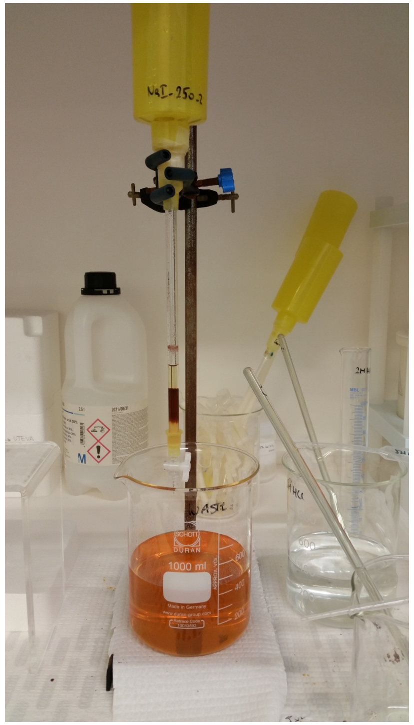
Advanced Metrology via Atom Counting



Michaela Froehlich
Research School of Physics
The Australian National University

1 MV VEGA accelerator, ANSTO

2023 CDM Annual Workshop - Collaboratively striving for success.

Cultural background

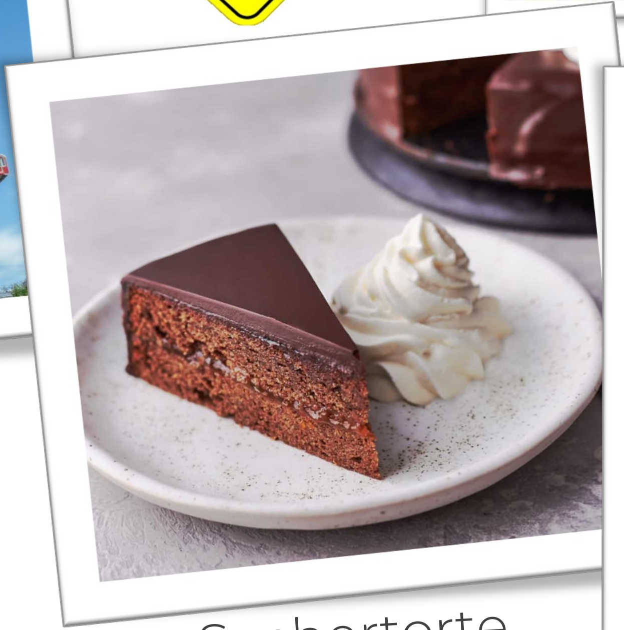
Austria - Australia

Cultural background

Austria - Australia

Snow in December!

Wiener Riesenrad



Sachertorte

Tiergarten Schönbrunn

HUH?!???

ARC CENTRE OF EXCELLENCE FOR DARK MATTER PARTICLE PHYSICS

Advanced Metrology

Determining radioactivity in detector components.

Why do we care?

Dark matter detector components & surrounding laboratory need to be constructed with minimal inherent (internal) radioactivity.

→ minimise interference with the expected signal from the detector should a dark matter particle be detected.

Advanced Metrology

Determining radioactivity in detector components such as ^{40}K, ^{129}I, ^{210}Pb, ^{232}Th, ^{238}U, …

Ways to measure the radioactive content, e.g.,

- Inductively Coupled Mass Spectrometry (ICP-MS)
- Gamma Spectrometry
- Accelerator Mass Spectrometry (AMS)

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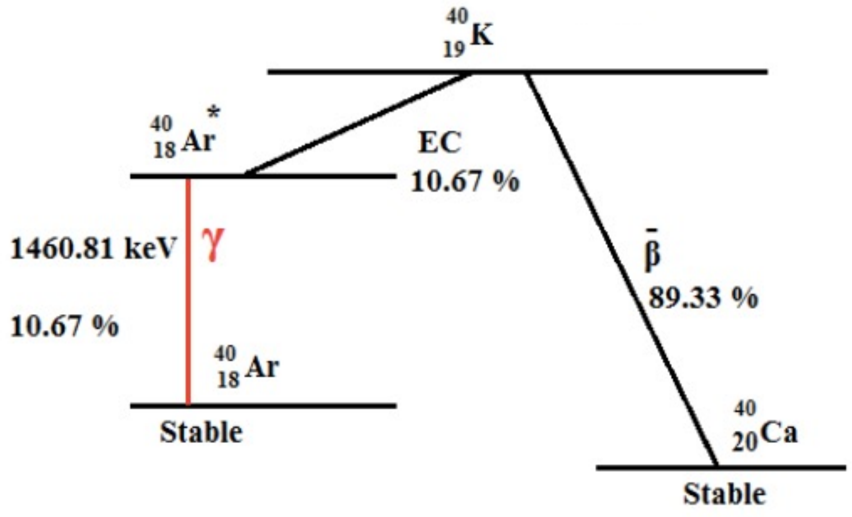
Potassium-40, half-life = 1,250 Myrs

Primordial radionuclide.

^{40}K: 0.012%, ^{39}K: 93.3% → ^{40}K/^{39}K = 1.29x10^{-4}.

Decay via EC to ^{40}Ar (1460 keV gamma, low-energy Auger electrons) and via beta decay to ^{40}Ca.

Measurement capability: ICP-MS.



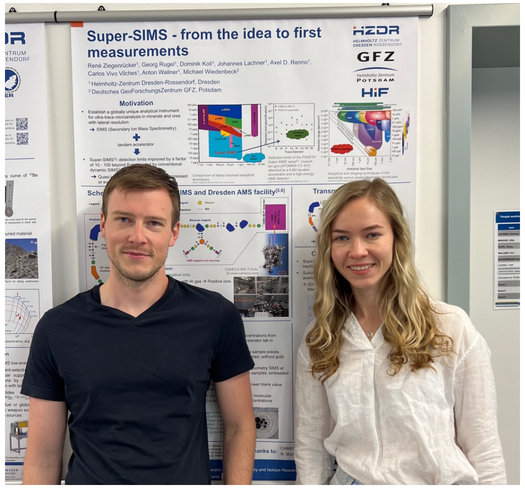
Potassium-40, half-life = 1,250 Myrs

BUT: ICP-MS: background from molecules & $^{38,40}Ar$ from the plasma, dilution.

SOLUTION: SIMS: solid crystals, small samples, no dilution, combine AMS with SIMS → Super-SIMS

Super-SIMS to characterize ultra-low ^{40}K concentrations in NaI crystals

(Centre funding: Assoc. Investigator Dr D. Koll, HZDR & Dr Z. Slavkovská, ANU).



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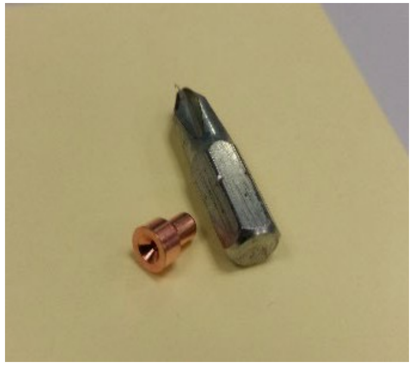
Iodine-129, half-life = 15.7 Myrs

^{129}I routine AMS isotope: isotope ratio $^{129}I/^{127}I$

No isobaric interference.

AMS background at ANU: $^{129}I/^{127}I < 10^{-14}$.

^{129}I/NaI …. $^{129}I/^{127}I$ isotope ratio independent of sample mass. Only need a few mg!



AgI + Ag → AMS target

Iodine-129, half-life = 15.7 Myr

^{129}I routine AMS isotope: isotope ratio ^{129}I/^{127}I

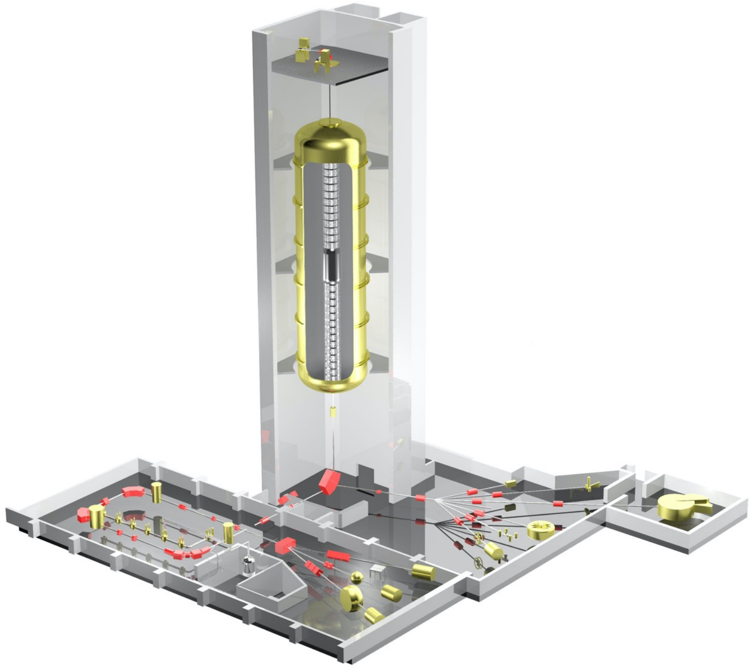
No isobaric interference.

AMS background at ANU: ^{129}I/^{127}I $<10^{-14}$.

^{129}I/NaI …. ^{129}I/^{127}I isotope ratio independent of sample mass. Only need a few mg!

Compare Bernabei et al., 2008 (DAMA/LIBRA): ^{129}I/I = $(1.7 \pm 0.1)\times10^{-13}$.

Convert to specific activity → 1 mBq/kg.



14UD tandem accelerator, ANU

Iodide compound	^{129}I/I [$\times10^{-13}$]
NaI (Analytical Grade)	1.5 ± 0.1
NaI (Growth Grade)	1.9 ± 0.1
NaI (Astro Grade)	1.6 ± 0.1

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Advanced Metrology

Determining radioactivity in detector components such as ^{40}K, ^{129}I, ^{210}Pb, ^{232}Th, ^{238}U, …

Ways to measure the radioactive content, e.g.,

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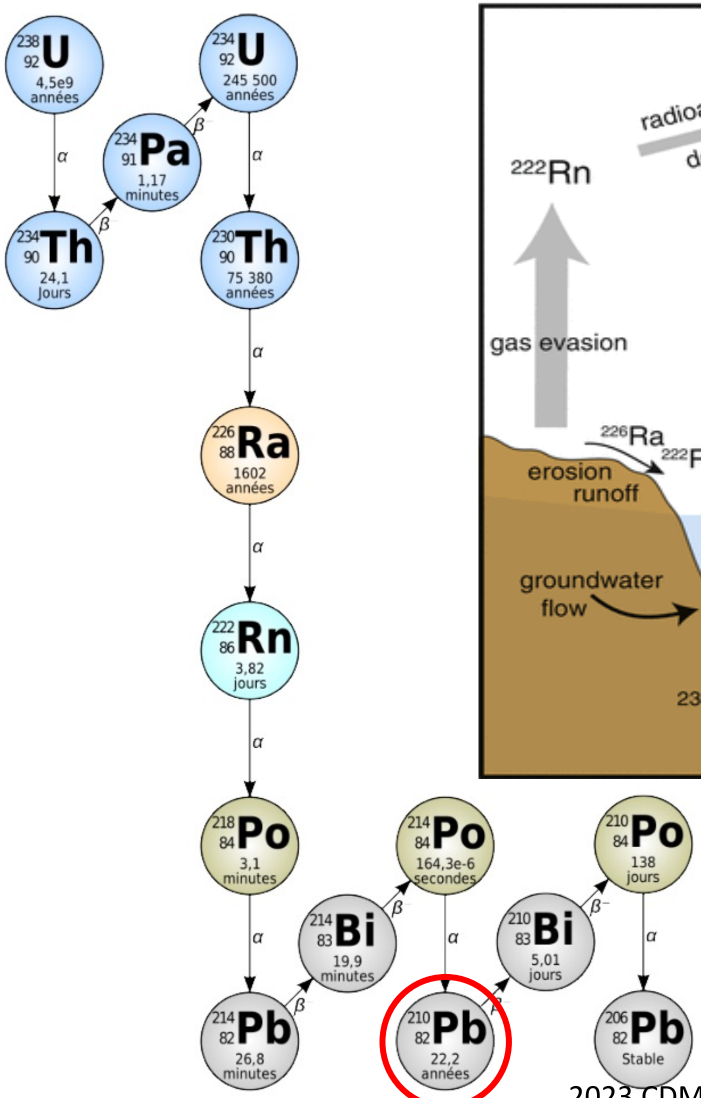
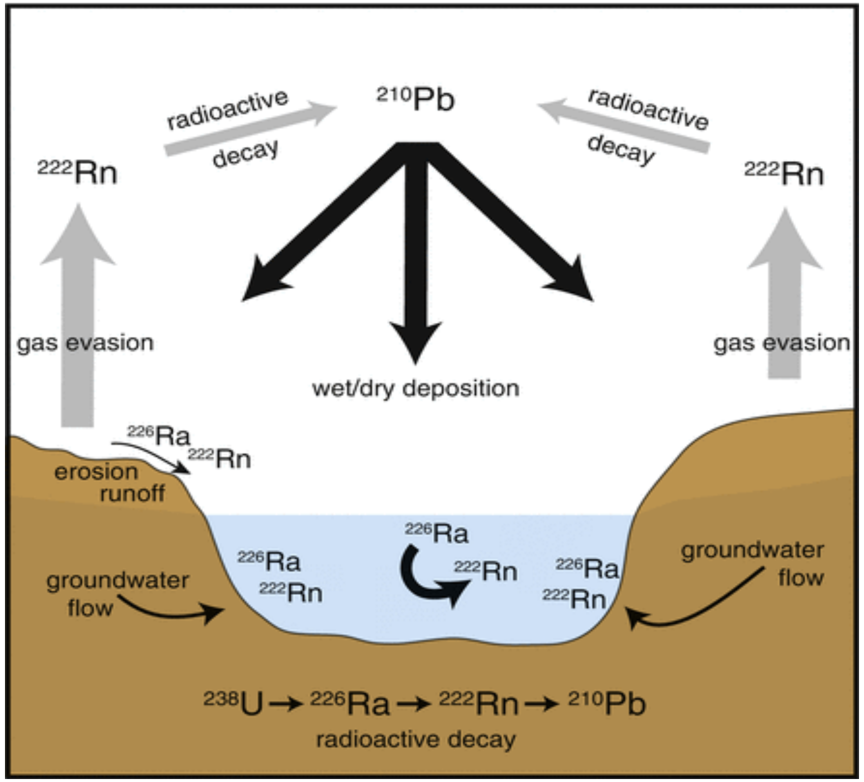
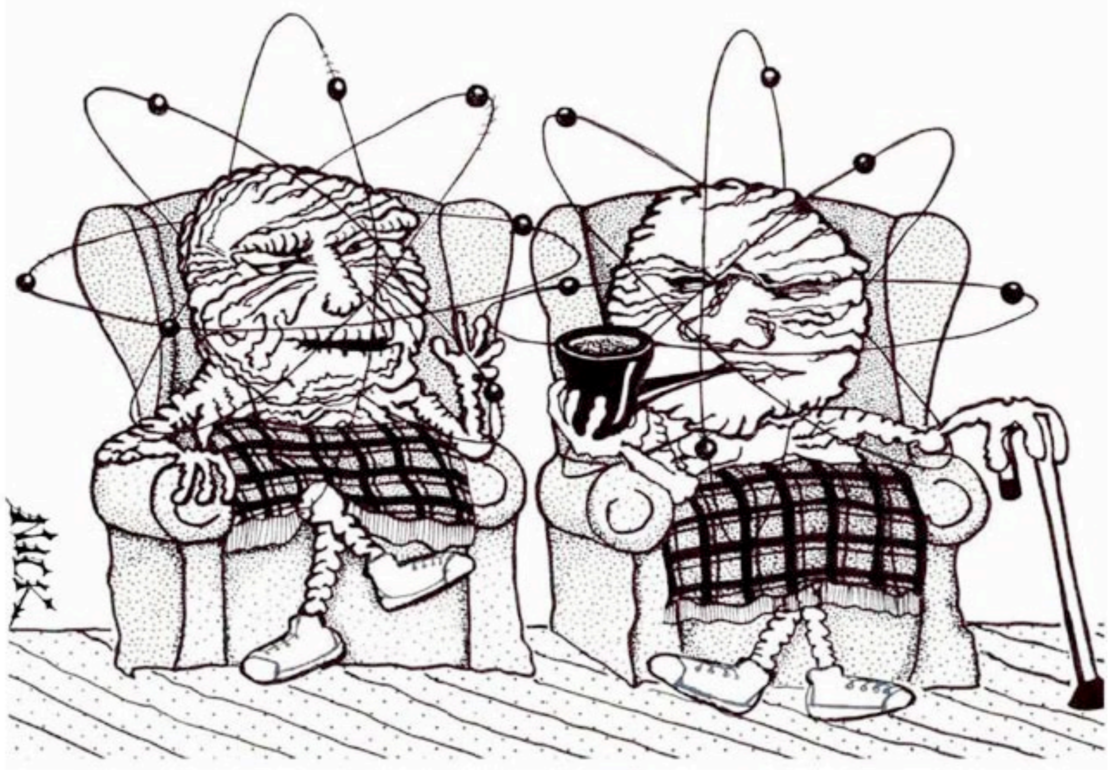
Lead-210, half-life = 22.2 yrs







"When I was young I used to feel so alive and dangerous! Would you believe I started life as a uranium-238? Then one day I accidentally ejected an alpha particle. Now look at me—a spent old atom of lead-206. It seems that all my life since then has been nothing but decay, decay, decay…"

Lead-210 screening via atom counting

"Lead is among one of the most frustrating elements that we have encountered." by Roy Middleton in *A Negative-Ion Cookbook* (1989).

Advantages of AMS:

Low ratios of 10^{-10} – 10^{-17}.
Low number of atoms: 10^{5} – 10^{8} atoms.

Progress over the last 20 years:

University of Vienna	^{210}Pb/Pb = 5.7×10^{-11}	(3MV accelerator, 2001).
University of Ottawa	^{210}Pb/Pb = 2.5×10^{-12}	(3MV accelerator, 2015).

NaI: Expected ^{210}Pb < 0.03 mBq/kg → ^{210}Pb/Pb = < 7.0×10^{-15}.

Antonello et *al*., Astroparticle Physics, 2019; 106:1-9.

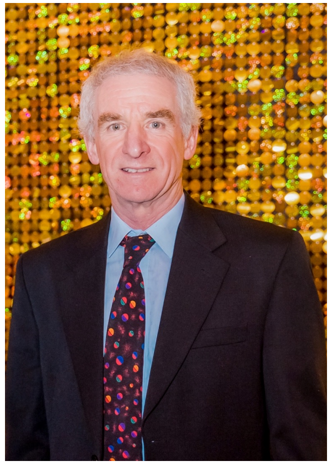
Accelerator Mass Spectrometry (AMS)

The art of identifying and counting rare atoms one by one.

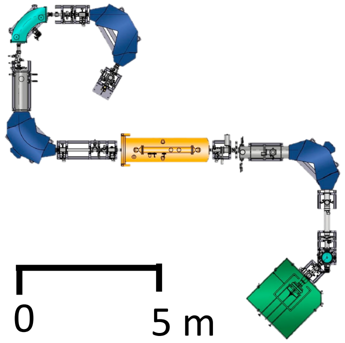
Main benefits of AMS:

- ✓ No isobaric background.
- ✓ Isotope ratios: 10^{-12} – 10^{-17}.
- ✓ Ultrasensitive.

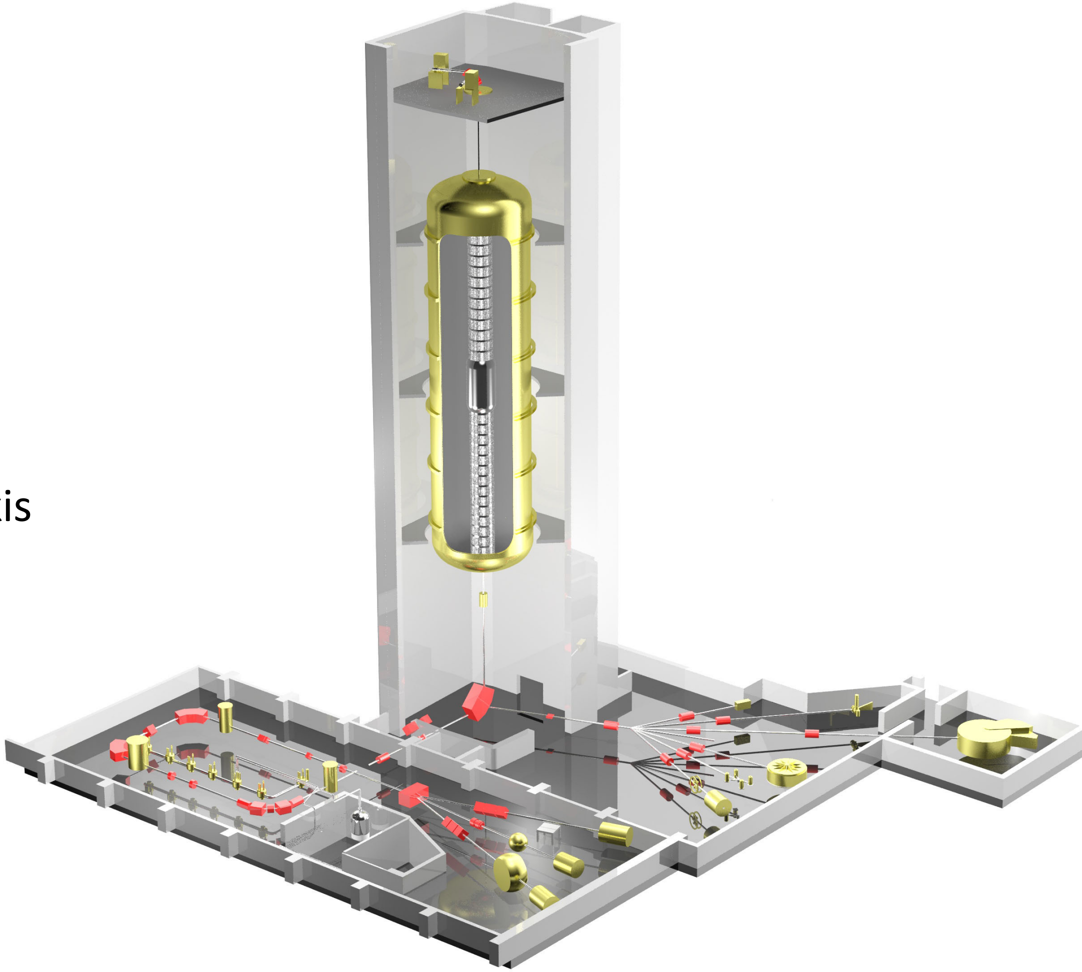
→ Sample-dependent chemistry!



Dr Michael Hotchkis



1 MV VEGA accelerator, ANSTO



14UD tandem accelerator, ANU

How do we get ^{210}Pb?

Dissolving NaI

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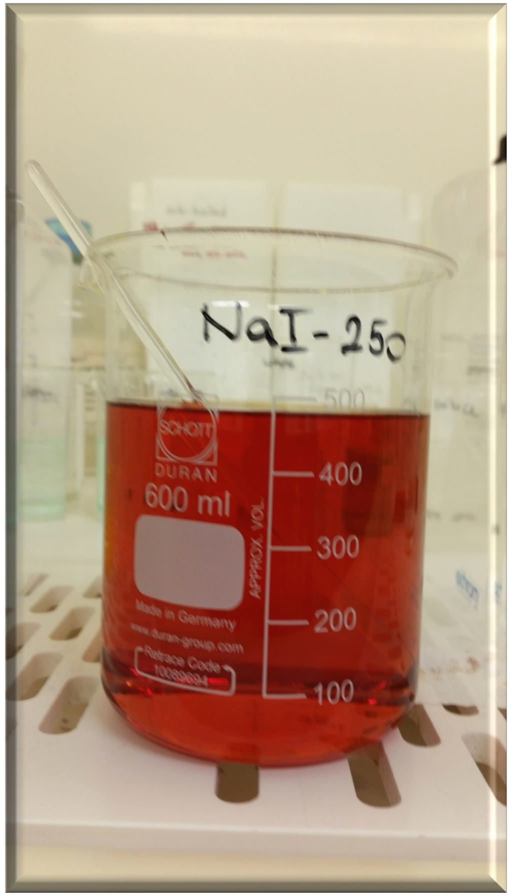
Dissolving NaI



How do we get ^{210}Pb?

Dissolving NaI + adding a Pb carrier.

Any Pb compound in our lab.

Enriched Pb.

18th century church roof piece.

Kanazawa Castle (Japan).

Pb from Canadian AMS group.

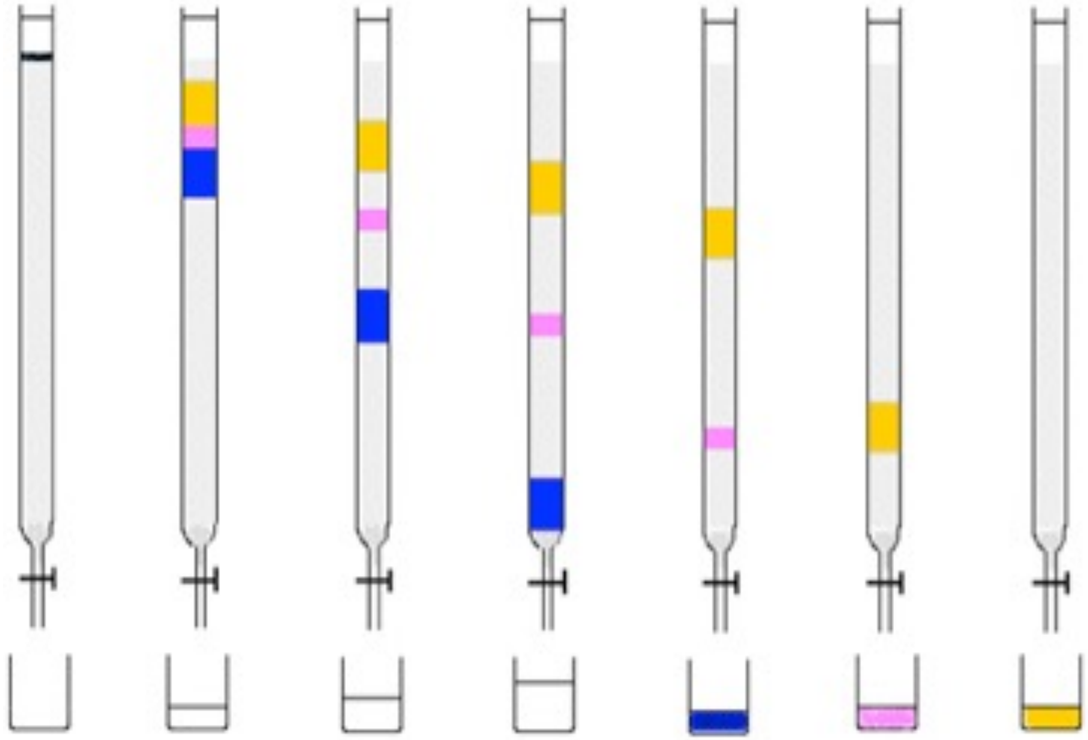
LNGS.

Pb material from underground labs in Germany.

Pb compound	Pb compound
Pb (metal powder)	Japan
PbO	Hampton Court Palace roof
PbO_2	LANL stock & Chicago stock
Pb_3O_4	U. Chicago Spanish lead
PbF_2	PNNL ancient lead
ICP-MS Pb	LNGS material
Pb-204	Munich
Pb-206	Brick (Felsenkeller)
Pb_20001 (roof)	

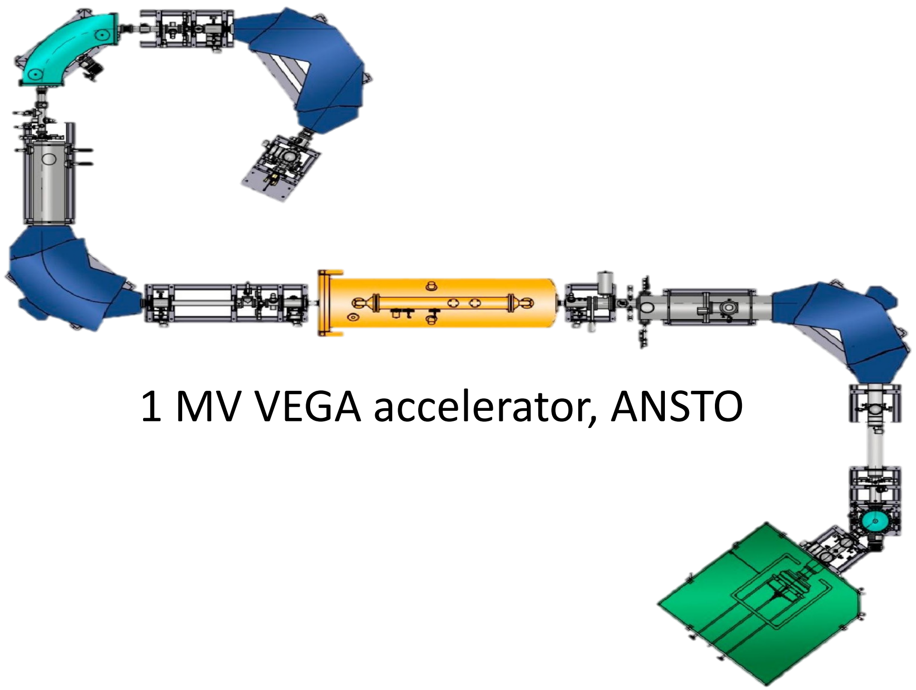
How do we get ^{210}Pb?

Dissolving NaI + adding a Pb carrier + column chromatography.



How do we get ^{210}Pb?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.



1 MV VEGA accelerator, ANSTO

So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

EASY!!

NaI is highly soluble in H_2O and HCl.

So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

Need to find a suitable Pb carrier, i.e., $^{210}Pb/Pb$ = < 7.0×10^{-15}.

Reminder: ^{210}Pb is everywhere!

Pb compound	$^{210}Pb/Pb$	Pb compound	$^{210}Pb/Pb$
Pb (metal powder)	-	ICP-MS Pb	9.6×10^{-14}
PbO	1.2×10^{-14}	Brick	-
PbO_2	1.1×10^{-14}	LNGS	-
Pb_3O_4	4.8×10^{-14}	U. Chicago Spanish lead	9.1×10^{-15}
PbF_2	3.9×10^{-14}	PNNL ancient lead	2.7×10^{-15}
Pb_20001 (roof)	7.3×10^{-15}	Hampton Court Palace roof	2.5×10^{-15}
^{204}Pb	-	LANL stock	1.7×10^{-15}
^{206}Pb	9.1×10^{-15}	U. Chicago stock	2.3×10^{-14}

So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

Need to find a suitable Pb carrier, i.e., $^{210}Pb/Pb$ = < $7.0×10^{-15}$.

Pb compound	$^{210}Pb/Pb$	Pb compound	$^{210}Pb/Pb$
Pb (metal powder)	-	ICP-MS Pb	$9.6×10^{-14}$
PbO	$1.2×10^{-14}$	Brick	-
PbO_2	$1.1×10^{-14}$	LNGS	-
Pb_3O_4	$4.8×10^{-14}$	U. Chicago Spanish lead	$9.1×10^{-15}$
PbF_2	$3.9×10^{-14}$	PNNL ancient lead	$2.7×10^{-15}$
Pb_20001 (roof)	$7.3×10^{-15}$	Hampton Court Palace roof	$2.5×10^{-15}$
^{204}Pb	-	LANL stock	$1.7×10^{-15}$
^{206}Pb	$9.1×10^{-15}$	U. Chicago stock	$2.3×10^{-14}$

So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

Need to find a suitable Pb carrier, i.e., $^{210}Pb/Pb$ = < 7.0×10^{-15}.

Pb compound	$^{210}Pb/Pb$ [$\times10^{-15}$]
LANL stock	2.08 ± 0.42
Hampton Court Palace roof	1.41 ± 0.28
PNNL ancient lead	0.15 ± 0.03
LNGS	0.24 ± 0.05
Brick	1.13 ± 0.23
Munich	0.26 ± 0.05
Pb metal powder	27.5 ± 0.55
Japan	1.17 ± 0.23

So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

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ARC CENTRE OF EXCELLENCE FOR DARK MATTER PARTICLE PHYSICS

This brings us back to …

Progress over the last 20 years:

University of Vienna $^{210}Pb/Pb = 5.7\times10^{-11}$ (3MV accelerator, 2001).
University of Ottawa $^{210}Pb/Pb = 2.5\times10^{-12}$ (3MV accelerator, 2015).
ANSTO $^{210}Pb/Pb = 1.5\times10^{-16}$ (1MV accelerator, 2023)!!!

So, the procedure is simple, right?

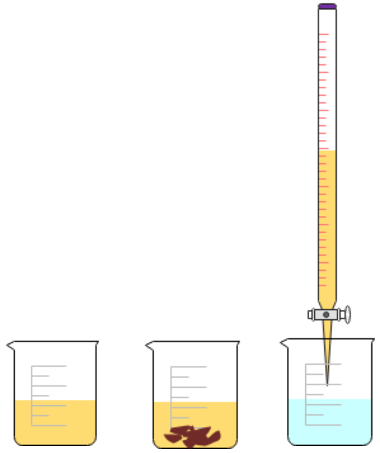
Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

1 kg NaI (4× 250 g); tests with analytical grade NaI

Option 1: dissolve NaI

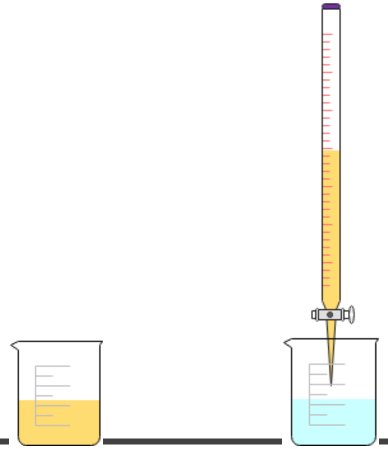
pre-concentration via Fe/Pb precipitation

column chromatography (AE resin) → Pb



Option 2: dissolve NaI

column chromatography (Sr resin) → Pb



Chemistry Challenge

Option 1 – Advantage:

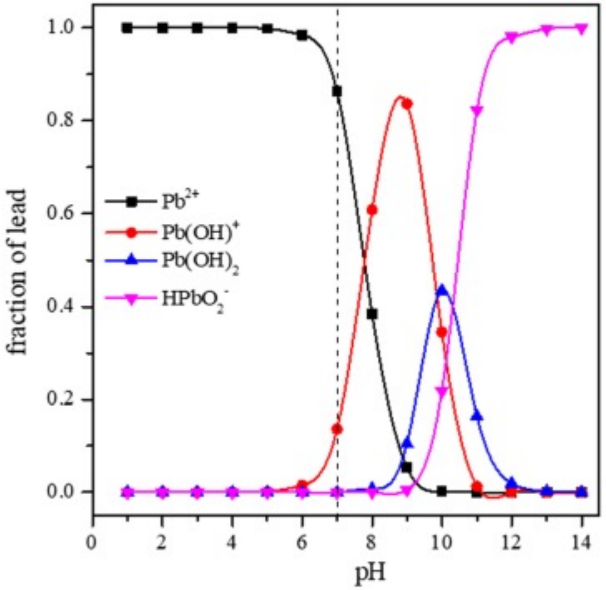
Pre-concentration

Less chemicals

Disadvantage:

pH dependant

System	logβ
$Pb^{2+} + OH^- \rightleftharpoons Pb(OH)^+$	6.2
$Pb^{2+} + 2OH^- \rightleftharpoons Pb(OH)_2$	10.3
$Pb^{2+} + 3OH^- \rightleftharpoons HPbO_2^- + H_2O$	14.0



Oliveira L.C., et *al.*, Water Air Soil Pollut (2019) 230: 191.

Option 2 – Advantage:

Directly on the column

Sr resin → higher Pb retention

Disadvantage:

85% iodide (I^-)

Break-through & bleeding

ARC CENTRE OF EXCELLENCE FOR DARK MATTER PARTICLE PHYSICS

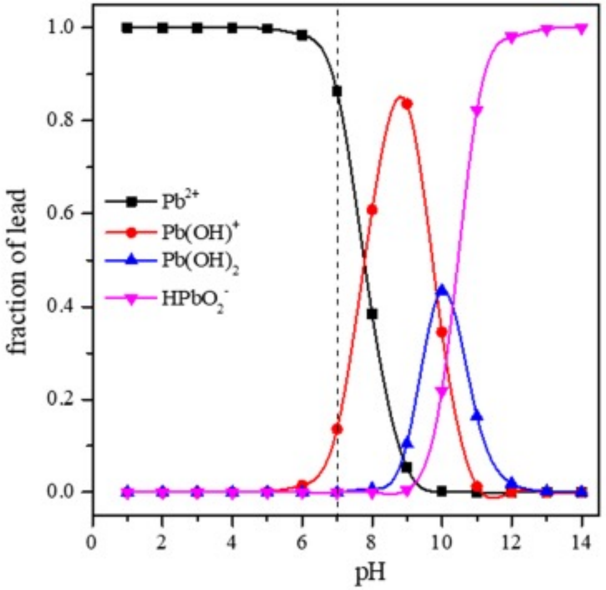
Chemistry Challenge

Option 1 – Advantage:
Pre-concentration
Less chemicals

Disadvantage:
pH dependant

Still in progress

System	logβ
$Pb^{2+} + OH^- \rightleftharpoons Pb(OH)^+$	6.2
$Pb^{2+} + 2OH^- \rightleftharpoons Pb(OH)_2$	10.3
$Pb^{2+} + 3OH^- \rightleftharpoons HPbO_2^- + H_2O$	14.0



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So, the procedure is simple, right?

Dissolving NaI + adding a Pb carrier + column chromatography + AMS.

Table 1. Potential interferences during Pb isotope measurements

Pb isotope	Mass-to-charge interferences at injection magnetic	Mass-to-charge interference(s) in final detector
$^{204}Pb^{a}$	$^{68}Zn_3^{57}Fe^-$	$^{68}Zn^{+1}$
$^{205}Pb^{b}$	$^{205}TlF_3^-$	$^{205}Tl^{+3}$
$^{208}Pb^{a}$	–	–
$^{210}Pb^{b}$	$^{70}Zn^{140}Ce^{19}F_3^-$	Coincidence counting of $^{70}Zn^{+1}$ and $^{140}Ce^{+2}$

[a]Collected in a Faraday cup.
[b]Collected in a gas ionization chamber.

A. Sookdeo et *al*. Rapid Commun Mass Spectrom 30 (2015) 867.

Table 1
Ratios of $PbXY^-$ compounds mixed with fluorinating agents or Ag and the currents measured from the respective mixtures.

Target composition	Ratio by weight	Average ^{208}Pb molecular anion current (nA)	Molecular anions
$PbF_2 + AgF_2$	1:1	75	$^{208}PbF_3^-$
$PbF_2 + AgF_2 + CsF$	9:6:10	175	$^{208}PbF_3^-$
$PbO + AgF_2$	3:5	63	$^{208}PbOF^-$
$PbO + PbF_2$	3:5	23 53	$^{208}PbF_3^-$, $^{208}PbOF^-$
$PbO + PbF_2 + AgF_2$	4:5:3	27.5 120	$^{208}PbF_3^-$, $^{208}PbOF^-$
$PbO + PbF_2 + AgF_2 + CsF$	5:5:4:4	145 25	$^{208}PbF_3^-$, $^{208}PbOF^-$
$Pb(SCN)_2 + Ag$	2:3	0.6	$^{208}Pb(SCN)_2^-$
$Pb(SCN)_2 + AgF_2$	7:6	60	$^{208}PbF_3^-$
$Pb(NO_2)_2 + Ag$	5:2	0	–
$PbSO_4 + Ag$	4:5	4	$^{208}PbO_2^-$ & $^{208}PbS^-$
$PbCO_3 + Ag$	5:8	12.5	$^{208}PbO_2$

A. Sookdeo et *al*. NIMB 361 (2015) 450.

AMS challenge

Samples do not last, i.e., beam drops after 15-30 min.

AMS challenge

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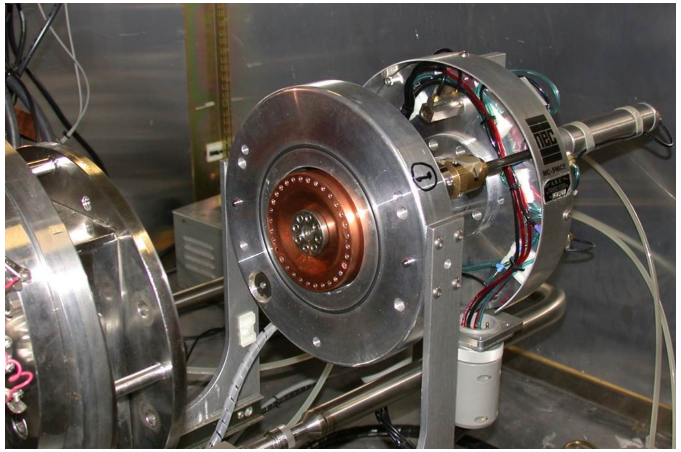
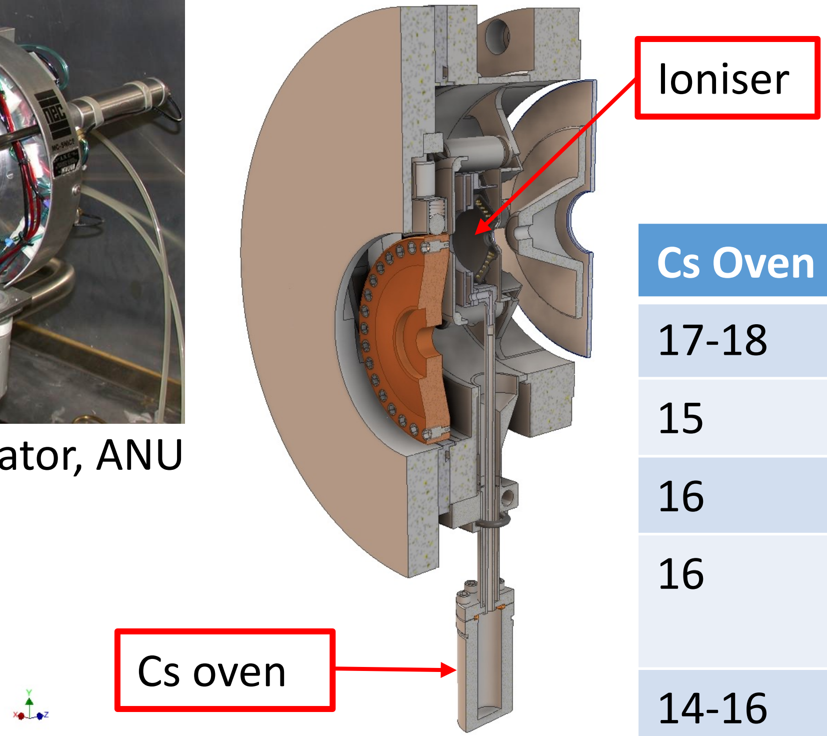
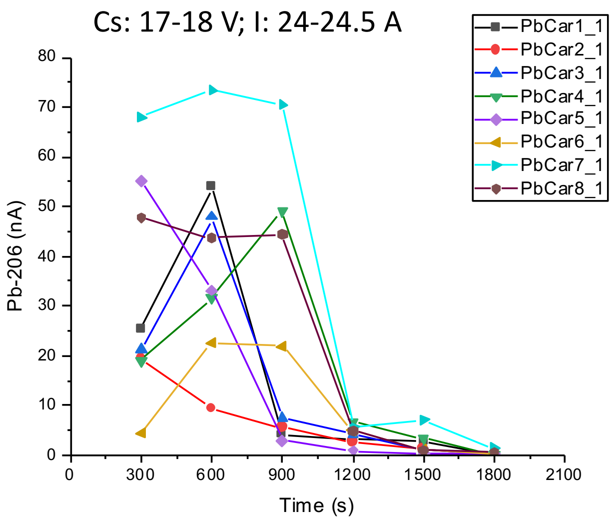


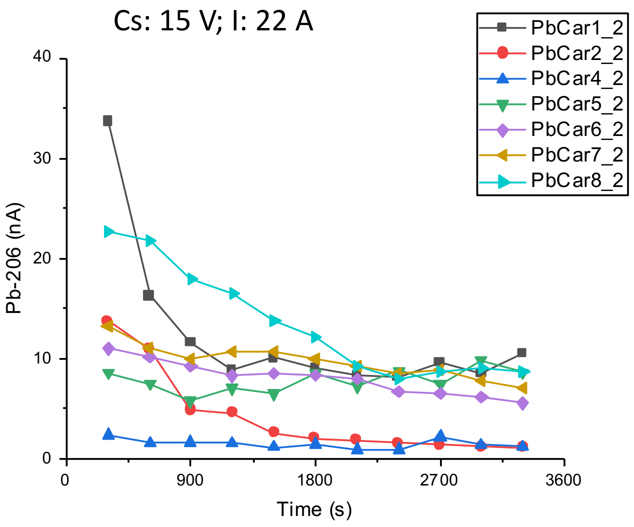
Ion source, 14UD accelerator, ANU

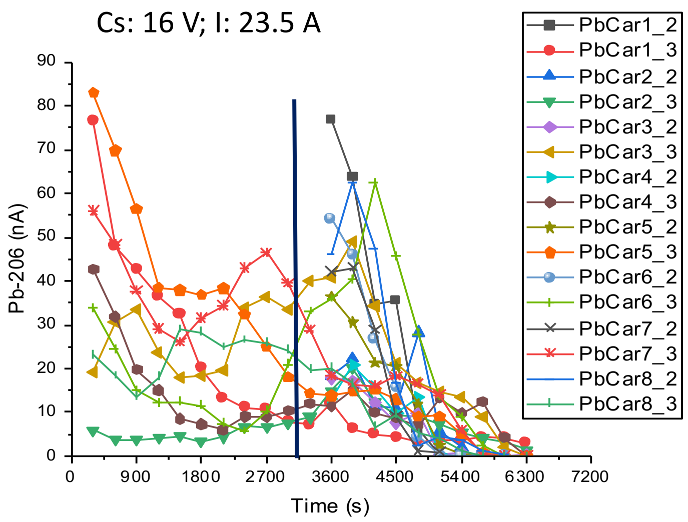


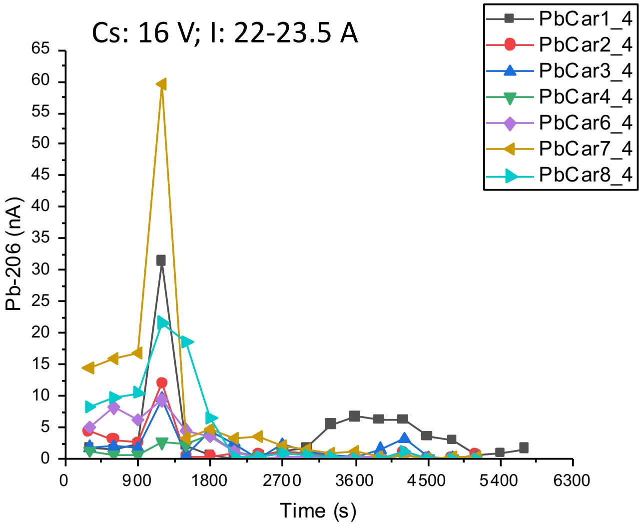
Cs Oven [V]	Ioniser [A]	Comment
17-18	24-24.5	Standard setting (high)
15	22	Low source setting
16	23.5	Medium source setting
16	22-23.5	Low ioniser, medium oven; ioniser increased in steps
14-16	21-23	Start low, progressively increase both ioniser and oven

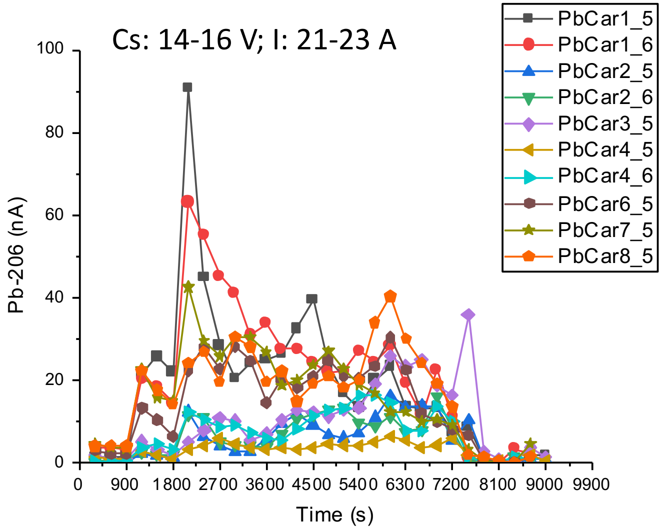
Results – different ioniser settings





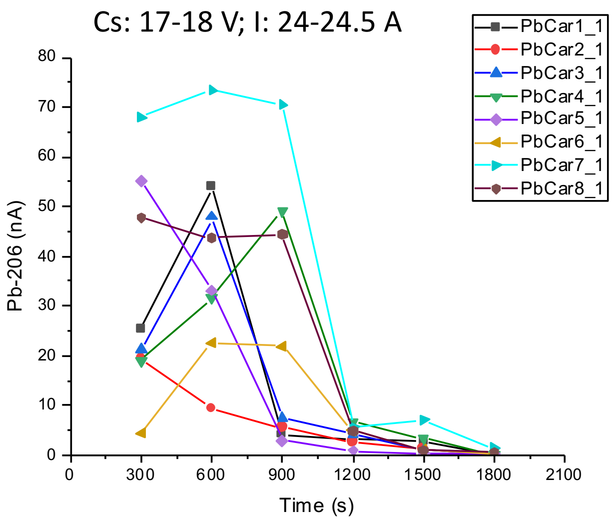


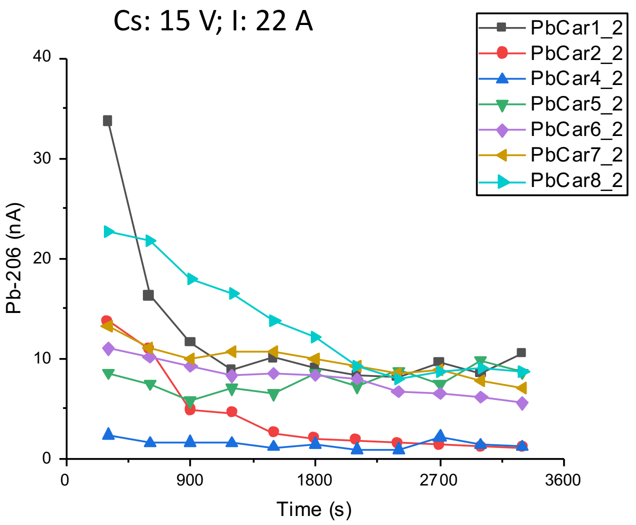


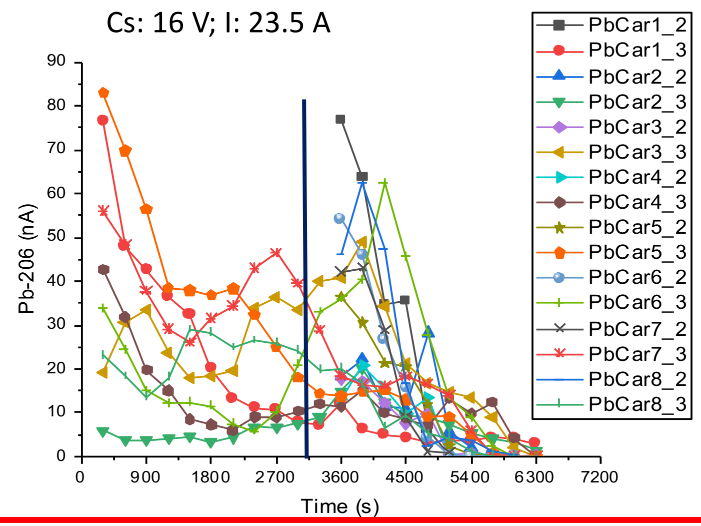


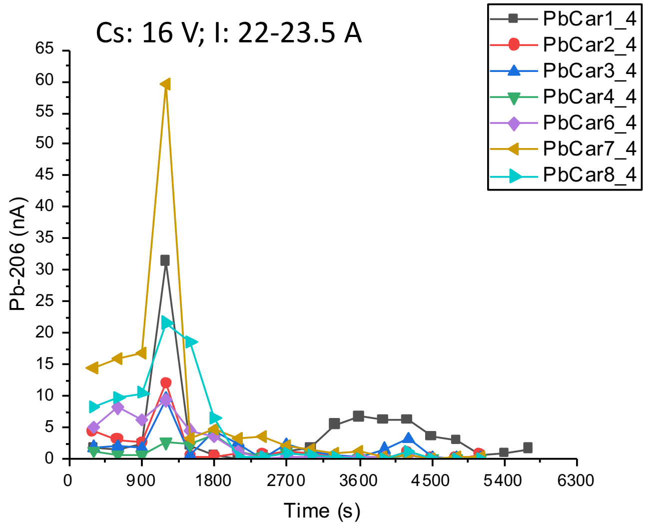
ARC CENTRE OF EXCELLENCE FOR DARK MATTER PARTICLE PHYSICS

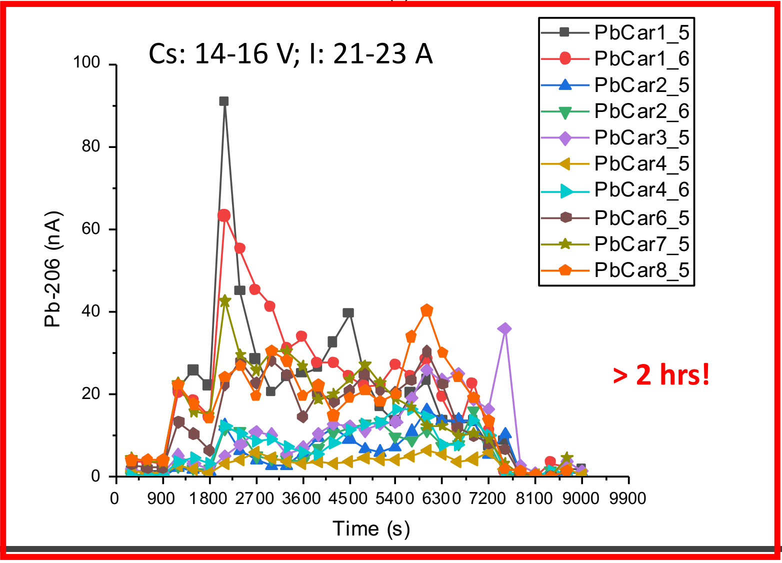
Results – different ioniser settings











Summary

Clean materials are required to construct and operate many low-background physics experiments → identify background (^{40}K, ^{129}I, ^{210}Pb, …).

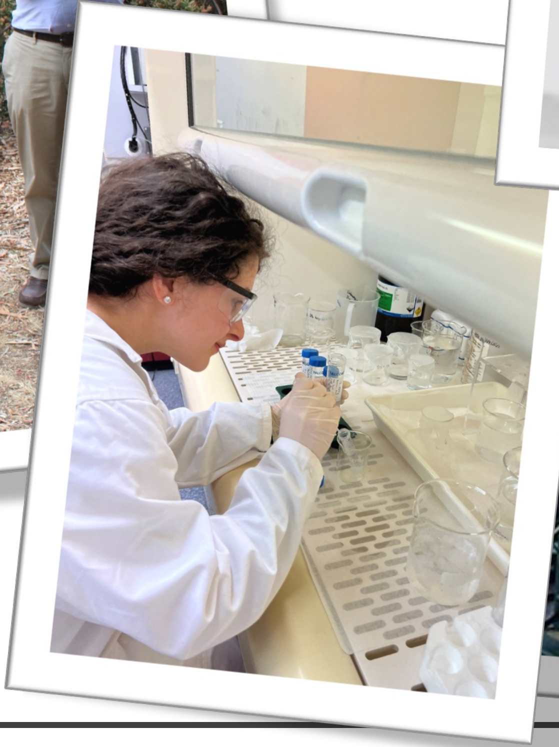
Some are easier than others:

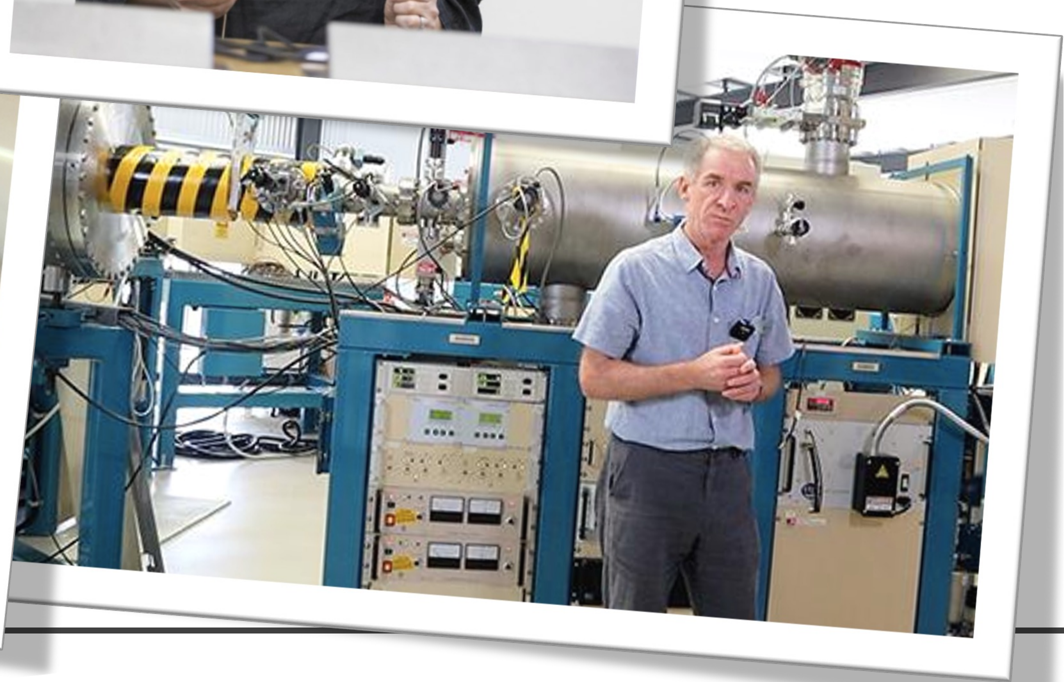
^{40}K → Super-SIMS?

^{129}I → done by AMS.

^{210}Pb → optimise chemical extraction procedure for AMS.

This could not have been done without a great team!





Happy festive season!

ARC CENTRE OF EXCELLENCE FOR DARK MATTER PARTICLE PHYSICS