)**
  ~ 1 mBq/m^2 (surface)





First direct measurements of ^{210}Po in Cu:

Appl. Rad. Isot. 126 (2017) 165

NIM A. 884 (2018) 157

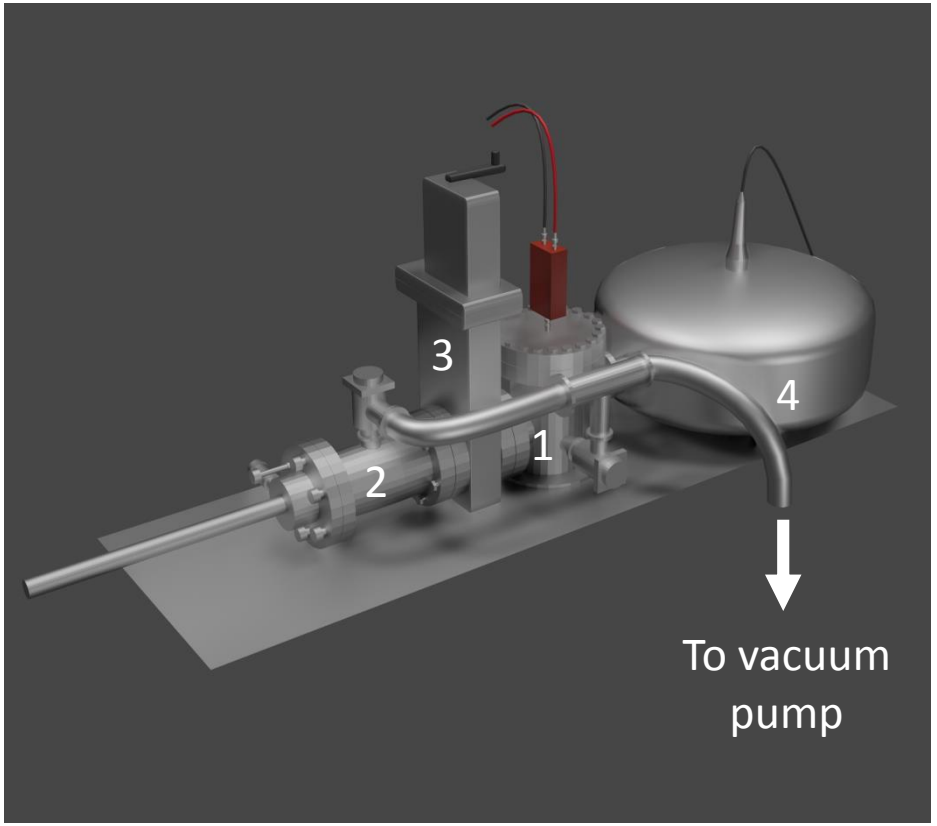
Radiochemical separation of ^{210}Po

- Preparation of 50 g Cu piece and 120 ml of *aqua regia* (HCl:HNO_3 3:1)
- Surface cleaning: etching sample in concentrated HNO_3
- Addition of tracer (30 mBq of ^{209}Po) and dissolution of copper sample in *aqua regia*
- Separation of polonium from copper matrix by ion exchange chromatography using strongly basic resin (Dowex-1)
- Elution of fraction containing ^{209}Po and ^{210}Po with HNO_3
- Source preparation: spontaneous deposition of Po on silver disc from 0.5 N HCl solution
- Drying with ethanol
- Determination of ^{210}Po/ ^{209}Po activities deposited on disc using low background alpha silicon spectrometer

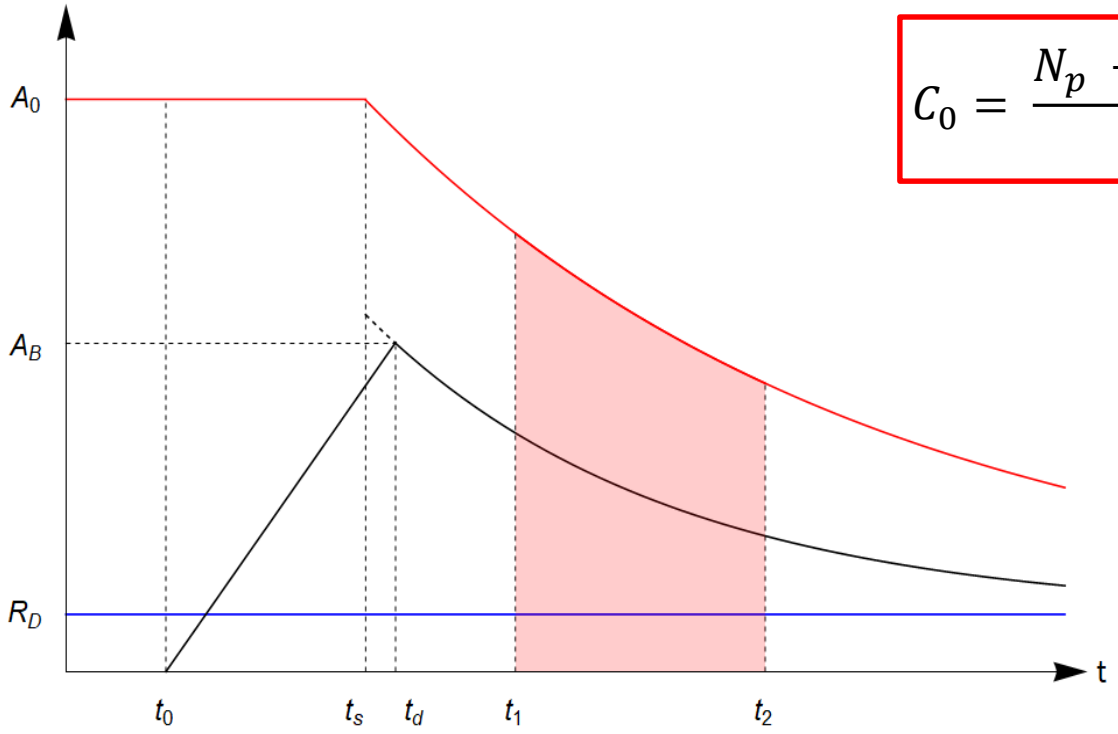
^{210}Pb specific activity is determined form a series of measurements of ^{210}Po in time (^{210}Pb and ^{210}Po achieve equilibrium after ~2 years)

Low background alpha spectrometer

- Based on an 40 mm diameter ORTEC Ultra-AS detector
- Air lock **(2)** with magnetically coupled UHV manipulator and gate valve **(3)** to introduce samples without venting the detector chamber **(1)**
- Vacuum buffer **(4)** for long-term (~2 weeks) measurements without pumping (holds 10^{-1} mbar)
- Source active area diameter: 21 mm
- Energy resolution of the spectrometer: ΔE = 20 keV
- Counting efficiency: ε_g = (33 ± 5) %
- Detector count rate in the ^{210}Po ROI: R_D = (2.2 ± 0.3) cpd



Sample activity calculation



$$C_0 = \frac{N_p - A_B \varepsilon e^{\lambda t_d} W - R_D t_p}{m W \varepsilon}$$

$$W = \frac{e^{-\lambda t_1}}{\lambda}\left(1 - e^{-\lambda t_p}\right)$$

$$\varepsilon = \varepsilon_g \cdot \varepsilon_c = \frac{N_z}{a_z\, m_z\, t_p}$$

N_p – registered number of counts in the counting time t_p

$t_p = t_2 - t_1$

A_0 – polonium activity in time

Detector background: R_D = (2.1 ± 0.2) cpd

Blank of the procedure: A_B = (0.24 ± 0.09) mBq

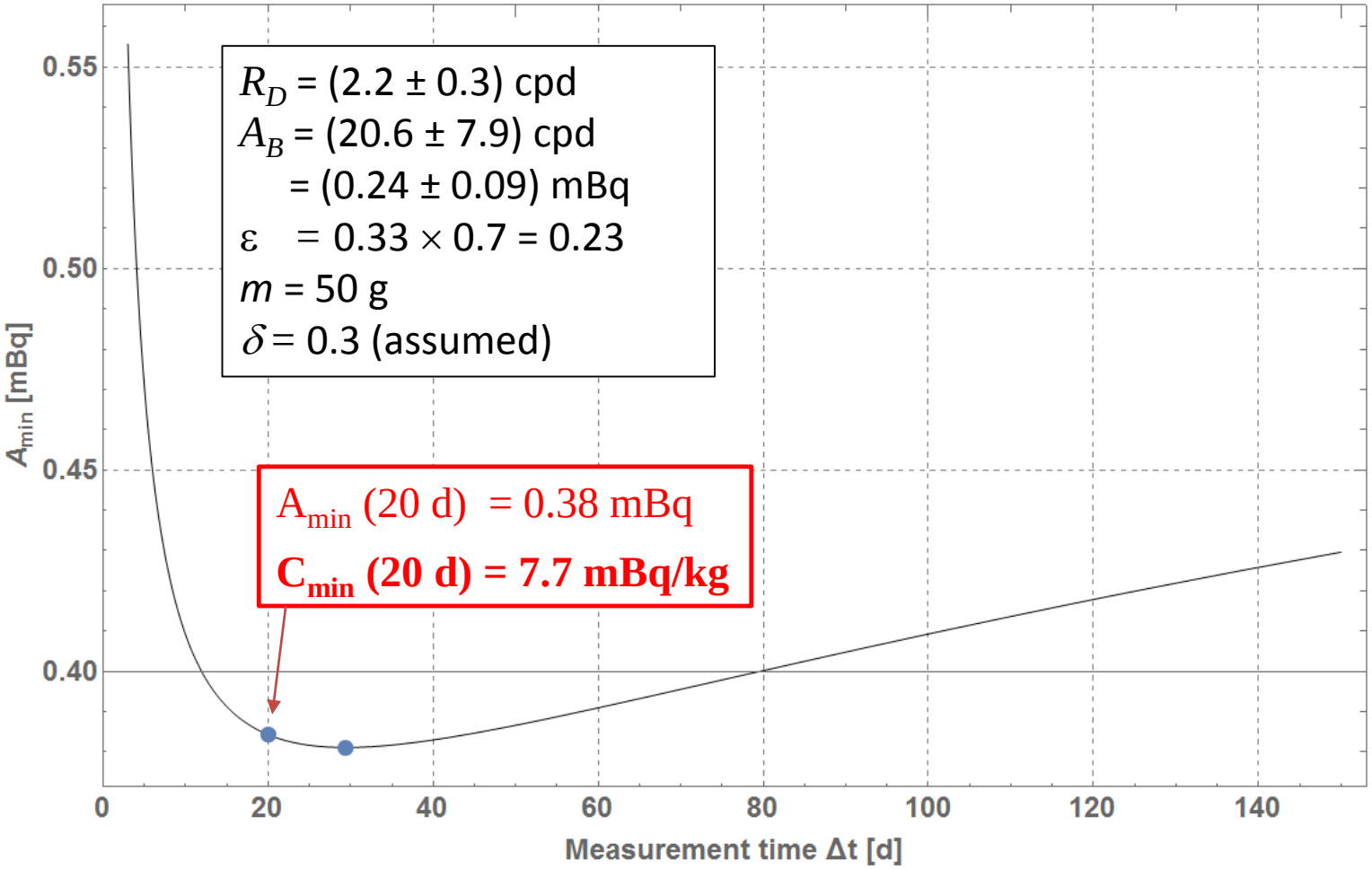
Detection efficiency: ε_g = (33 ± 5) %

Po recovery: ε_c = (50 – 80) % (measurement dependent)

Overall efficiency (ε) determined from the ^{209}Po tracer (a_z, m_z, N_z – added activity of the tracer, its mass and registered number of counts, respectively)

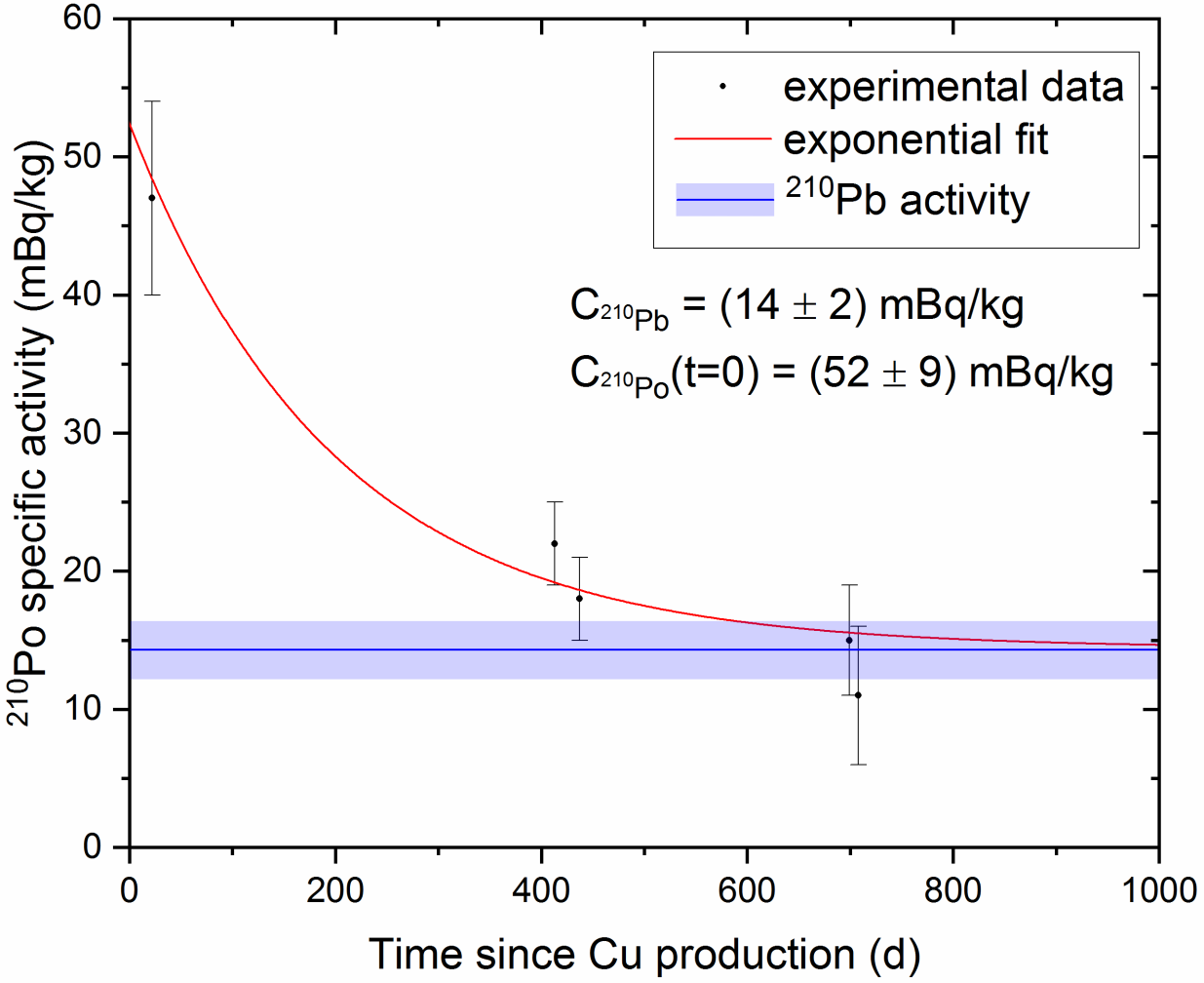
Sensitivity of the method

Sensitivity, in the sense of the Minimum Detectable Specific Activity, depends on the blank activity (A_B), background of the detector (R_D), overall efficiency for ^{210}Po determination including chemical recovery and detector efficiency (ε), amount of material processed (m) and assumed precision (δ).



^{210}Pb in HP copper from Aurubis

High purity copper procured from Aurubis AG (Germany) to be used as shielding material for HPGe spectrometer.



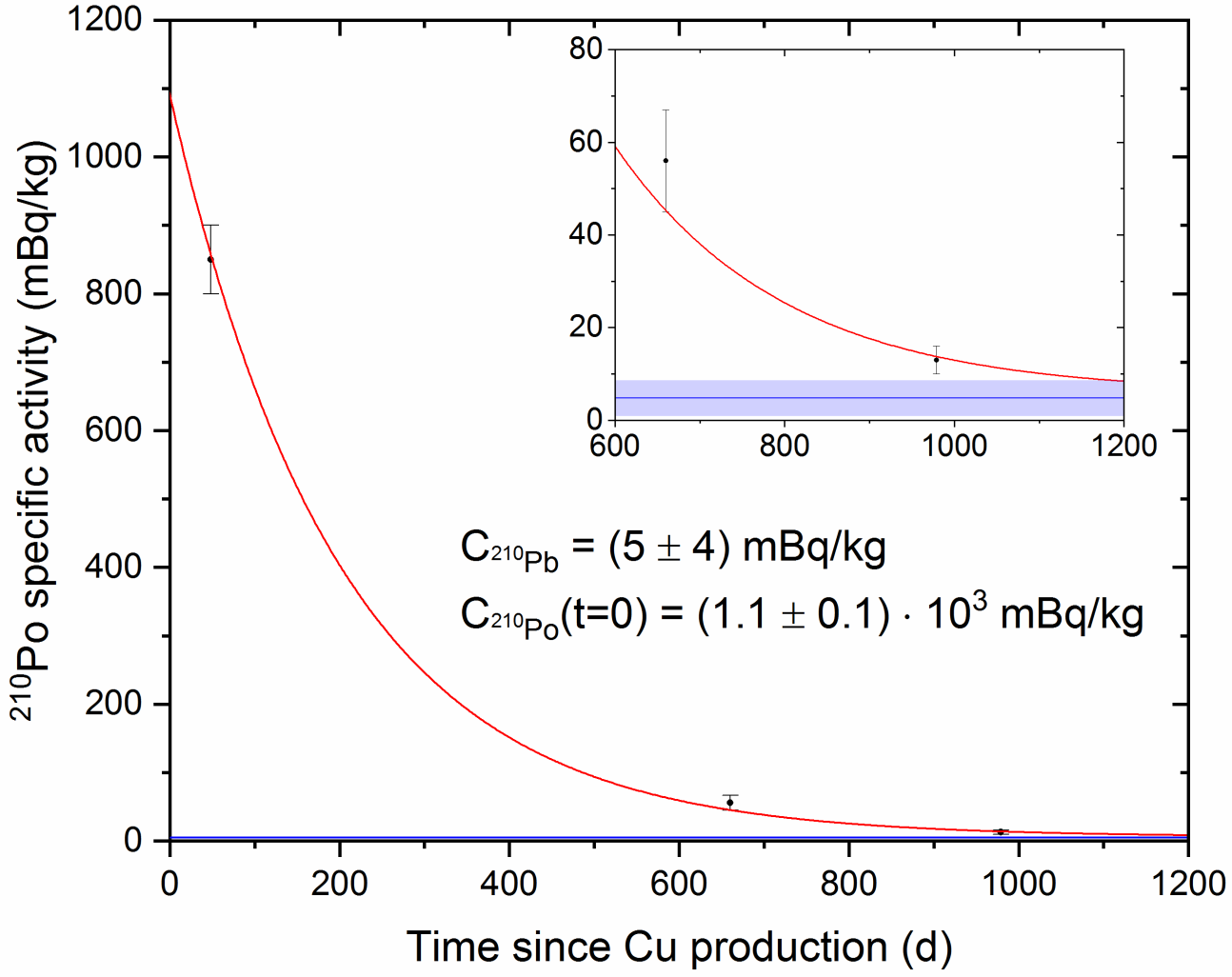
HP copper from Aurubis

Isotope	Specific activity [mBq/kg]	Comments
^{235}U	< 0.069	90 % C.L.
^{40}K	< 0.14	90 % C.L.
^{60}Co	$(14 \pm 4) \cdot 10^{-3}$	
^{234}Th	< 4.2	Upper ^{238}U sub-chain, 90 % C.L.
^{234m}Pa	< 0.45	Upper ^{238}U sub-chain, 90 % C.L.
^{228}Th	< 0.041	90 % C.L.
^{228}Ra	< 0.027	90 % C.L.
^{226}Ra	**$(29 \pm 8) \cdot 10^{-3}$**	Clear disequilibrium between the middle and the bottom ^{238}U sub-chain
^{210}Pb	**14 ± 2**	

HPGe spectrometry, M. Laubestein (LNGS)

^{210}Pb in HP cathode from KGHM

Electrolytic copper produced by KGHM (Poland). Cathode selected in terms of chemical purity.



Various copper samples

Sample	^{210}Pb specific activity [mBq/kg]	Comments
Aurubis HP Copper	14 ± 2	Determined form ^{210}Po decay profile
KGHM HP Cathode	< 11	Determined form ^{210}Po decay profile, 90 % C.L.
OFHC Cu	< 8.4	CF gasket, old sample, ^{210}Pb was in equilibrium with ^{210}Po, 90 % C.L.
30-μm Cu foil	38 ± 6	Result for ^{210}Po, only one measurement
Fire refined copper	$(10 \pm 0.6) \cdot 10^3$	Old sample, ^{210}Pb was in equilibrium with ^{210}Po
	$(9.6 \pm 0.8) \cdot 10^3$	Measured applying alpha spectrometry (XIA)

Conclusions

- Due to potential disequilibrium in the ^{238}U chain, the sub-chains (top, middle, bottom) should be assayed separately
- In the bottom sub-chain, the most relevant ^{210}Pb, may be assayed through ^{210}Po (alpha spectrometry or chemical separation)
- Presented method allows to determine ^{210}Po in Cu at a single mBq/kg level (presently, the most sensitive method)
- In high purity copper samples determined ^{210}Pb specific activity was up to 14 mBq/kg
- Despite the higher initial activity of ^{210}Po, the cathode from KGHM has significantly lower content of ^{210}Pb than the copper from Aurubis
- At least for one sample (Cu from Aurubis) a clear disequilibrium between ^{226}Ra (middle part of ^{238}U chain) and ^{210}Pb (bottom part of ^{238}U chain) has been observed
- Work in progress to improve sensitivity of the method

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