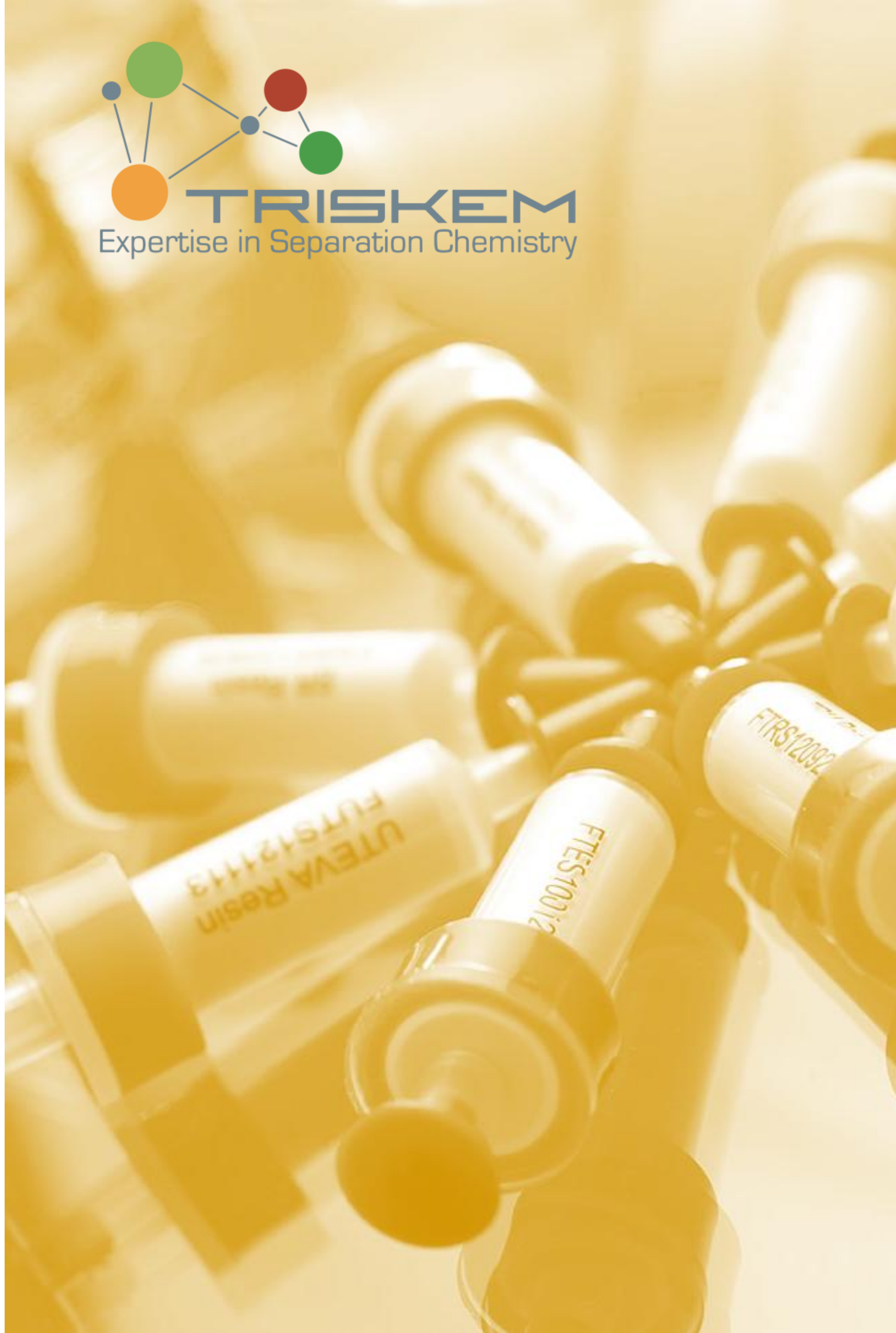
Optimization of the determination procedures to quantify DTM radionuclides in decommissioning samples

Inés Llopart, Steffen Happel, Alex Tarancón

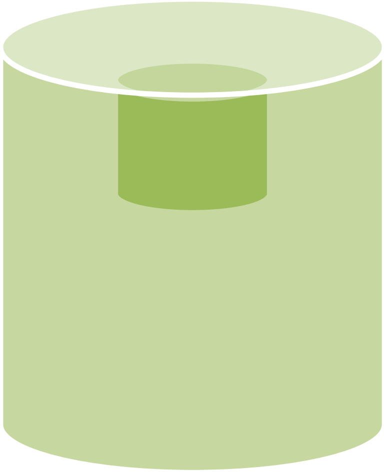
03-11-2025

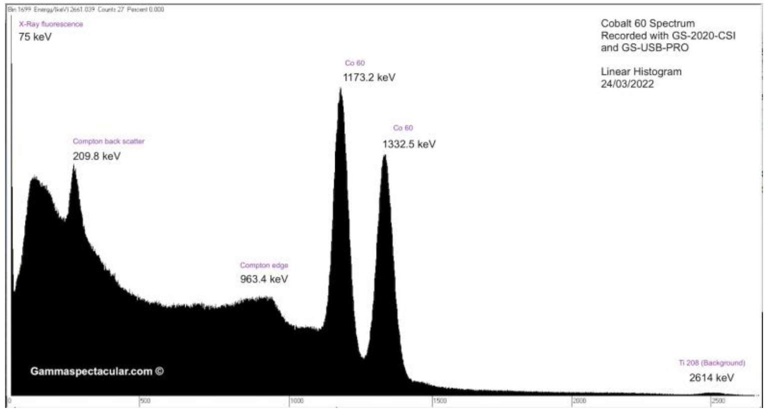


Difficult-to-measure radionuclides

*“a radionuclide whose radioactivity is difficult to measure **directly from the outside** of the waste packages by non-destructive assay means”*

ISO standard 24390:2023





Taken from Gamma Spectacular. (s. f.). Co60 Gamma Spectrum.

ETM radionuclides

- Gamma-ray and X-ray emitters
- High LOD
- Precise calibration needed
- Possible to use for screening

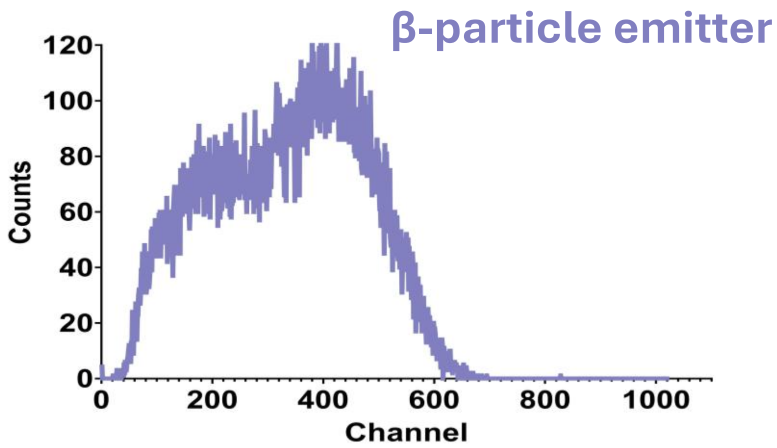
Difficult-to-measure radionuclides

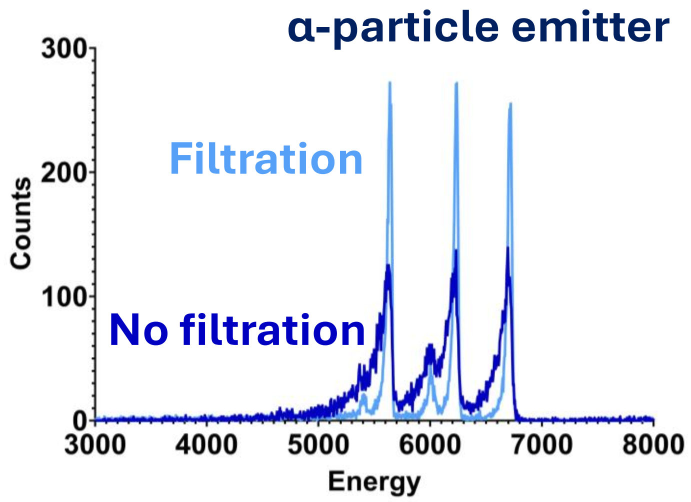
*“a radionuclide whose radioactivity is difficult to measure **directly from the outside** of the waste packages by non-destructive assay means”*

ISO standard 24390:2023

DTM radionuclides

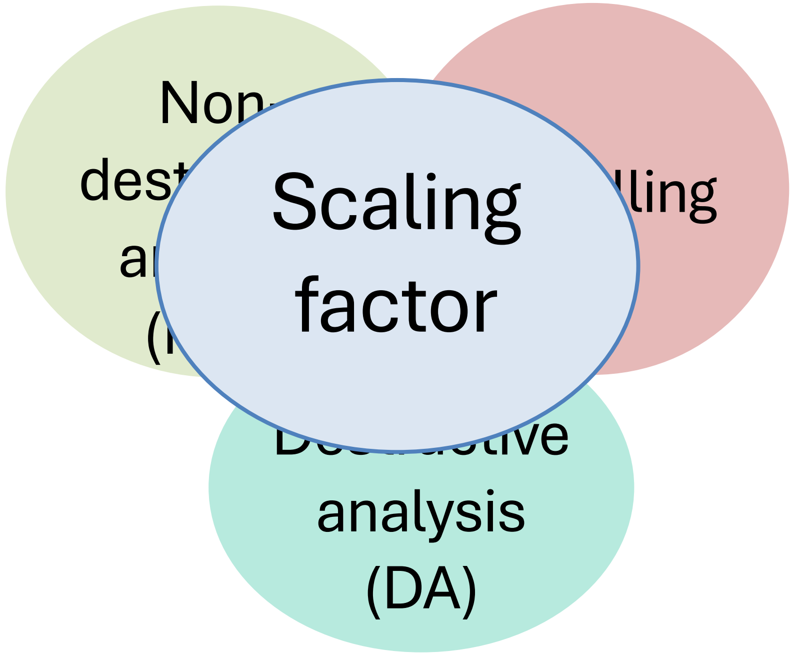
- Alpha and beta particle emitters
- Spectral overlap
- Sample treatment and chemical separation needed





Difficult-to-measure radionuclides

How is the activity of DTM radionuclides estimated?



Activity **ETM** radionuclides

γ-ray emitters

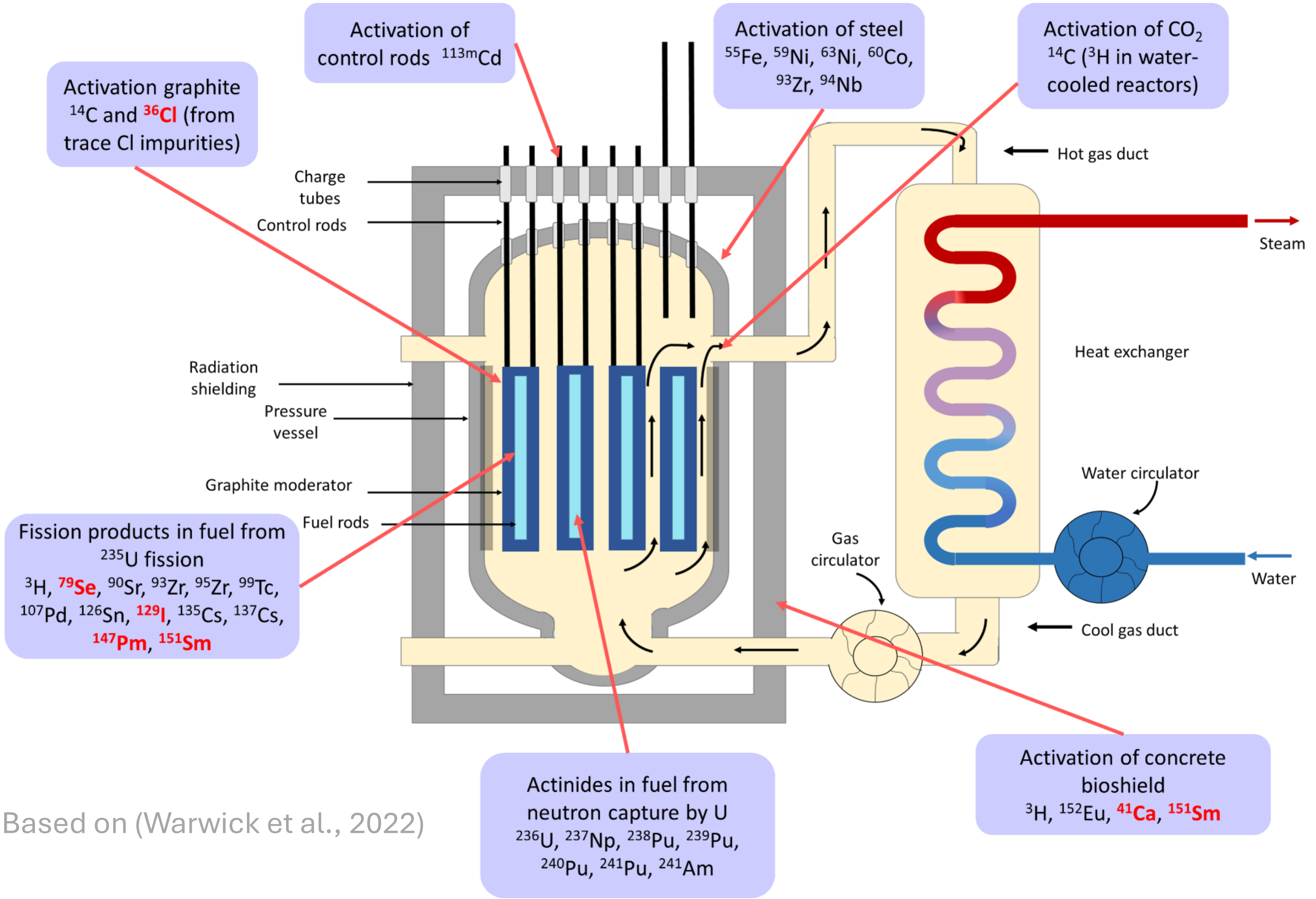
Activity **DTM** radionuclides

α and β particle emitters

Mathematical correlation

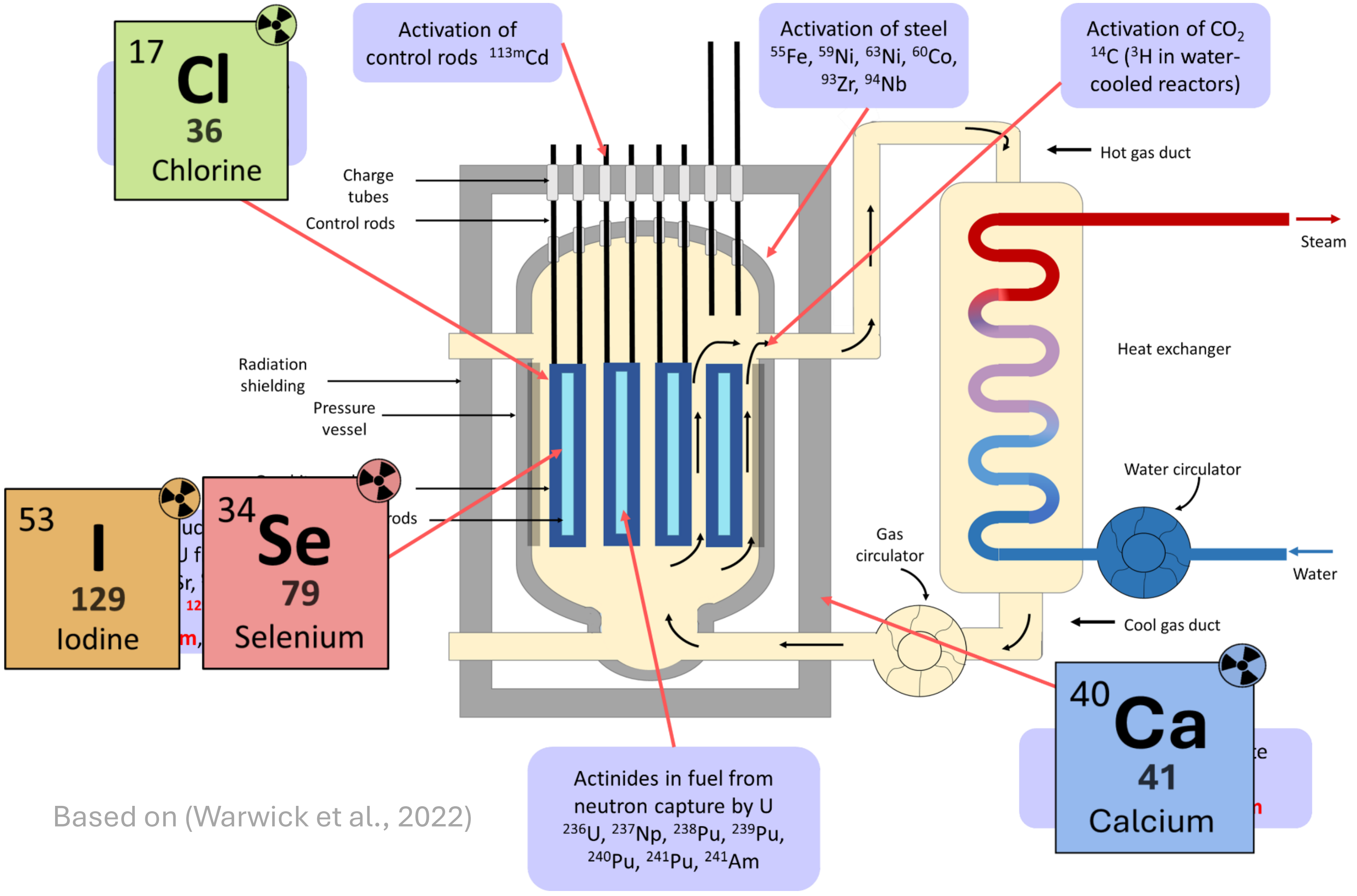
Validation using **experimental data**

Selection of DTM radionuclides



Based on (Warwick et al., 2022)

Selection of DTM radionuclides



Based on (Warwick et al., 2022)

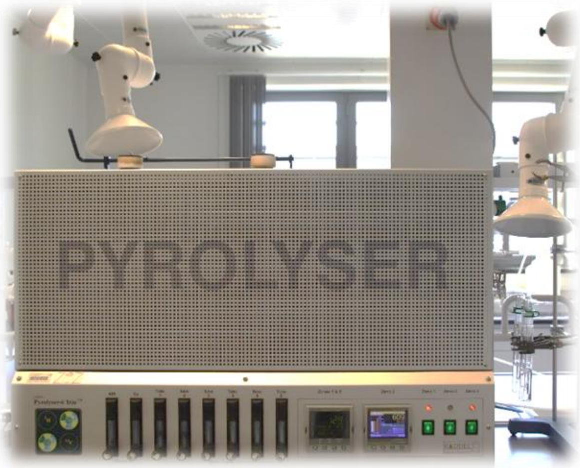
Determination of DTM radionuclides

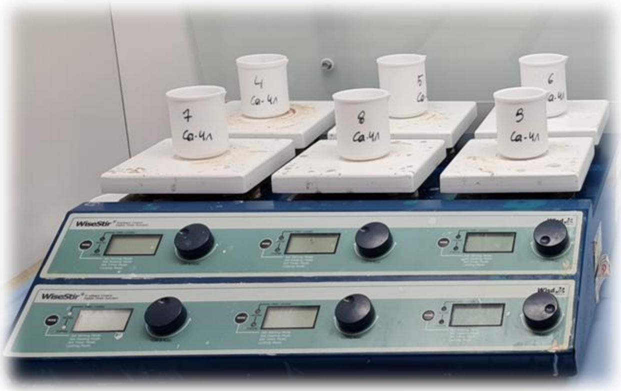
Sample treatment → Chemical separation → Measurement

Homogenization

Complete sample dissolution

Sample representativeness





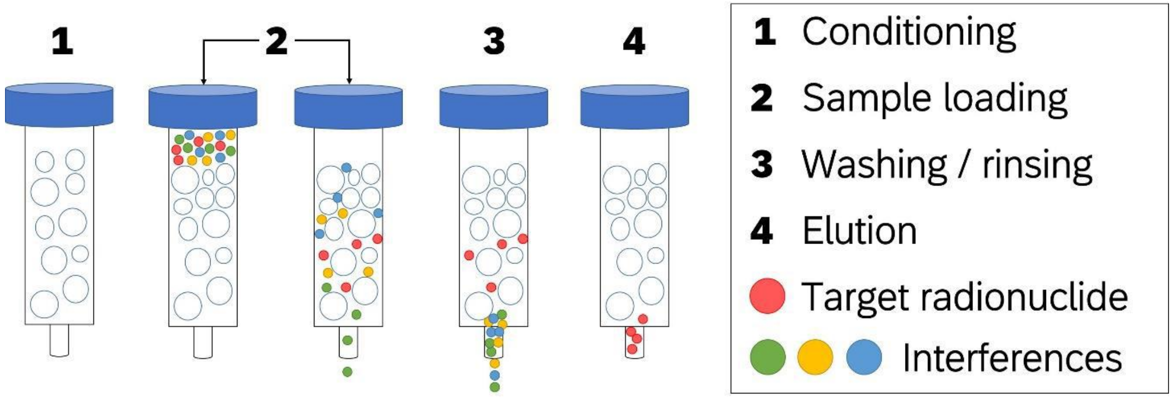
Determination of DTM radionuclides

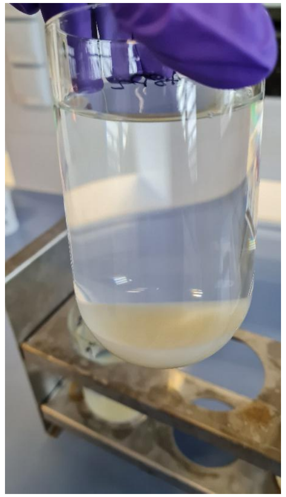
Sample treatment → **Chemical separation** → Measurement

Pre-concentration

Interference removal

Time needed for the separation procedure





Determination of DTM radionuclides

Sample treatment

Chemical separation

Measurement

Detection limit

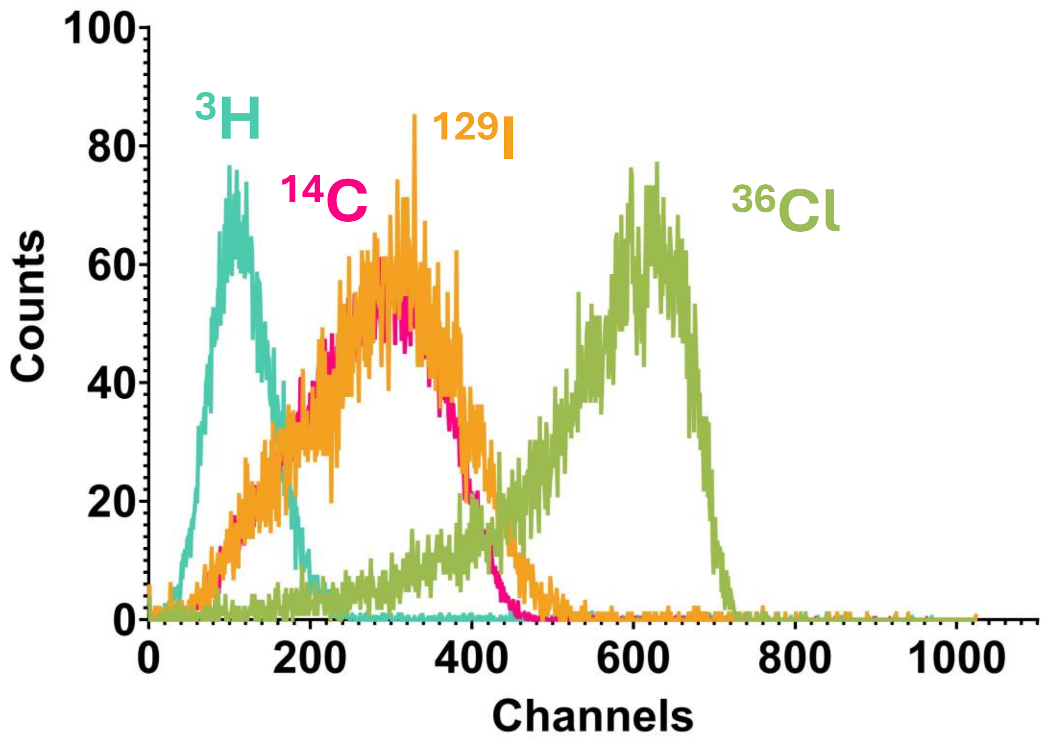
Effect of interferences

Time needed for measurement

Main challenges

Spectral interferences

17 Cl	53 I	34 Se	40 Ca
36	129	79	41
Chlorine	Iodine	Selenium	Calcium



Main challenges

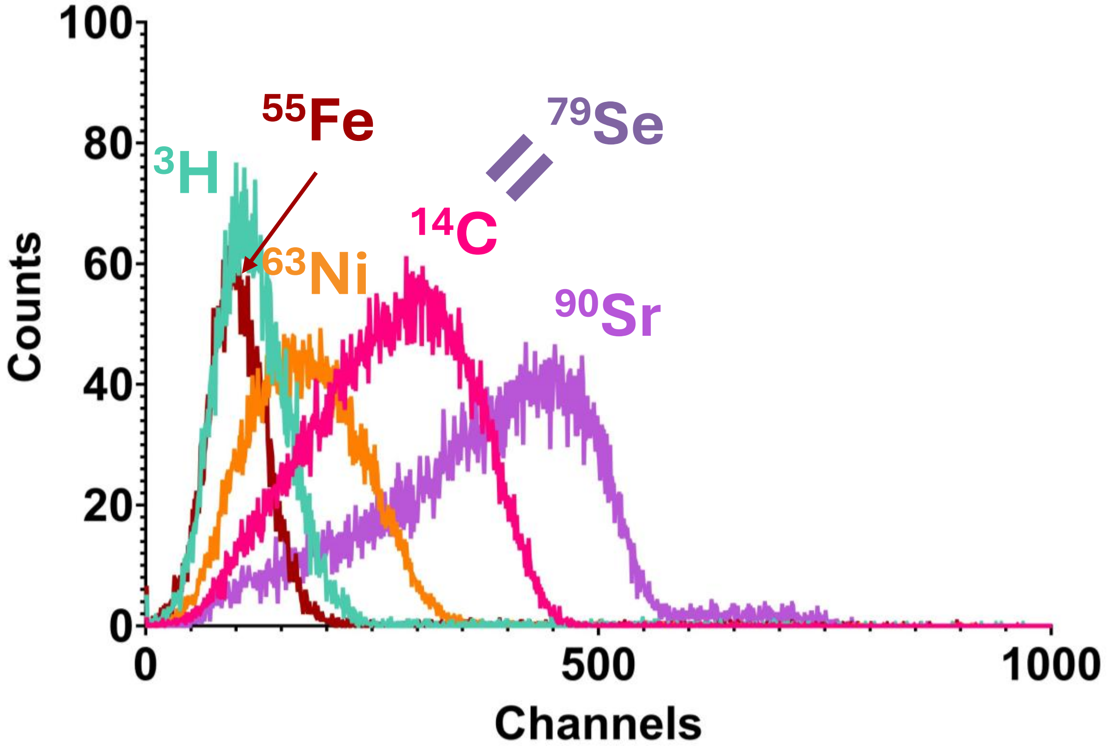
Spectral interferences

17 **Cl** 36 Chlorine

53 **I** 129 Iodine

34 **Se** 79 Selenium

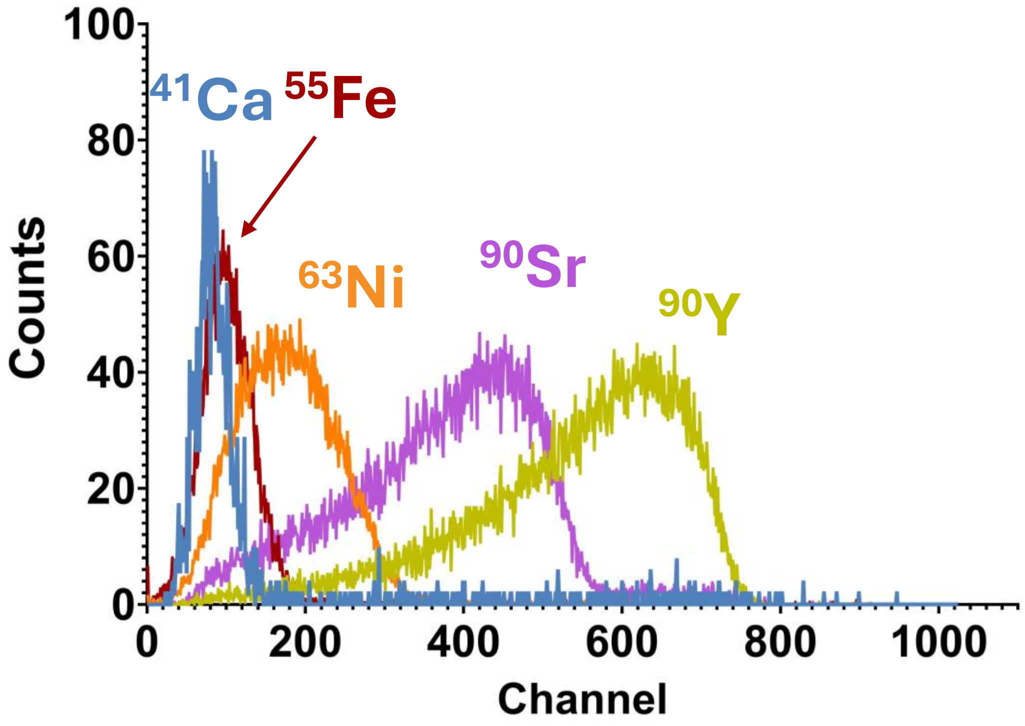
40 **Ca** 41 Calcium



Main challenges

Spectral interferences

17 **Cl** 36 Chlorine	53 **I** 129 Iodine	34 **Se** 79 Selenium	40 **Ca** 41 Calcium



Main challenges

Isobaric interferences

17 Cl 36 Chlorine	53 I 129 Iodine	34 Se 79 Selenium	40 Ca 41 Calcium
^{36}Ar ^{36}S	^{129}Xe ^{128}TeH	^{79}Br	^{41}K
^{35}Cl ^{37}Cl	^{127}I	^{78}Se	^{40}Ca
$^{18}O_2$	$^{127}IH_2$	$^{40}Ar^{39}K$ ^{78}SeH $^{40}Ca^{39}Cl$	^{40}CaH ^{40}ArH

Need to perform chemical separation before sample measurement

Main challenges

Reference material

17 Cl 36 Chlorine

✓ Standard solution

✗ No matrix RMs (soils, concretes, or leachates) certified for ^{36}Cl

53 I 129 Iodine

✓ IAEA materials containing ^{129}I but not certified

? Few matrix RMs

34 Se 79 Selenium

✗ No certified reference materials

✗ Very limited reference solutions

40 Ca 41 Calcium

? Few AMS isotopic standards

✗ No certified reference materials

Main challenges

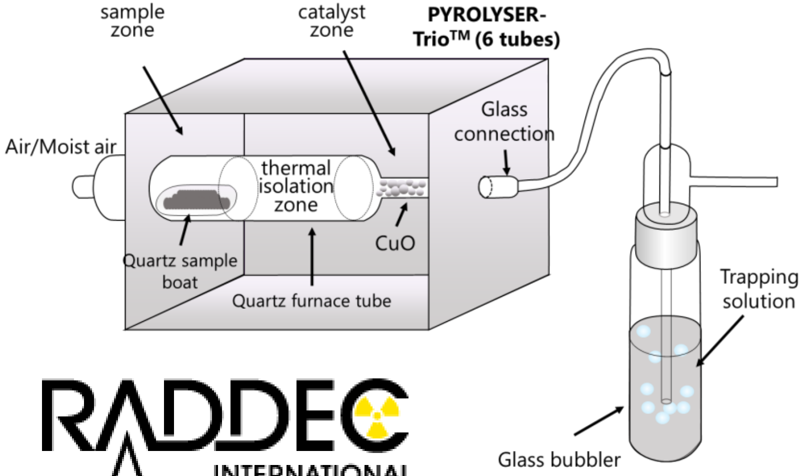
Turnaround time (TAT) and cost of the procedure

Sample decomposition	Chemical separation	Measurement
Expensive instruments	Chemical volume	Expensive instruments
Chemical needed	Amount of resin/cartridges	Sample source preparation
	Flow rates/steps involved	

^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

Pyrolisis (volatile elements)

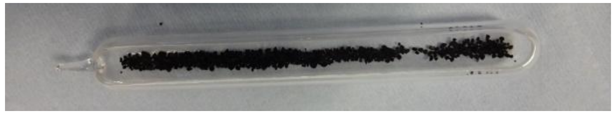


RADDEC INTERNATIONAL

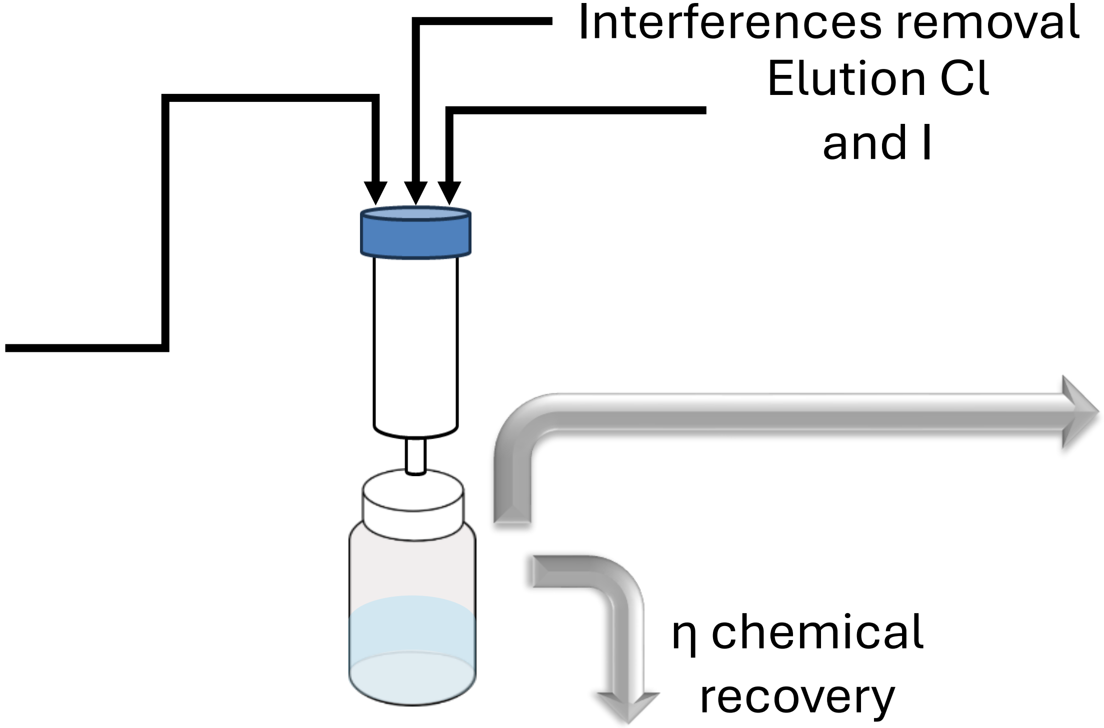
Based on Warwick et al. 2010

Concrete

Graphite



Chemical separation

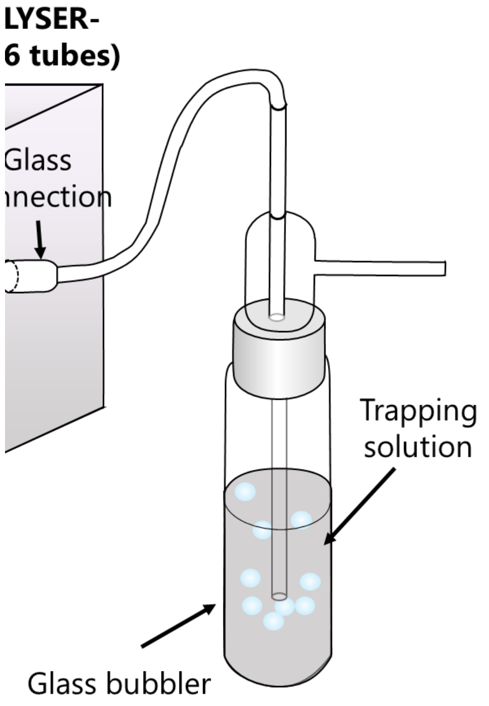


ICP-MS

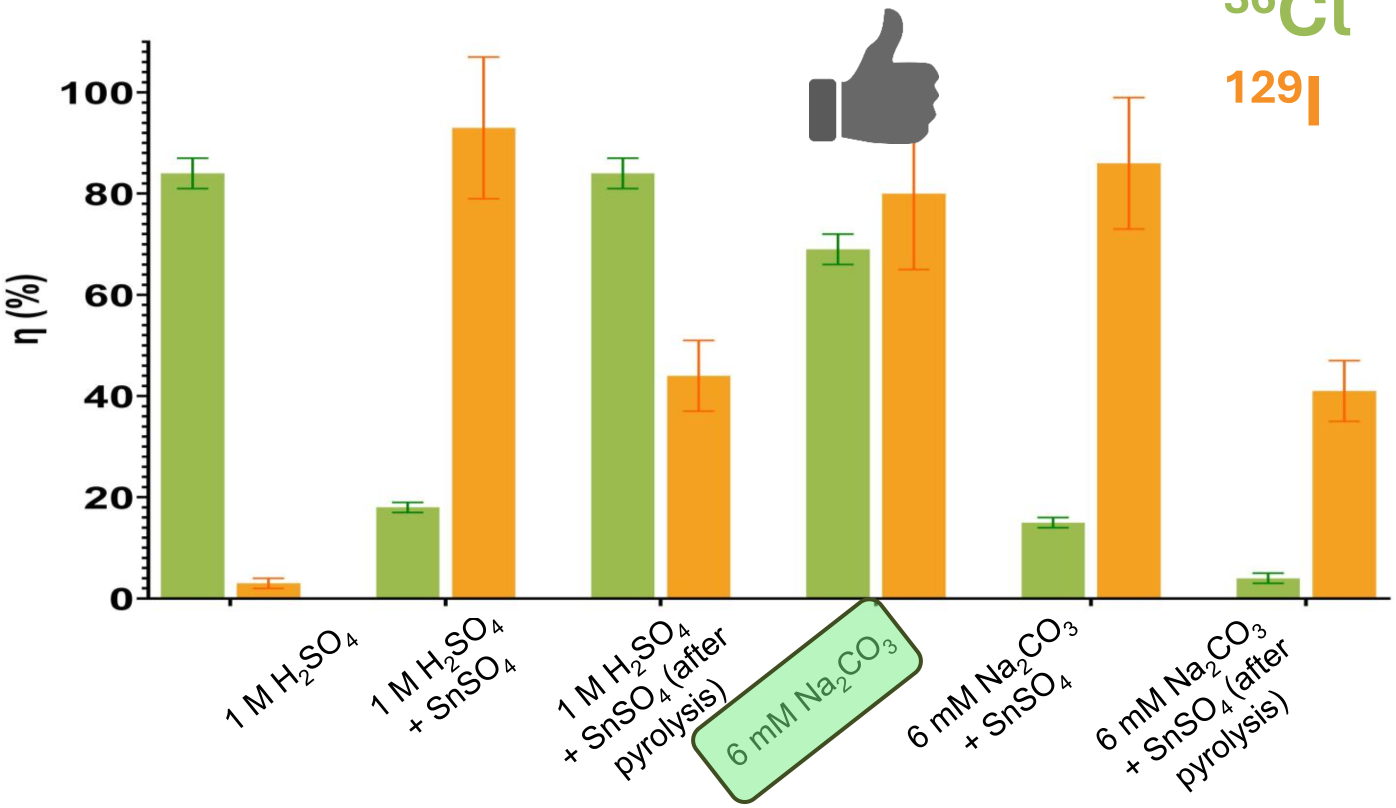
LSC

Wallac Quantulus 1220™

^{36}Cl and ^{129}I determination



Suitable trapping solution



^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine

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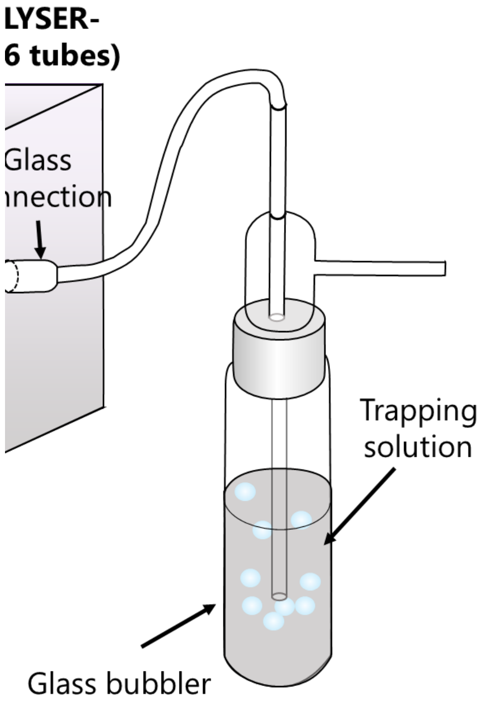
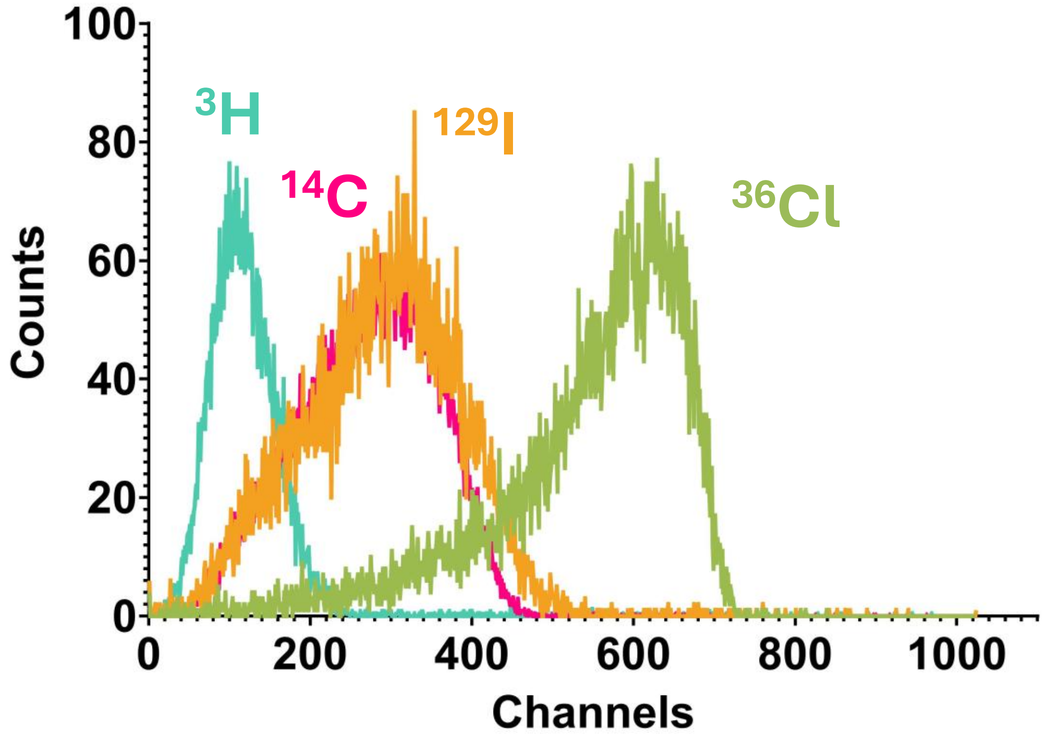
Cl and I chemical separation



Volatile elements collected



^{36}Cl and ^{129}I determination

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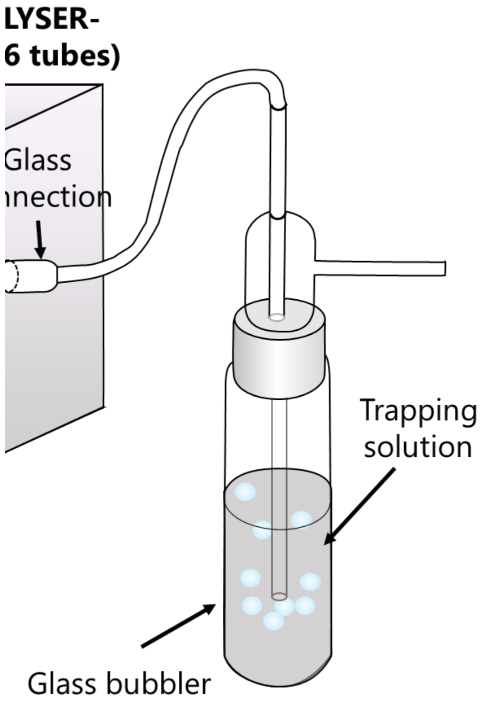

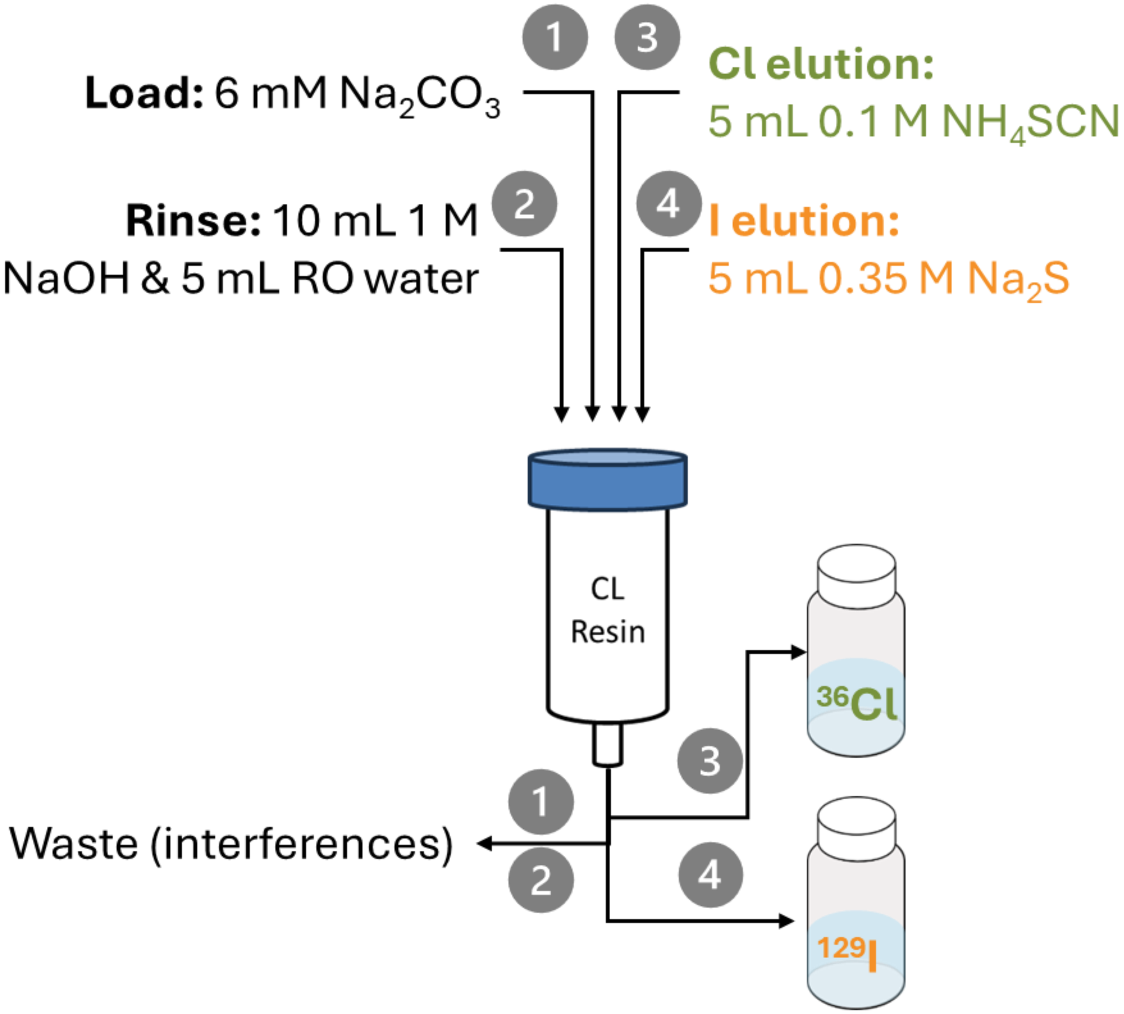

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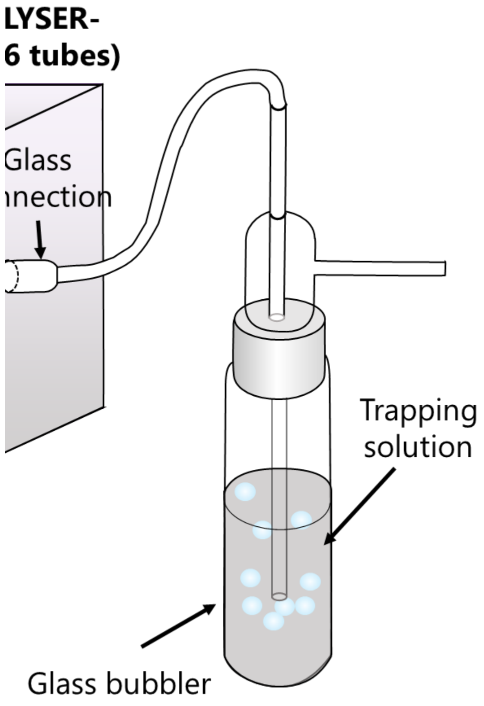
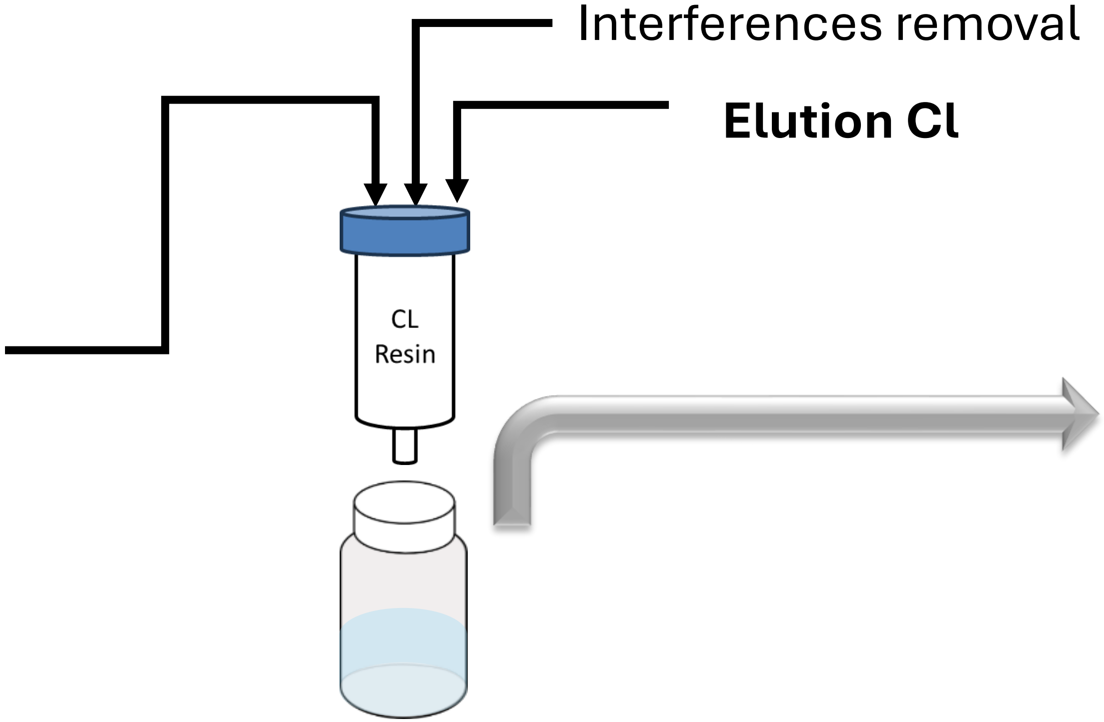


Chemical separation



LSC

Wallac Quantulus 1220™

Procedural blank samples

Spiked ^{36}Cl / ^{129}I — Blank — Spiked ^{36}Cl / ^{129}I

^{36}Cl and ^{129}I determination

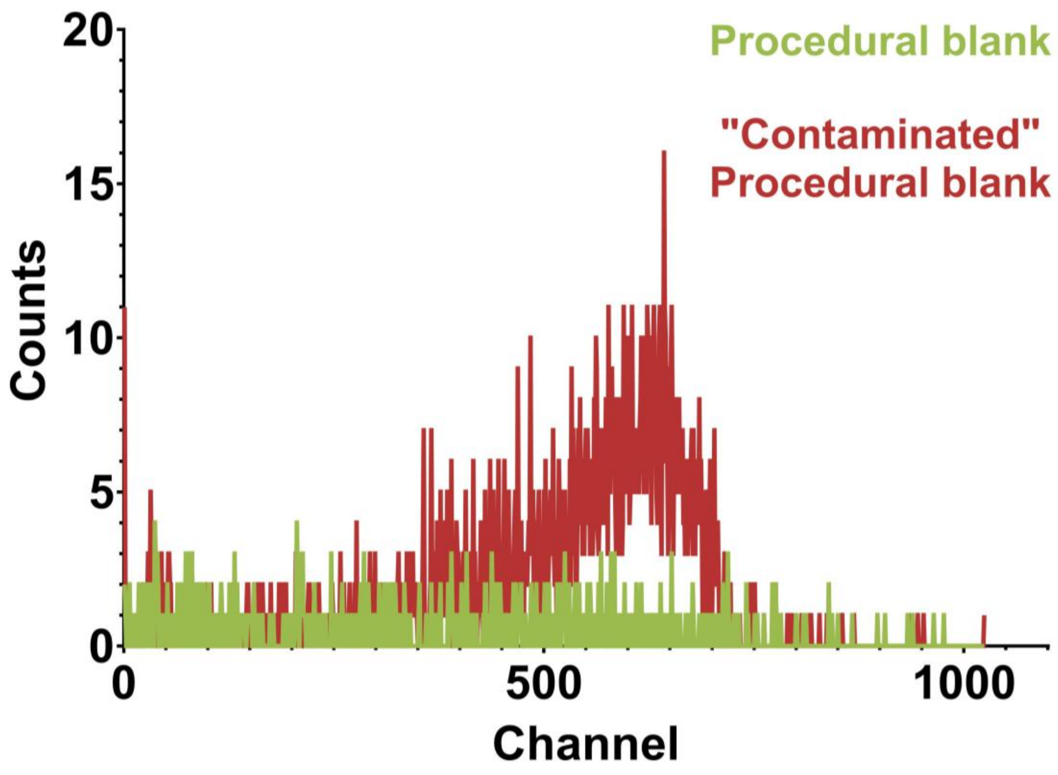
17 Cl 36 Chlorine

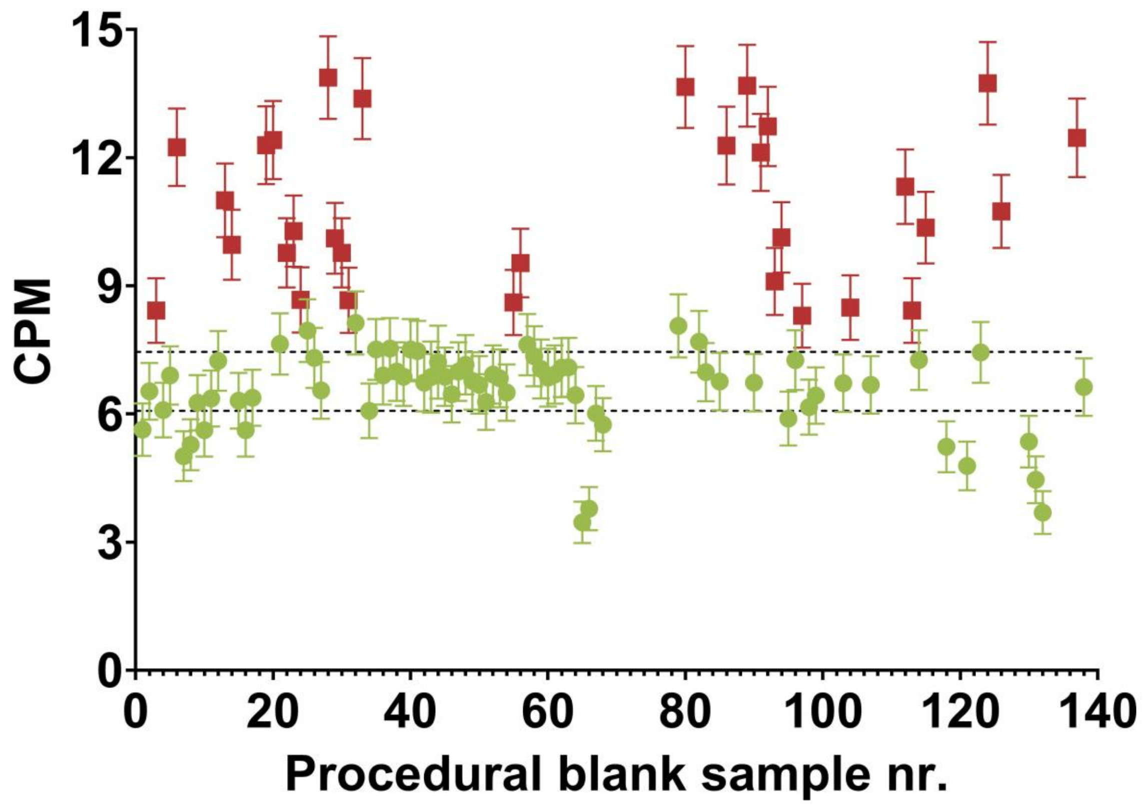
53 I 129 Iodine

34 Se 79 Selenium

40 Ca 41 Calcium

^{36}Cl detected on procedural blank samples



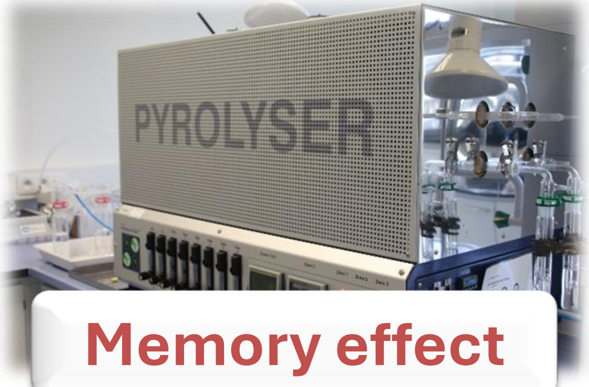


^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine	53 I 129 Iodine
34 Se 79 Selenium	40 Ca 41 Calcium

Where does ^{36}Cl comes from?

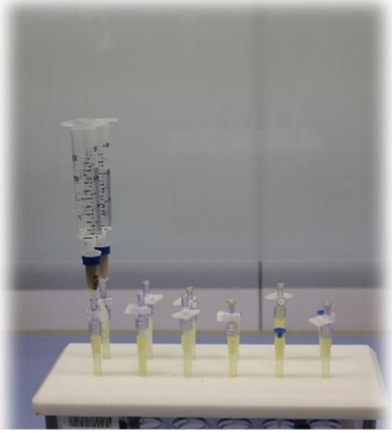
1. Sample combustion



Memory effect

Pyrolyser

2. Separation



Cross-contamination

CL Resin

3. Measurement

Liquid Scintillation Counting (LSC)

^{36}Cl and ^{129}I determination

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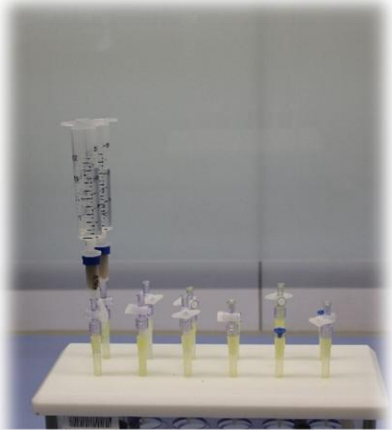
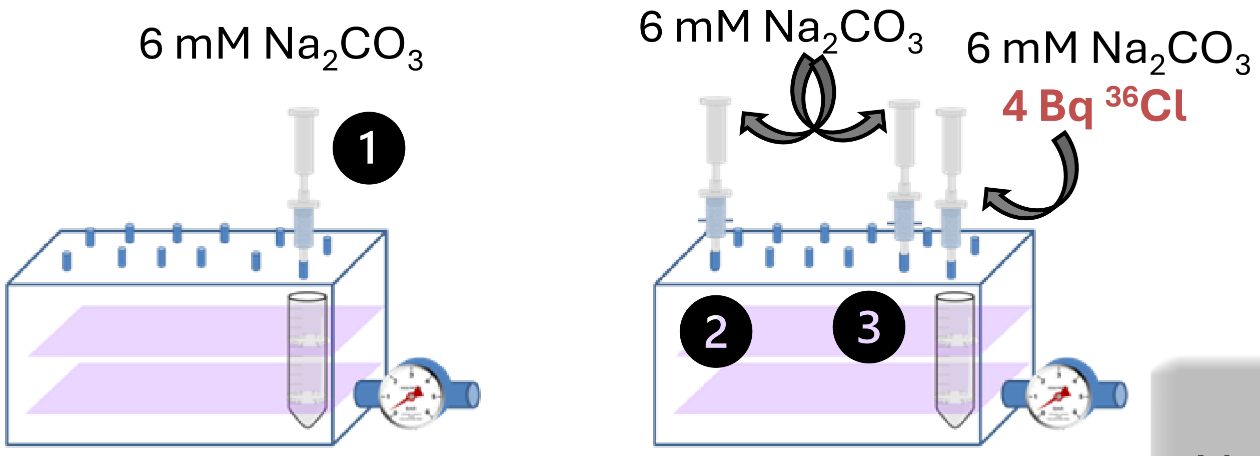
Where does ^{36}Cl comes from?

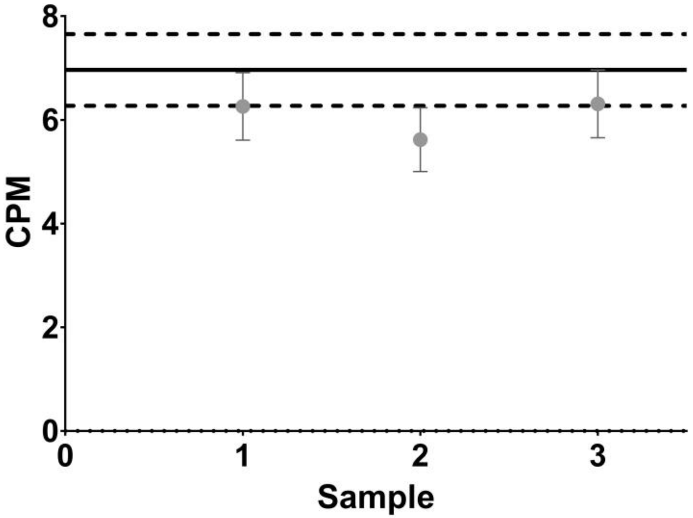
2. Separation



Cross-contamination

CL Resin





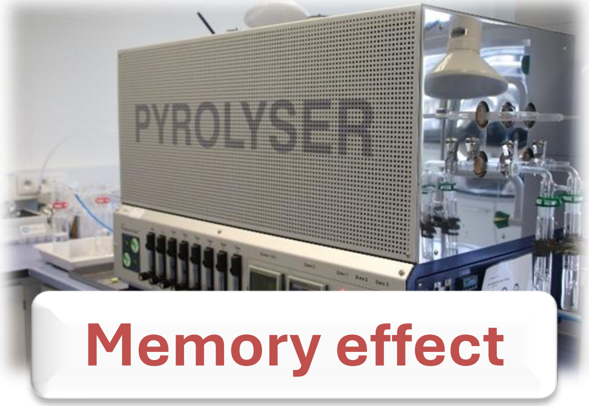
No ^{36}Cl detected
No differences in blanks

^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

Where does ^{36}Cl comes from?

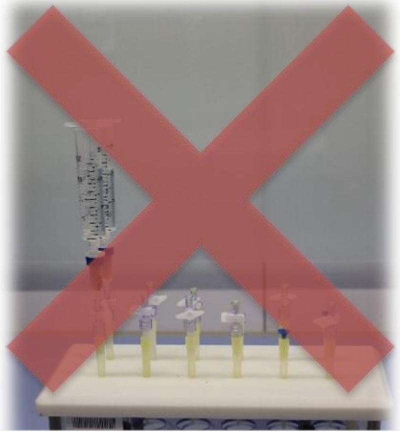
1. Sample combustion



Memory effect

Pyrolyser

2. Separation



Cross-contamination

CL Resin

3. Measurement

Liquid Scintillation Counting (LSC)

^{36}Cl and ^{129}I determination

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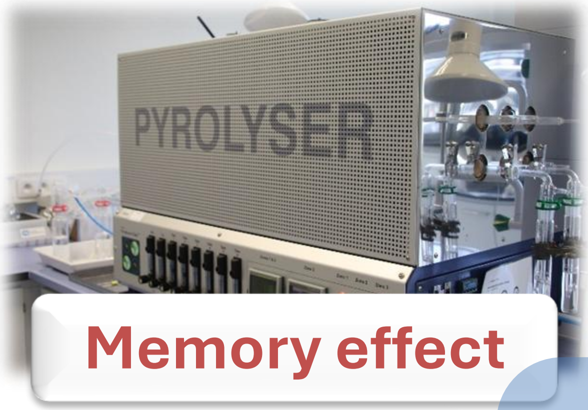
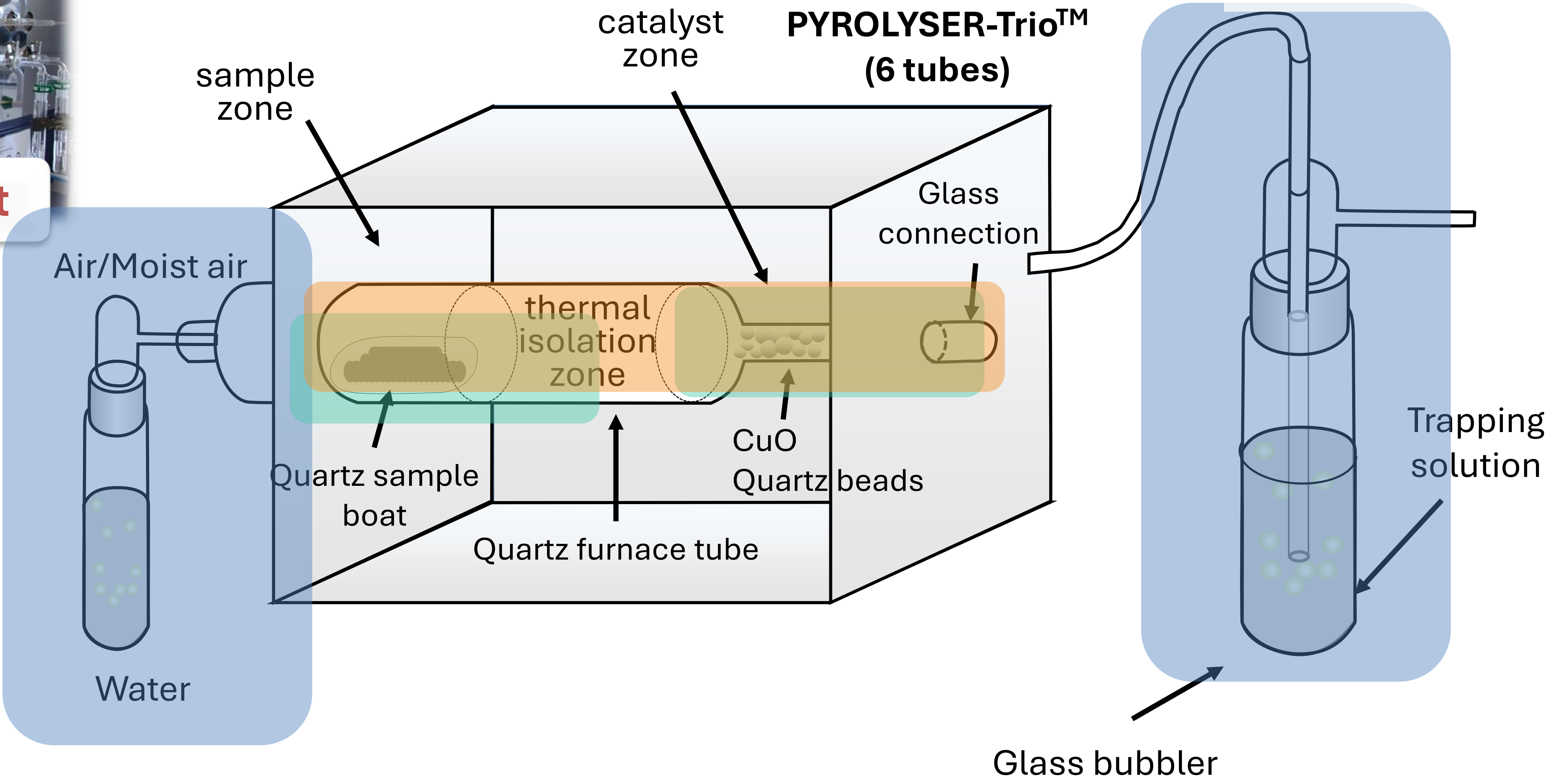
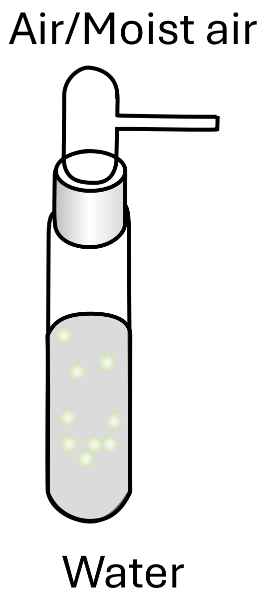


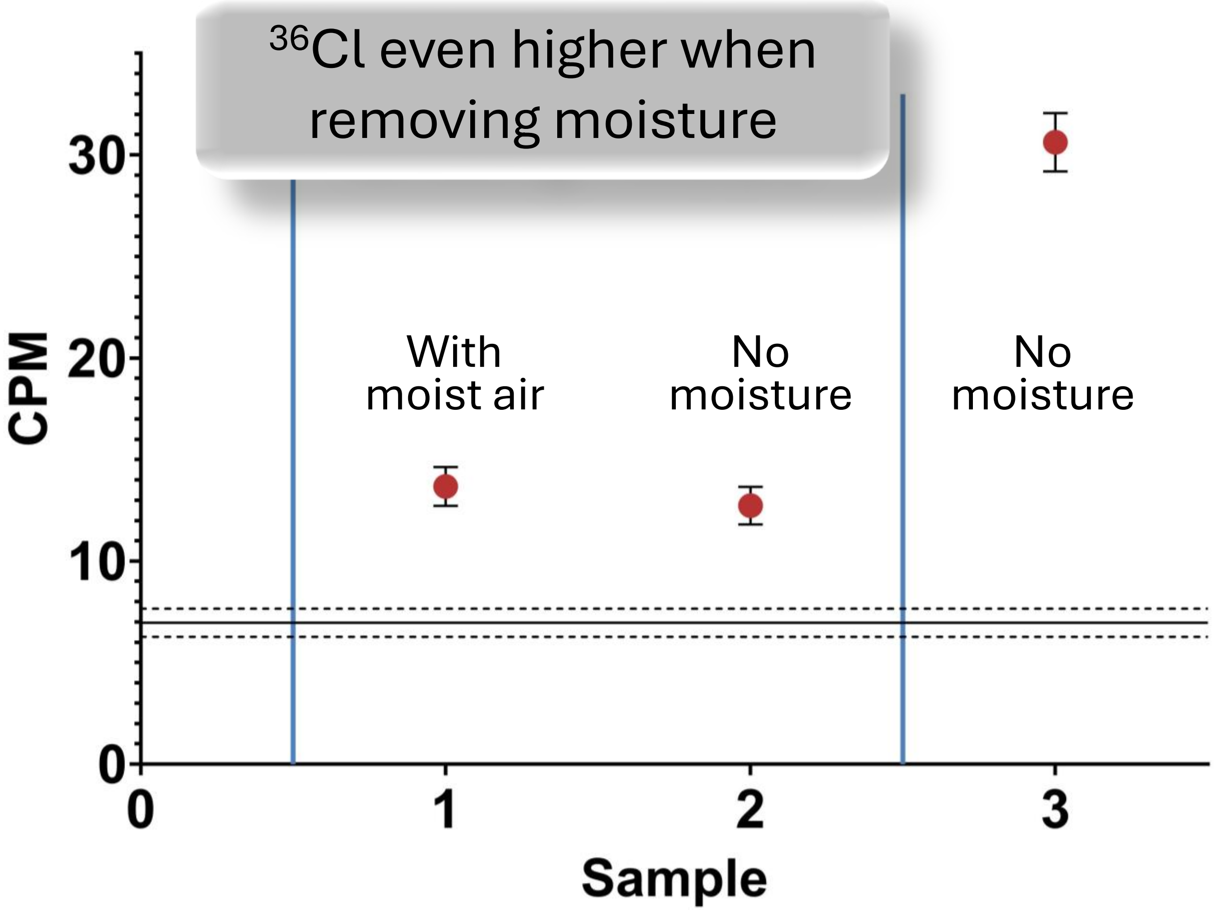
Pyrolyser



^{36}Cl and ^{129}I determination

1. Spiked 4 Bq ^{36}Cl
2. BLANK
3. Spiked 4 Bq ^{36}Cl
4. BLANK





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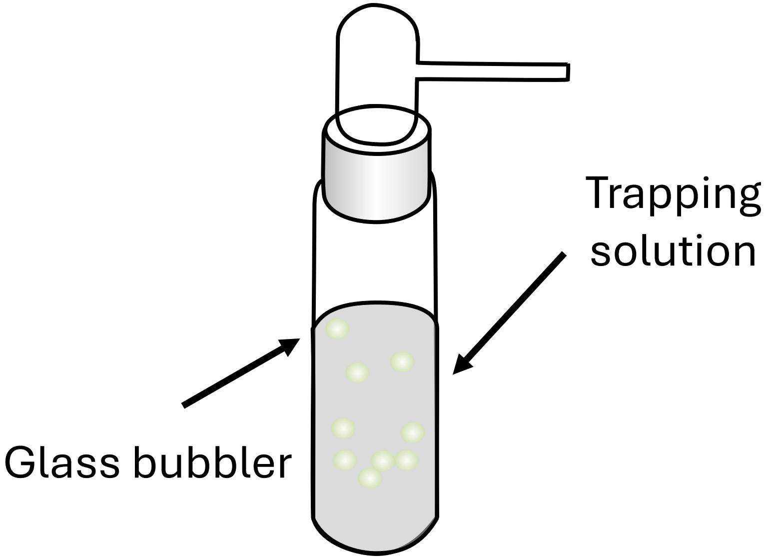

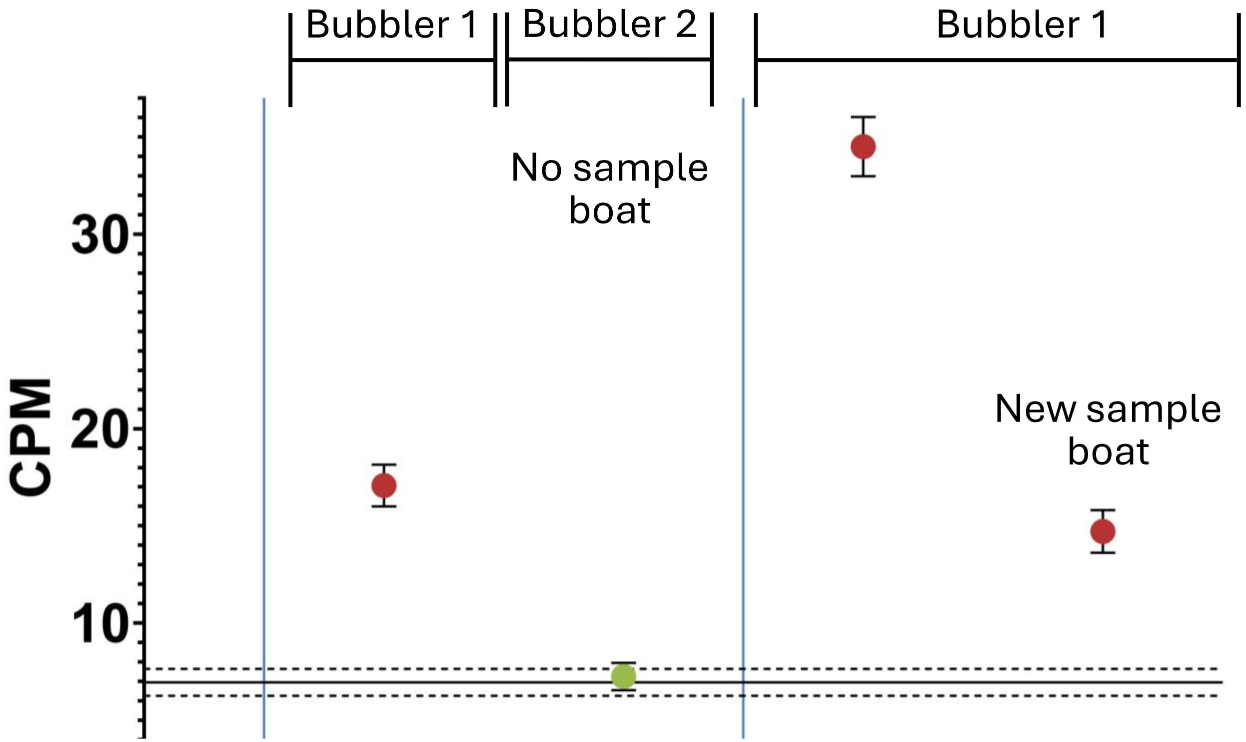

Glass bubblers contributed to ^{36}Cl memory effect
Not the only contributors

^{36}Cl and ^{129}I determination

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3. Spiked 4 Bq ^{36}Cl
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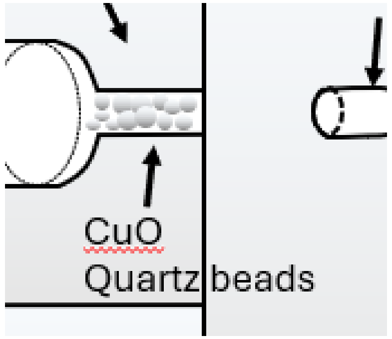
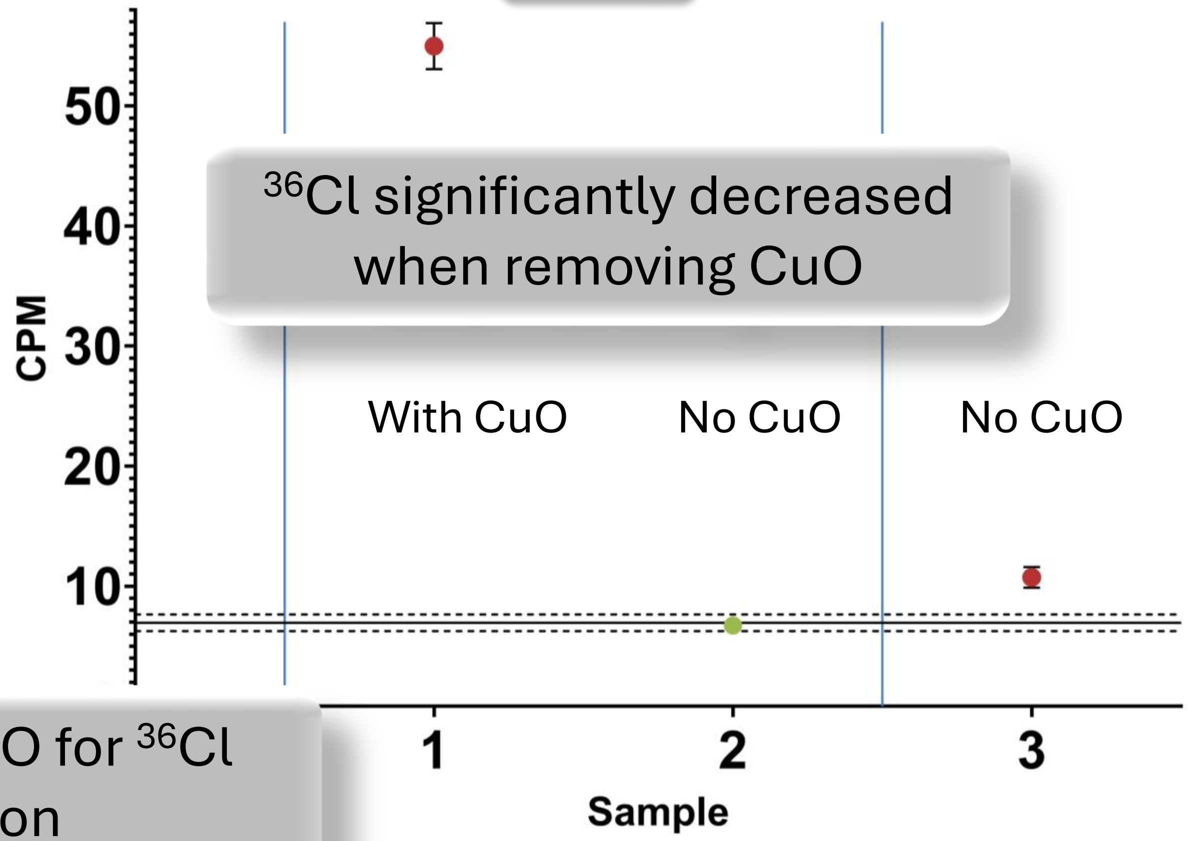


CuO



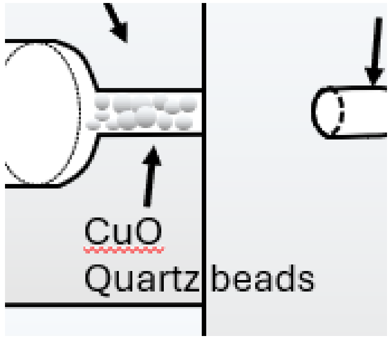
^{36}Cl significantly decreased when removing CuO

No need to use CuO for ^{36}Cl determination

^{36}Cl and ^{129}I determination

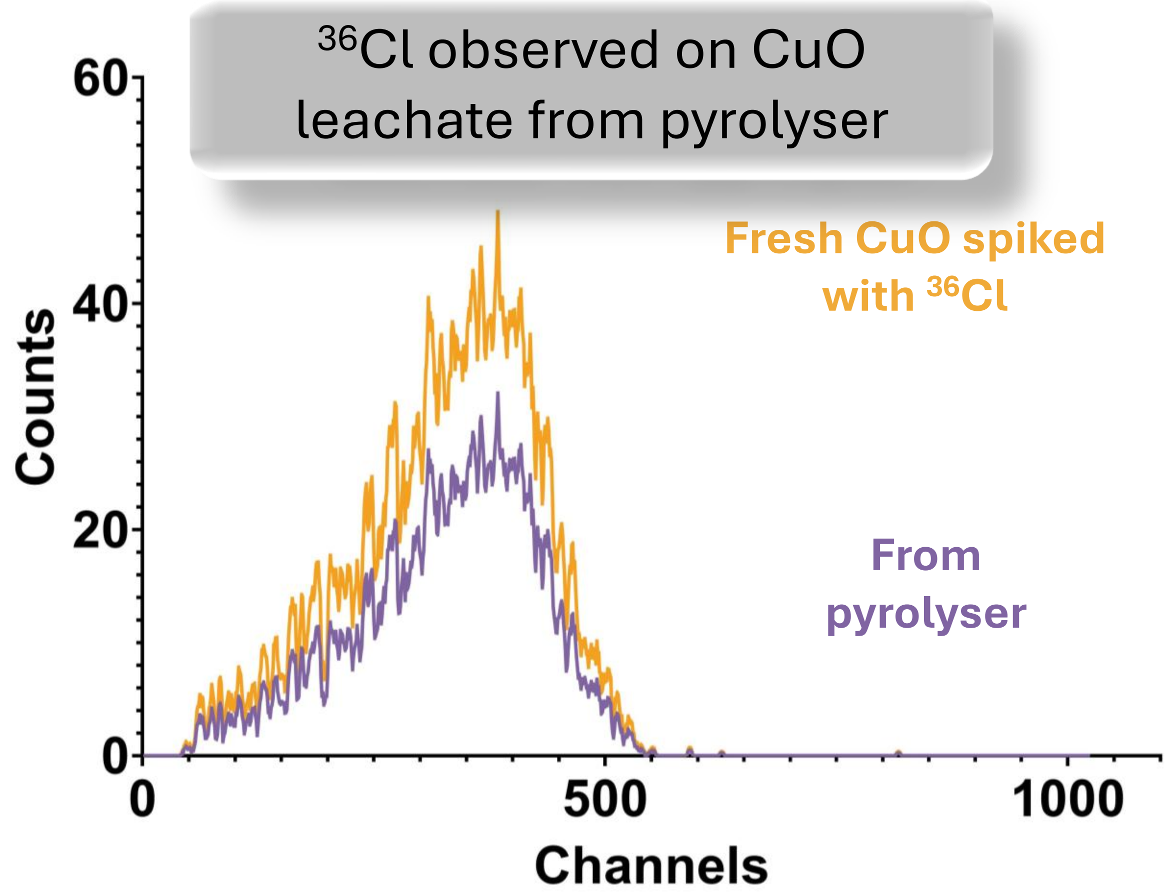
17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

1. Spiked 4 Bq ^{36}Cl
2. BLANK
3. Spiked 4 Bq ^{36}Cl
4. BLANK

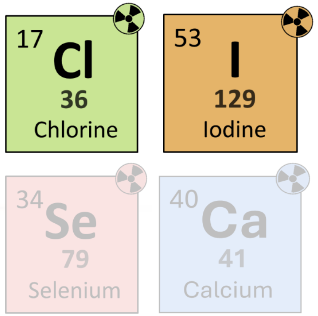


From pyrolyser

Fresh CuO spiked with ^{36}Cl



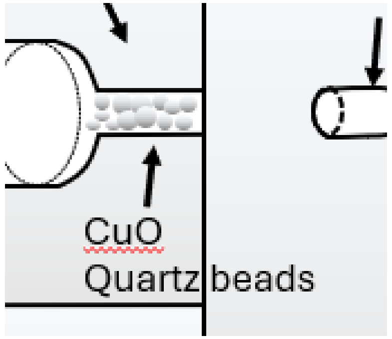
^{36}Cl and ^{129}I determination

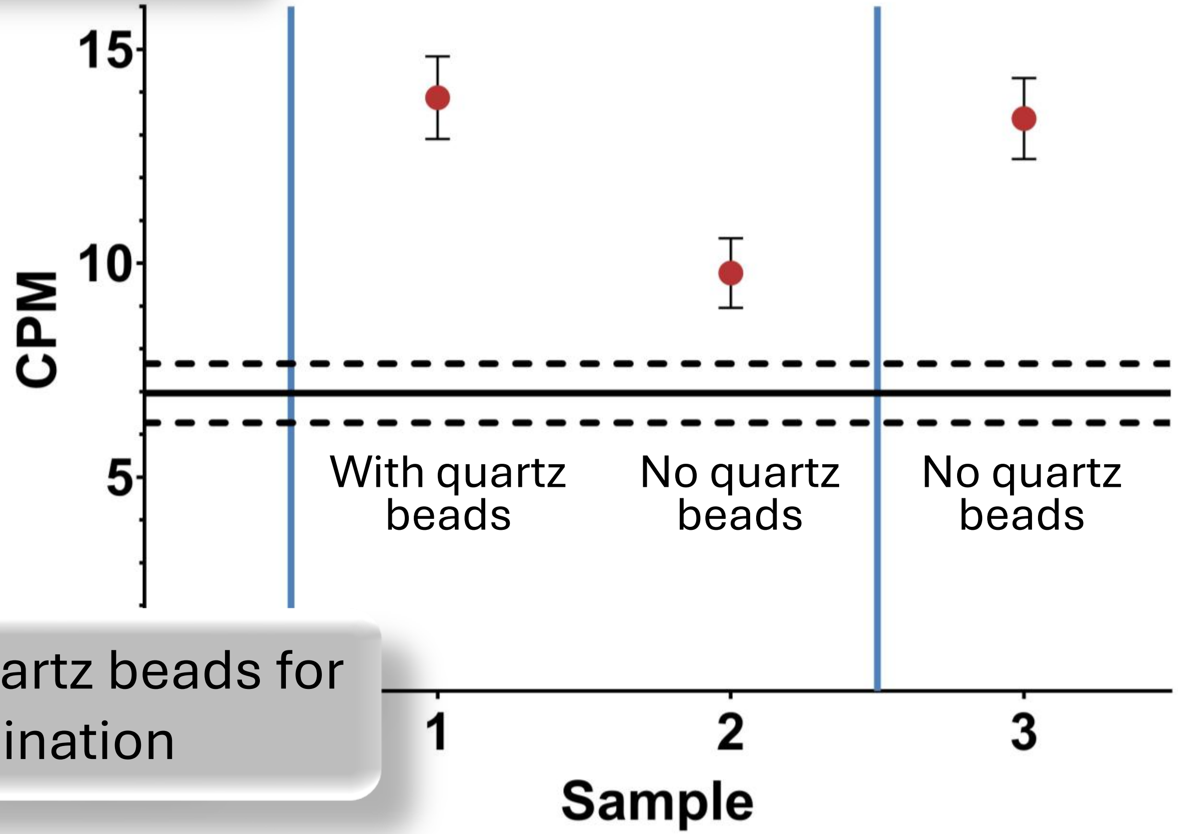


1. Spiked 4 Bq ^{36}Cl
2. BLANK
3. Spiked 4 Bq ^{36}Cl
4. BLANK

^{36}Cl decreased when removing quart beads

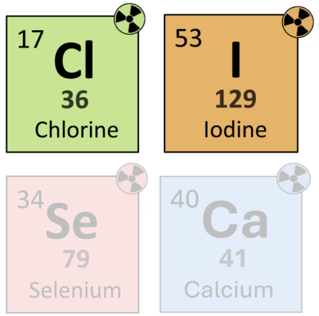
Quartz beads



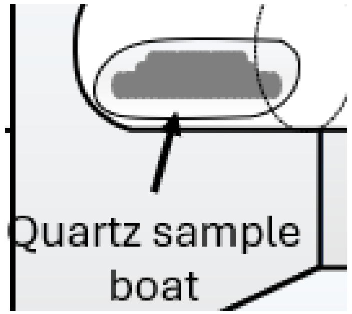


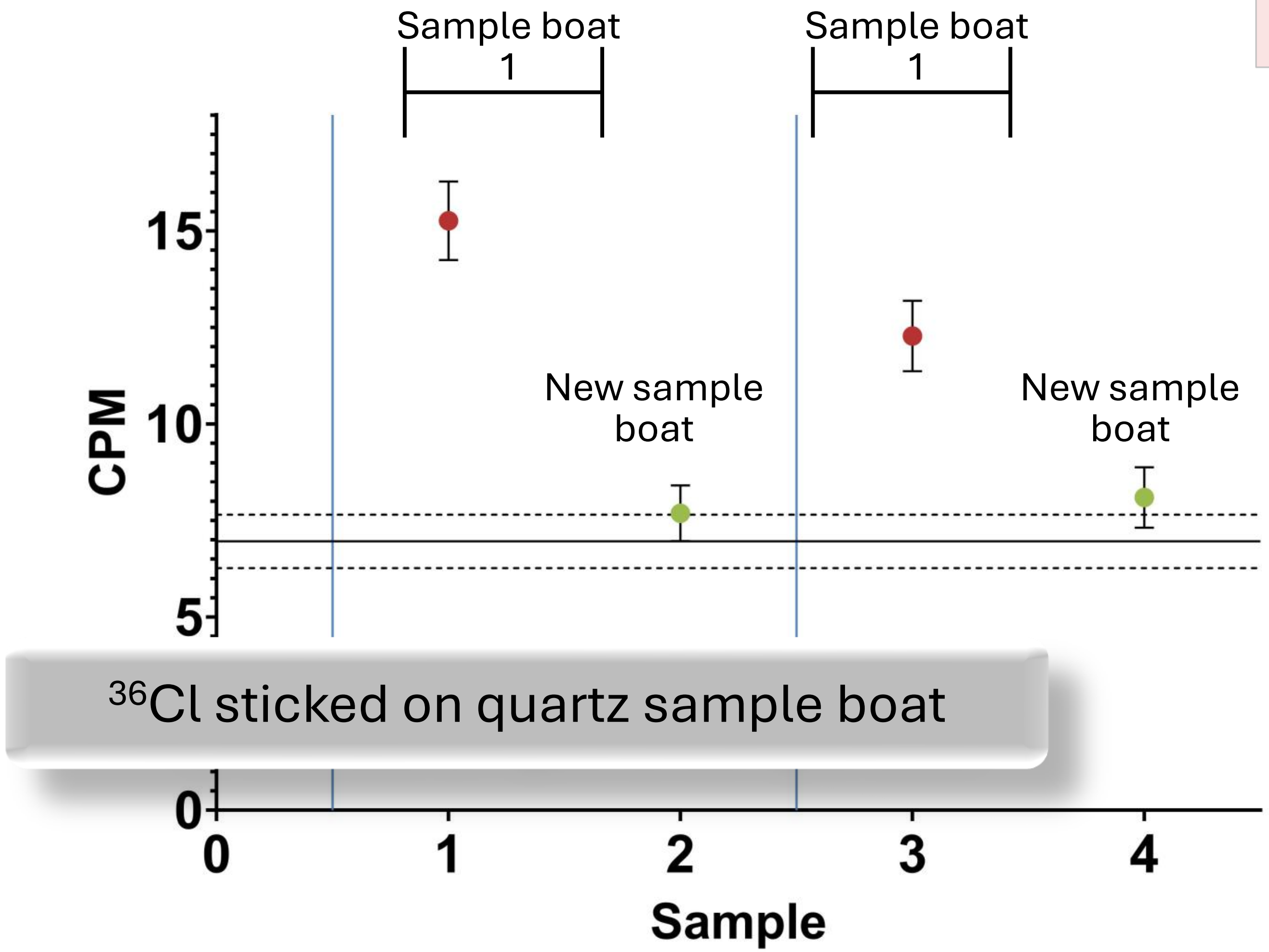
No need to use quartz beads for ^{36}Cl determination

^{36}Cl and ^{129}I determination

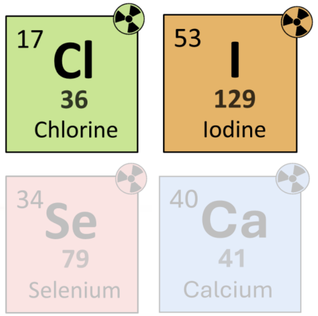


1. Spiked 4 Bq ^{36}Cl
2. BLANK
3. Spiked 4 Bq ^{36}Cl
4. BLANK





^{36}Cl and ^{129}I determination



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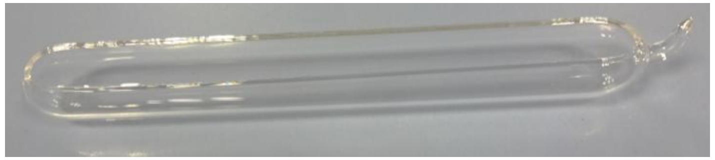
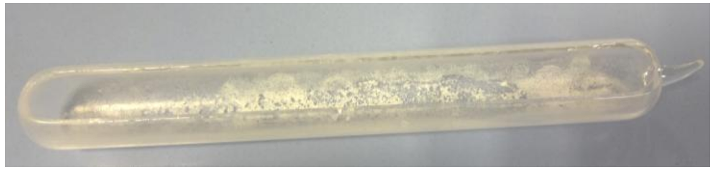
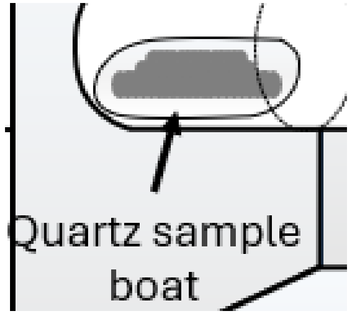
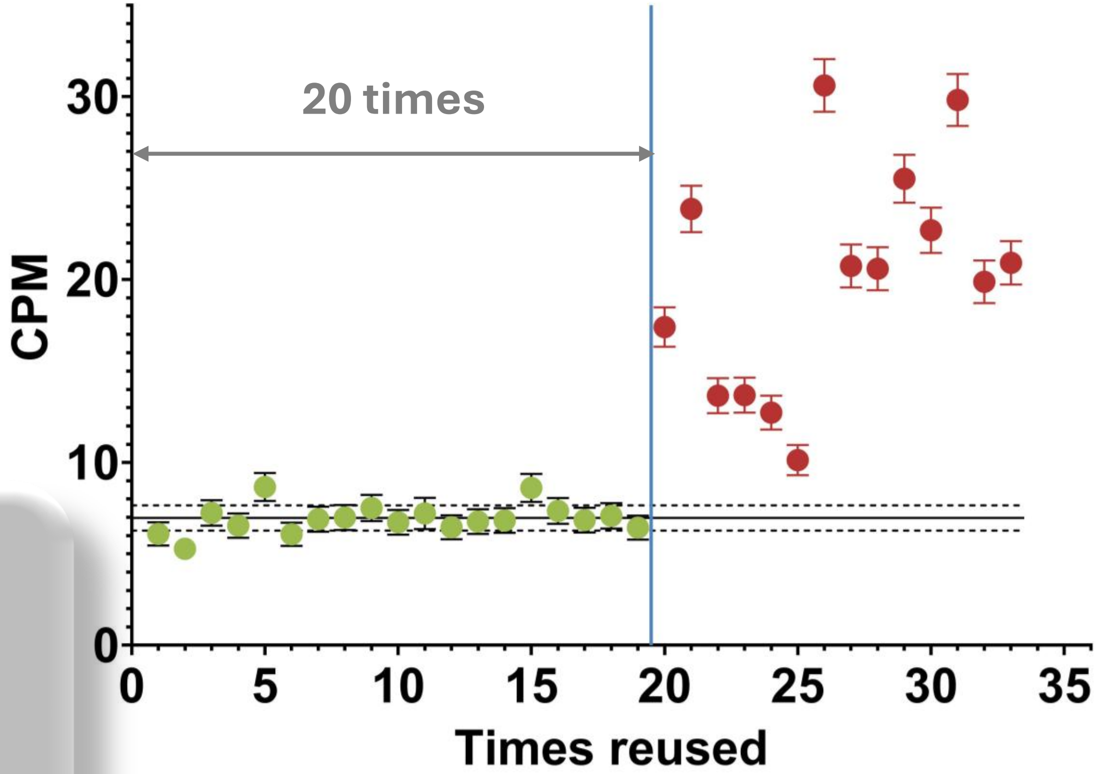




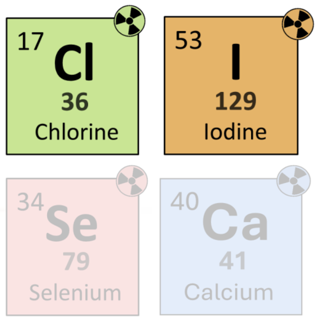


Ageing effect observed on quartz sample boats

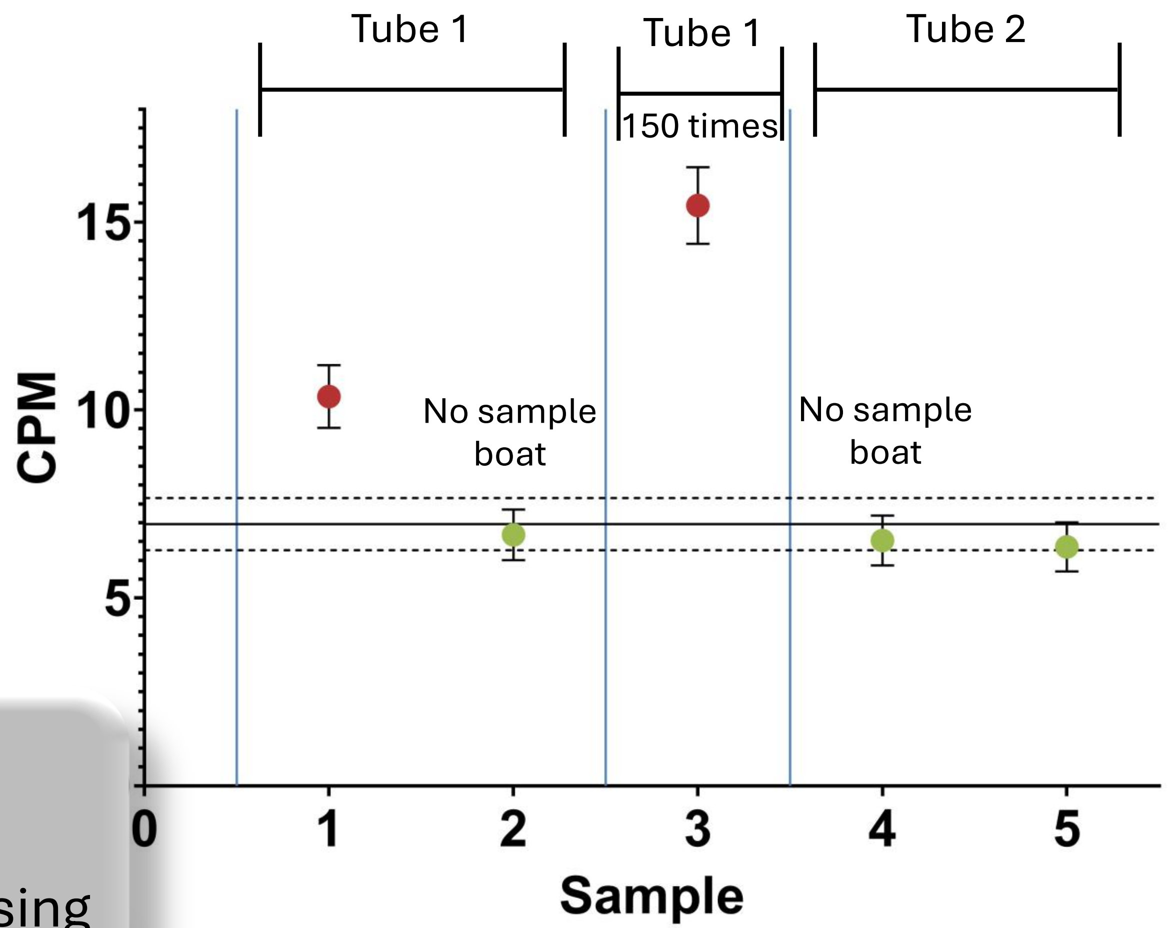
^{36}Cl measured after **20 times** reusing the materials

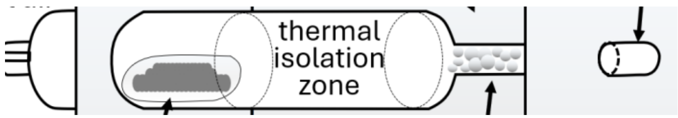


^{36}Cl and ^{129}I determination



1. Spiked 4 Bq ^{36}Cl
2. BLANK
3. Spiked 4 Bq ^{36}Cl
4. BLANK





Ageing effect observed on quartz tubes

^{36}Cl measured after **150 times** reusing the materials

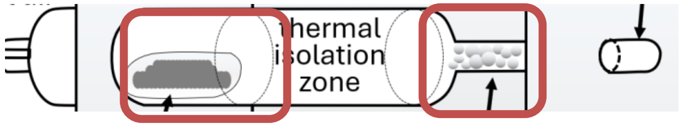
^{36}Cl and ^{129}I determination

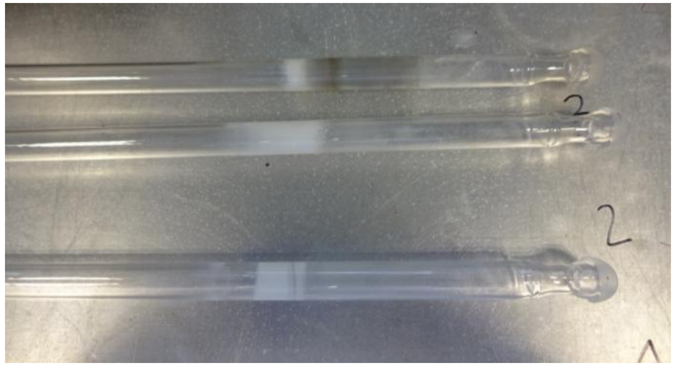
17 Cl 36 Chlorine

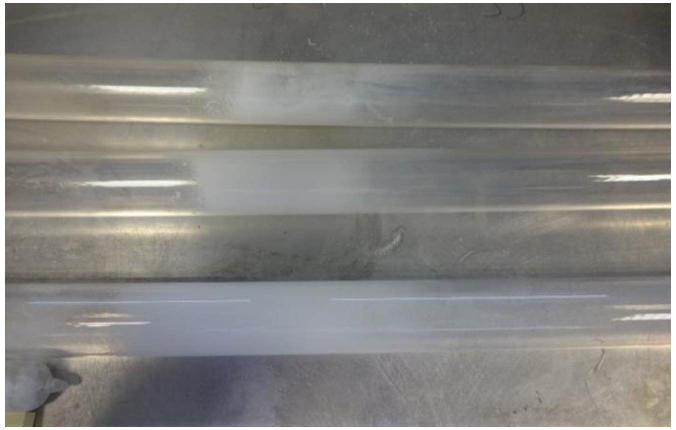
53 I 129 Iodine

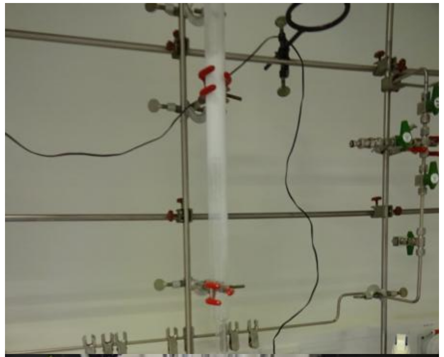
34 Se 79 Selenium

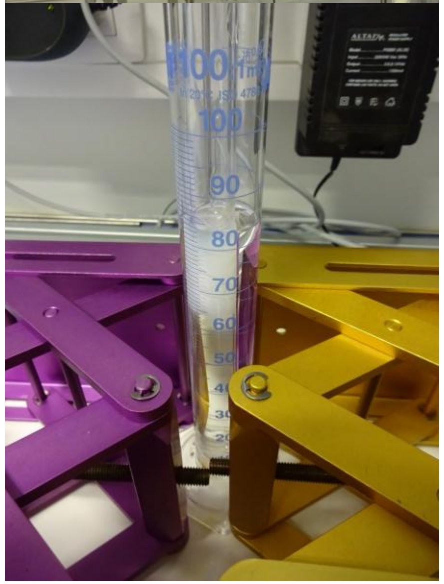
40 Ca 41 Calcium











Used for 163 experiments with ^{36}Cl-containing samples

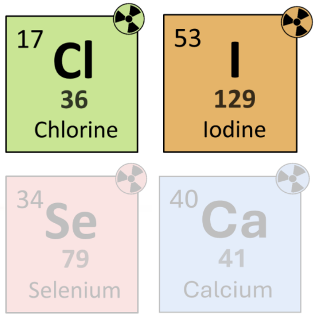
Addition $AgNO_3$

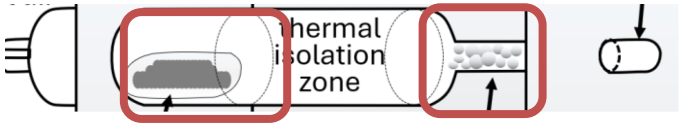
AgCl(s)

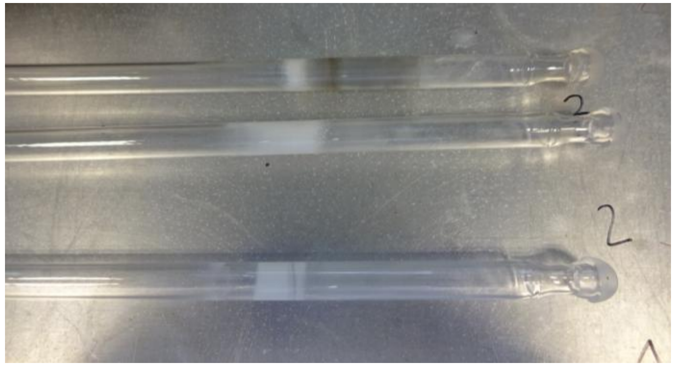
7 mL NH_3

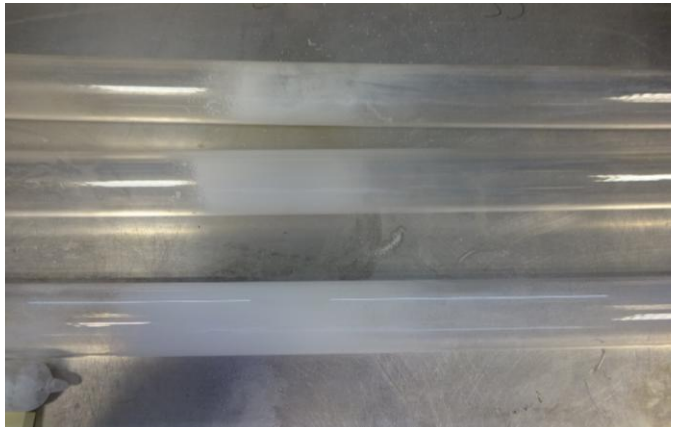
LSC measurement

^{36}Cl and ^{129}I determination

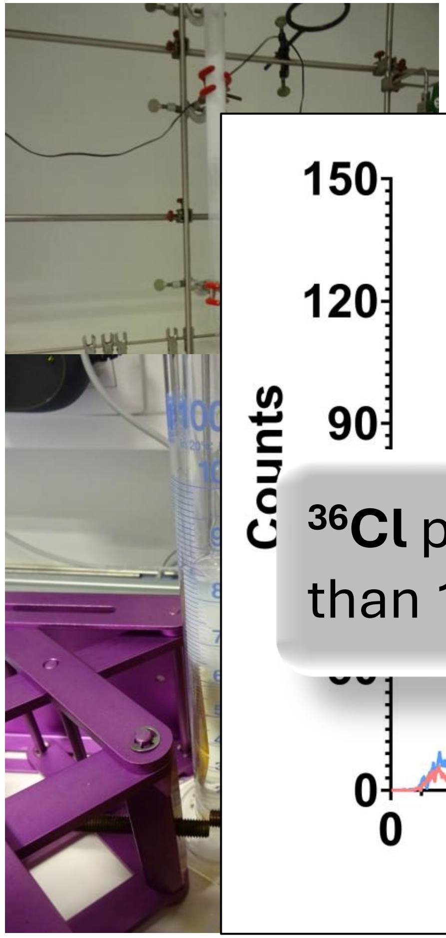


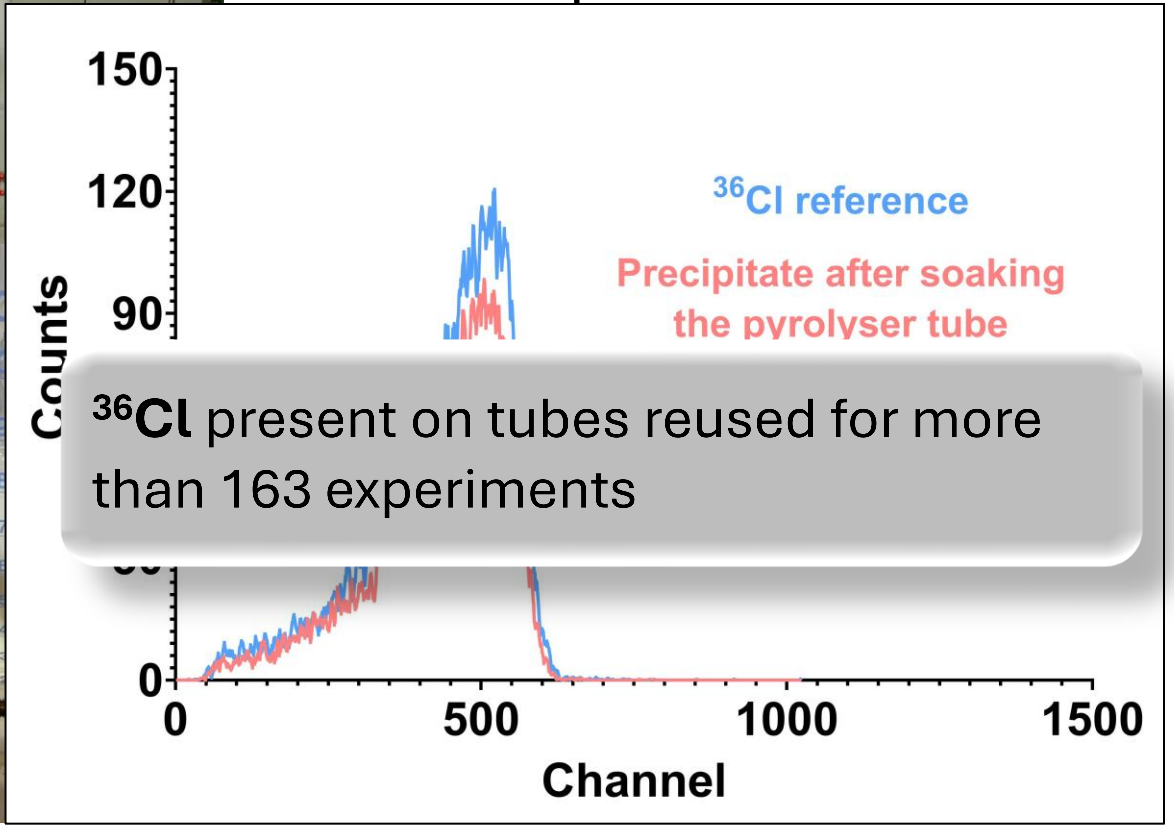






Used for 163 experiments with ^{36}Cl-containing samples





^{36}**Cl** present on tubes reused for more than 163 experiments

^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine

53 I 129 Iodine

34 Se 79 Selenium

40 Ca 41 Calcium

^{36}Cl memory effect on pyrolyser

adsorption or interaction of chlorine on the different surfaces comprising the pyrolysis system

Ageing effect

Moist air

Bubblers

Oxidant
(if needed)

Quartz wool/beads

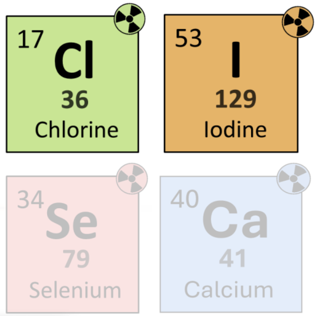
Quartz tubes

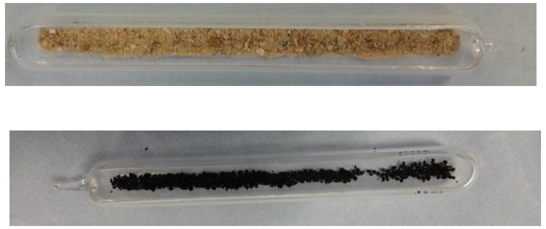
Sample boat

Can be removed

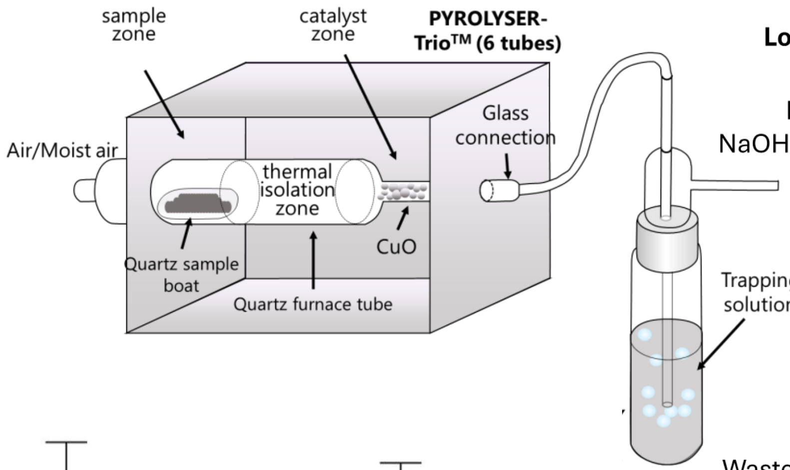
Major contributor

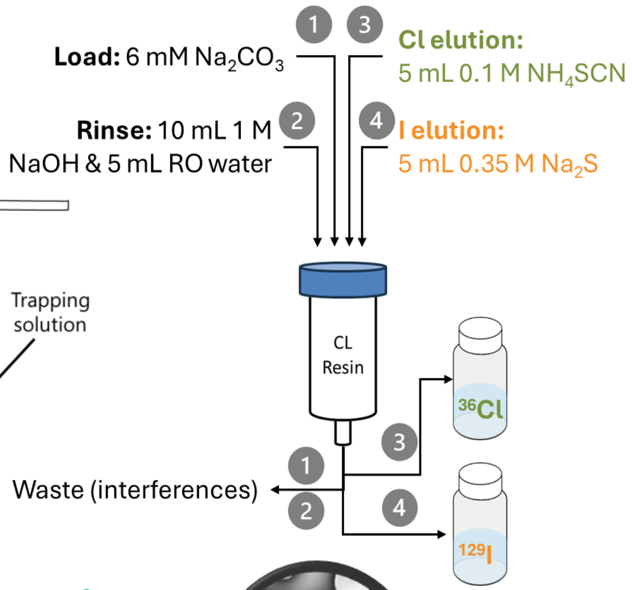
^{36}Cl and ^{129}I determination



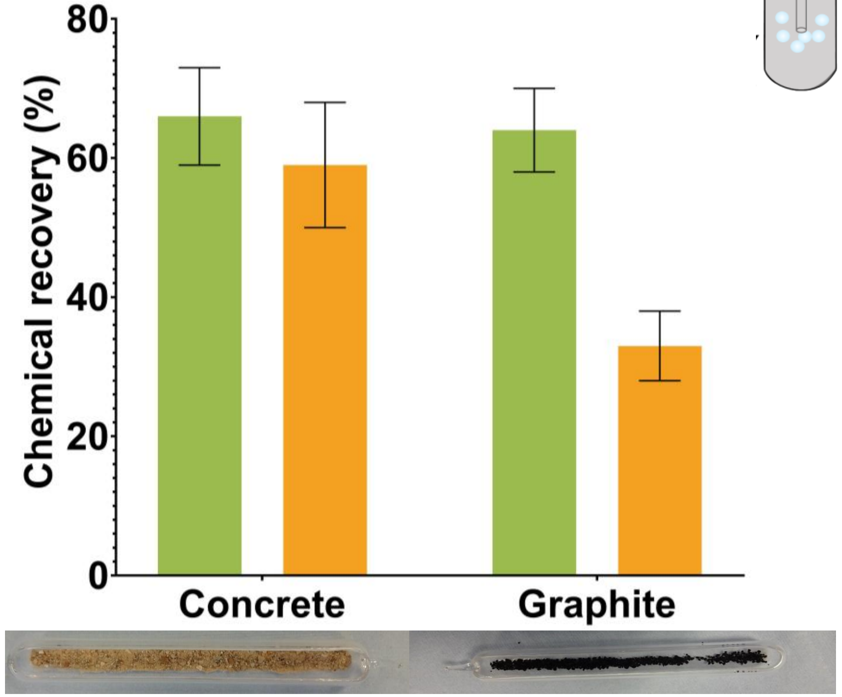


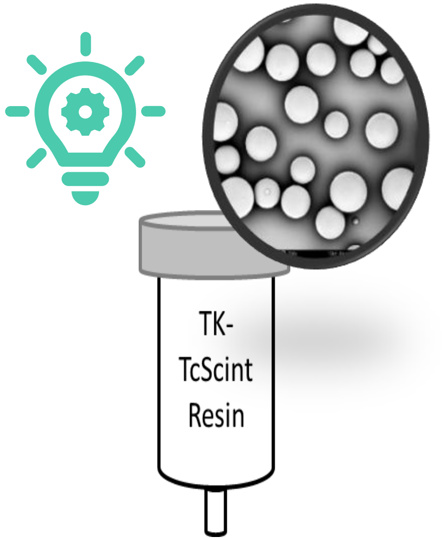
4 Bq ^{36}Cl/^{129}I
1 mg Cl and I





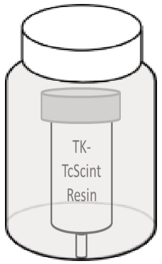
IC/ICP-MS



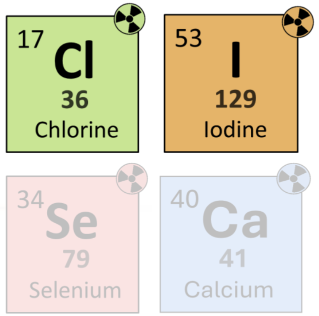


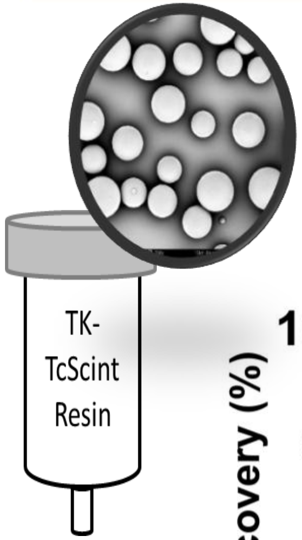
Turnaround time

Mixed wastes

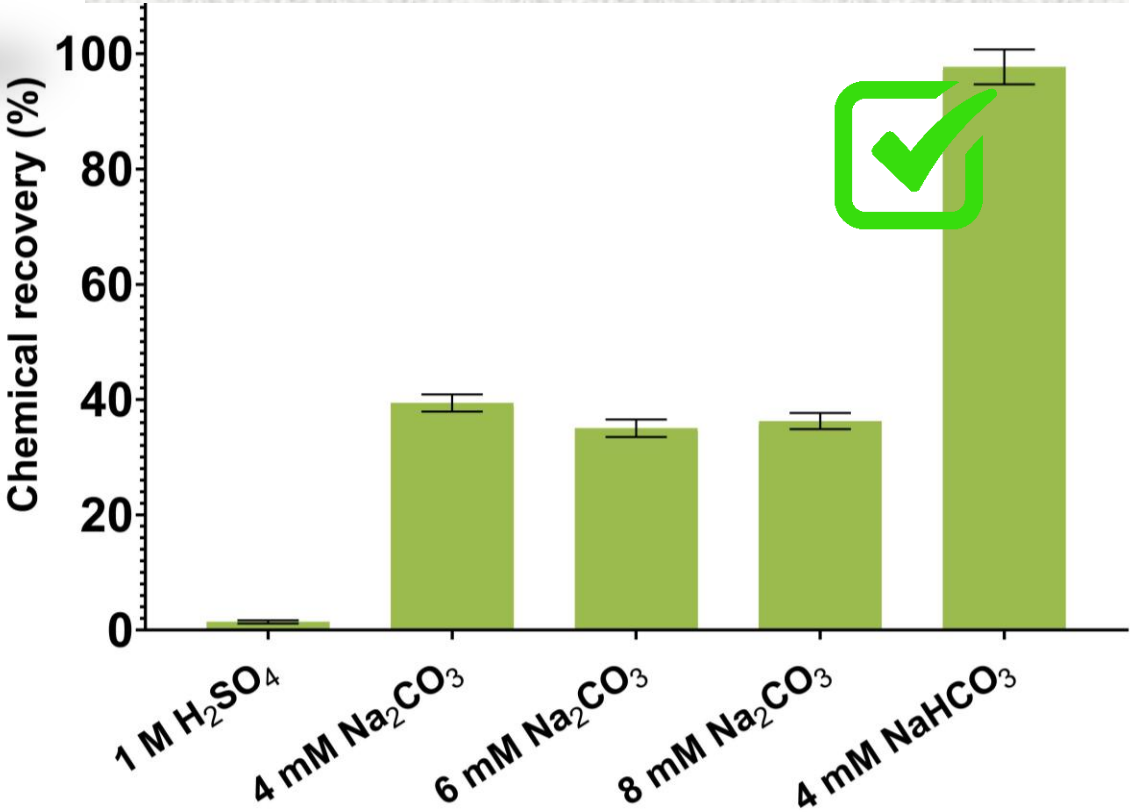


^{36}Cl and ^{129}I determination

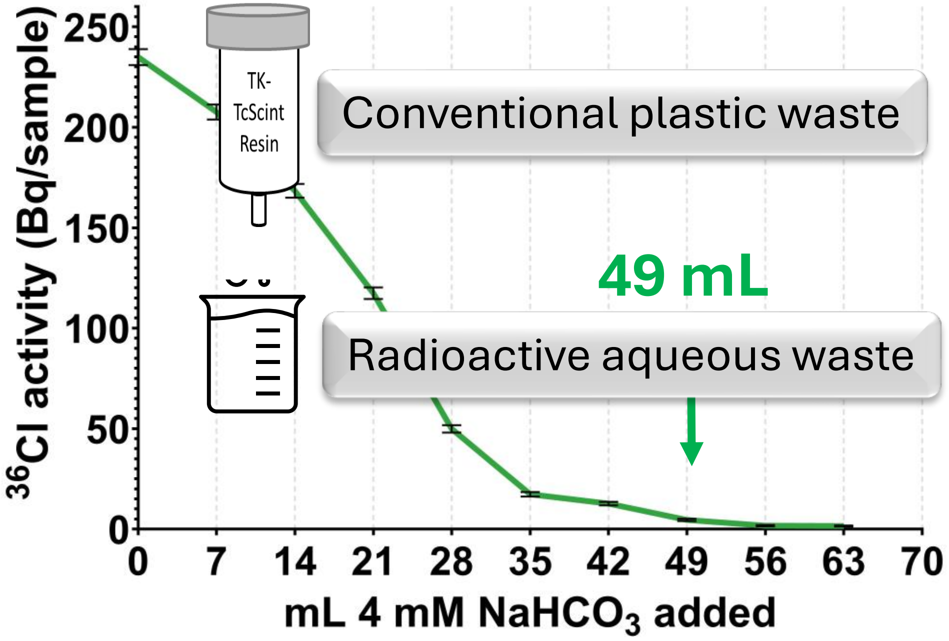




Suitable trapping media to retain ^{36}Cl on TK-TcScint Resin



Washing out ^{36}Cl activity from PS Resin using 4 mM $NaHCO_3$



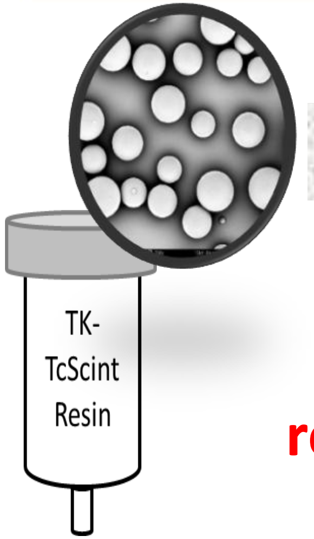
^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine

53 I 129 Iodine

34 Se 79 Selenium

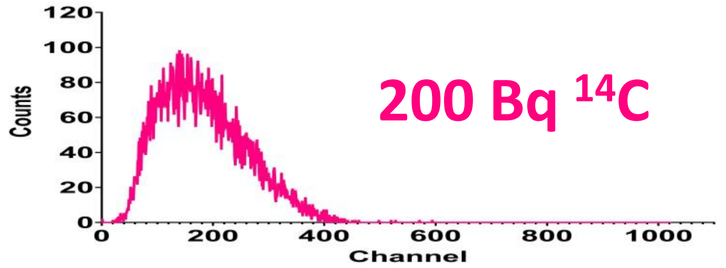
40 Ca 41 Calcium

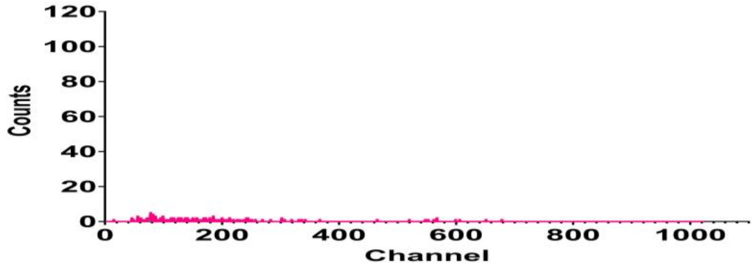


Interference removal

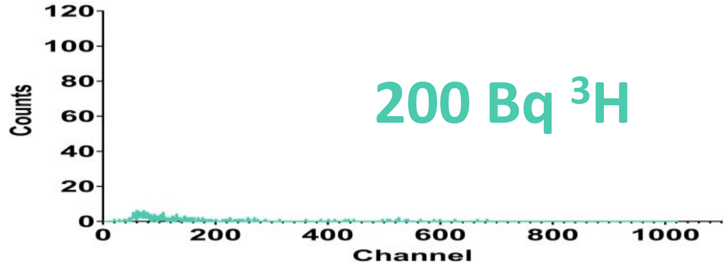
+ RO water

~2% retained

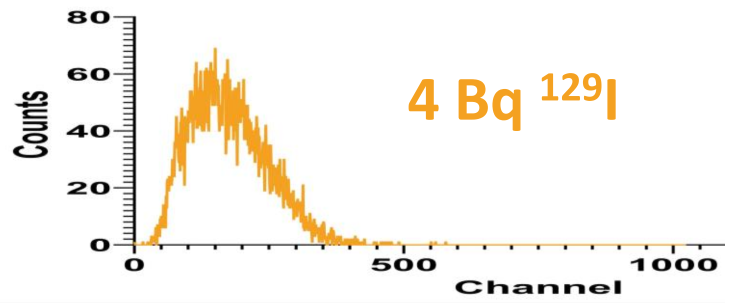


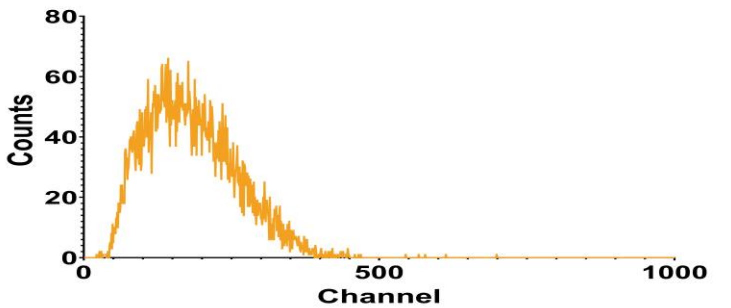


No retention



~100% retained





^{36}Cl and ^{129}I determination

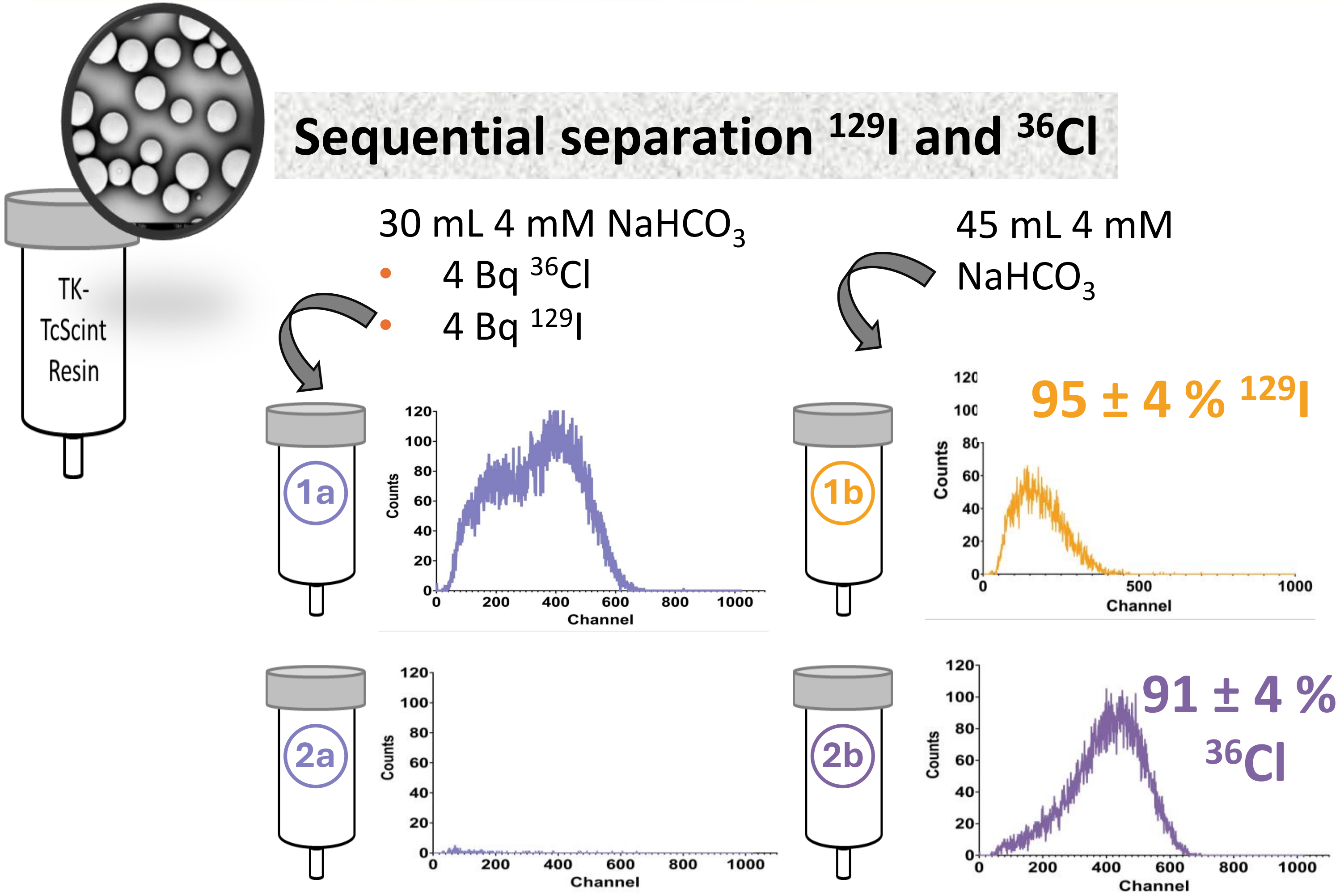
17 Cl 36 Chlorine

53 I 129 Iodine

34 Se 79 Selenium

40 Ca 41 Calcium

Sequential separation ^{129}I and ^{36}Cl



^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

Application of the procedures

LOD

Parameter	^{36}Cl	Clearance level by Belgian legislation	^{129}I	Clearance level by Belgian legislation
Count rate blank	3,8 CPM		3,5 CPM	
Counting time	100 min		100 min	
Mass of the sample	1 g		1 g	
Chemical recovery	64%		65%	
Counting efficiency	98%		92%	
LOD (α=β=0.05)	25 mBq g^{-1}	**<1000 Bq g^{-1}**	25 mBq g^{-1}	**>10 Bq g^{-1}**

^{36}Cl and ^{129}I determination

17 Cl 36 Chlorine
53 I 129 Iodine
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40 Ca 41 Calcium

Application of the procedures

Concrete from BR3

Scaling factor

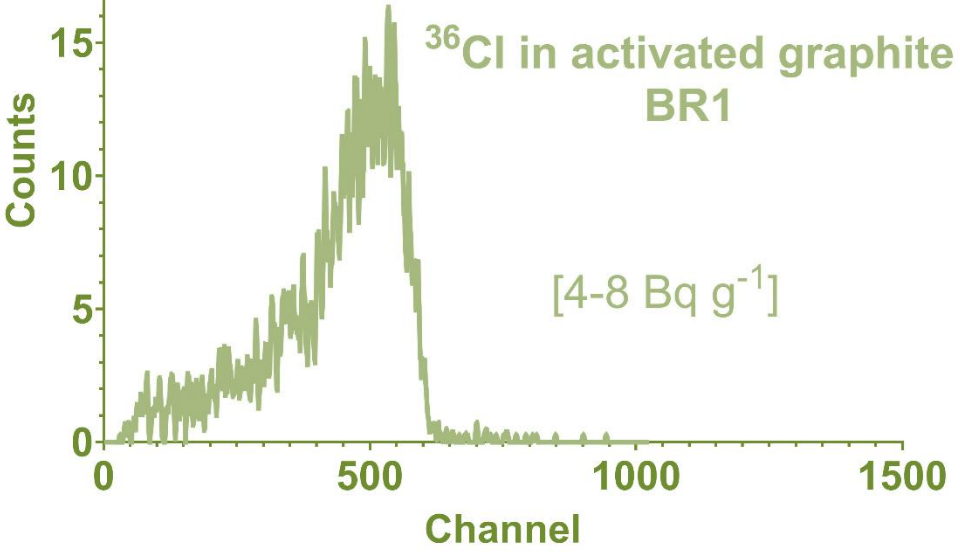
<3 mBq g^{-1}

^{36}Cl quantified <25 mBq g^{-1}

^{14}C, ^{3}H, ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu

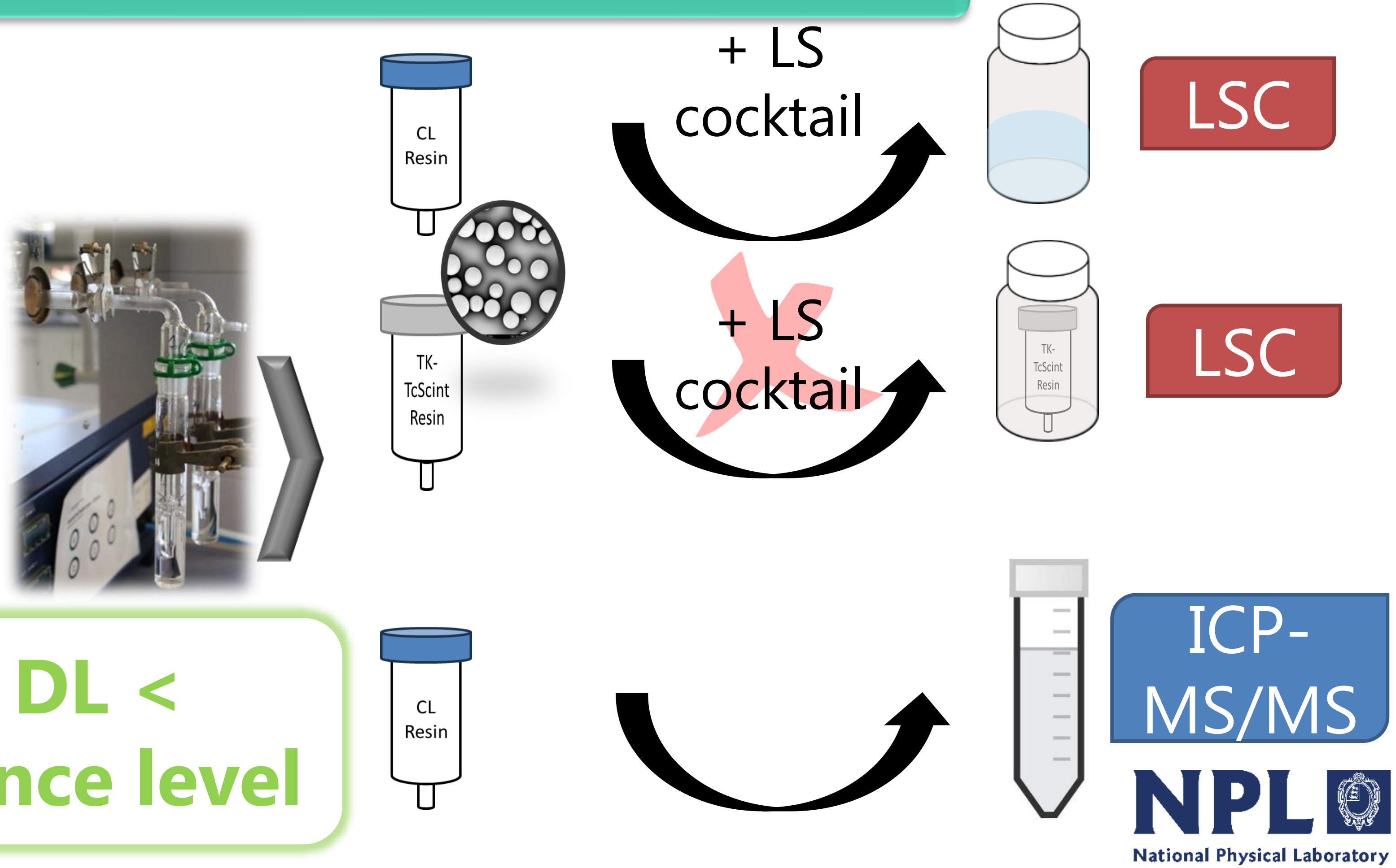
Graphite from BR1

^{14}C, ^{3}H, ^{60}Co, ^{133}Ba, ^{134}Cs, ^{152,154}Eu



^{36}Cl and ^{129}I determination

Comparison of different analytical techniques



No mixed waste
Fewer chemicals used

No mixed waste

All DL < clearance level

^{41}Ca in concrete samples

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

Physical barrier that protects against neutron and gamma-ray emissions from the nuclear reactor

Contains higher amount of shielding materials: boron, barite

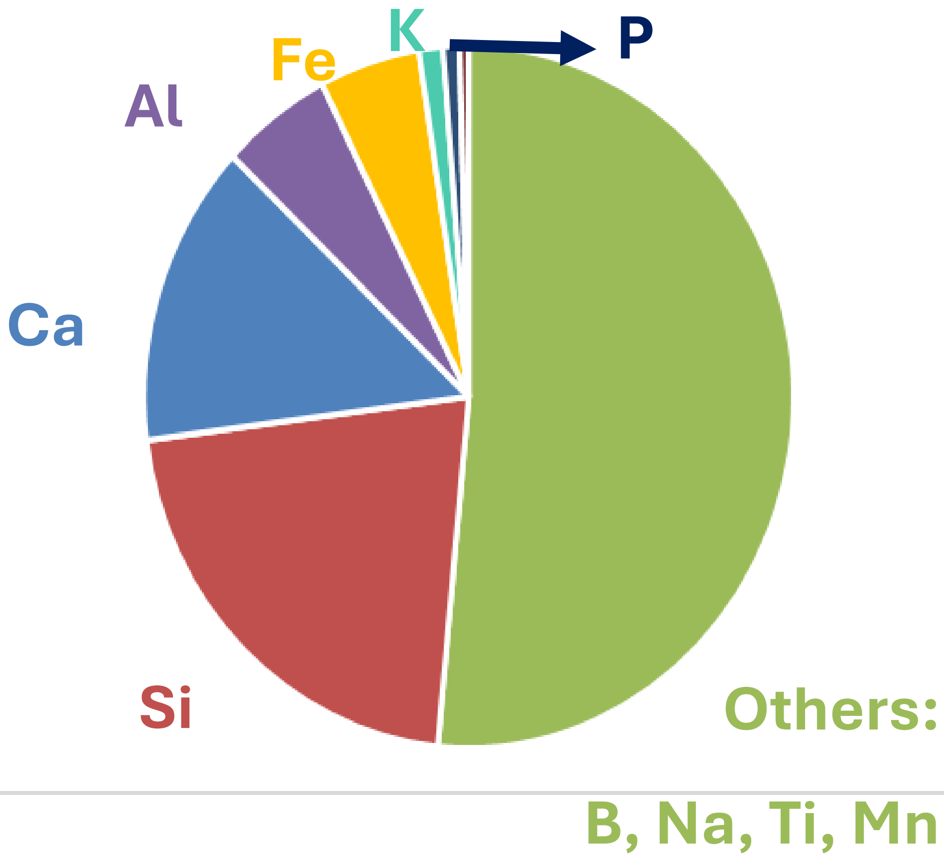
Calcium predominant component

(8 – 35%)

Silica

Barium

Chemical composition bioshield concrete



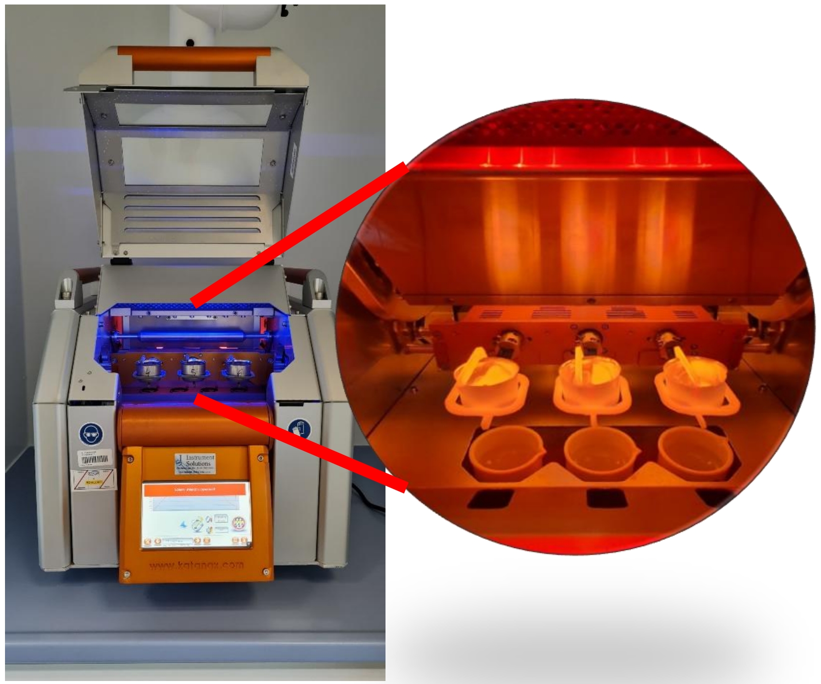
Evans, J.C et al. 1984. Long-lived activation products in reactor materials. Nureg/Cr-3474 1–185.

^{41}Ca in concrete samples

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

Complex procedures for **matrix dissolution**

Lithium borate fusion



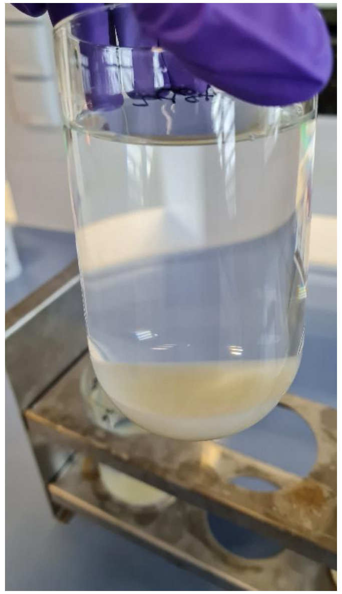
^{41}Ca in concrete samples

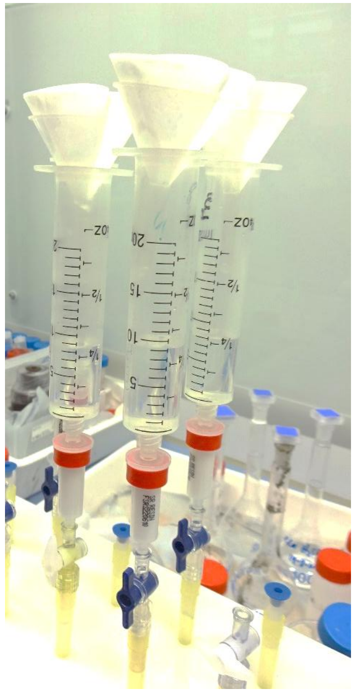
17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

Complex procedures for **matrix dissolution**

Long **radiochemical separation** procedures

Precipitation (matrix elements, metals) + Solid phase extraction





^{41}Ca in concrete samples

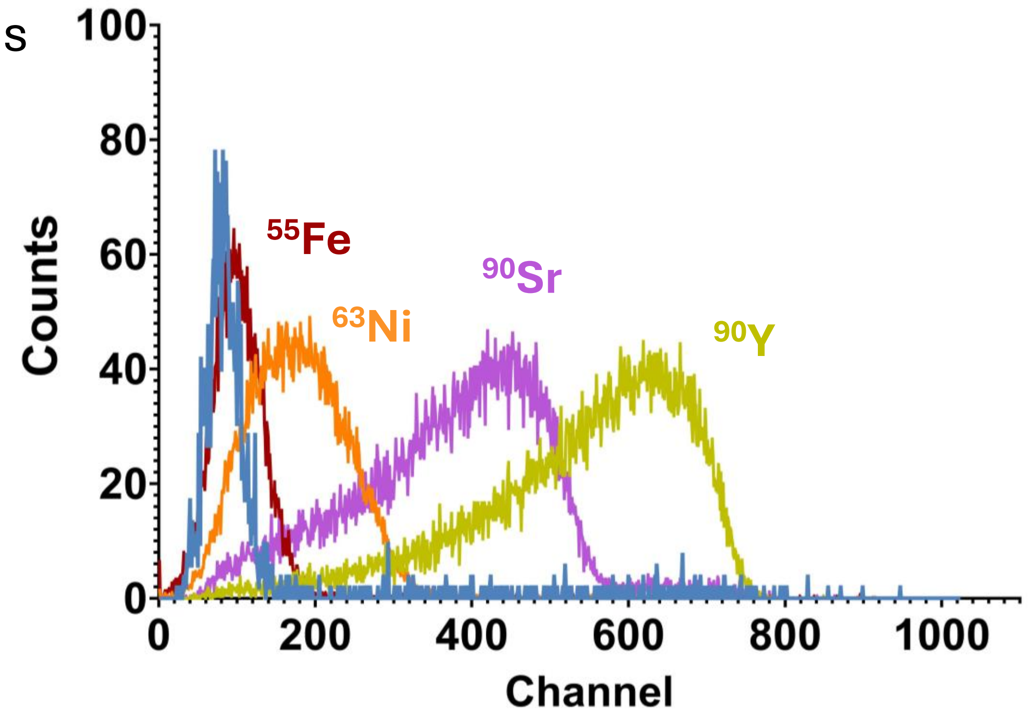
17 Cl 36 Chlorine

53 I 129 Iodine

34 Se 79 Selenium

40 Ca 41 Calcium

- Complex procedures for **matrix dissolution**
- Long **radiochemical separation** procedures
- Measurement of **^{41}Ca** by LSC



^{41}Ca in concrete samples

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

Fusion

Step	Procedure	Rocking angle	Speed	Time
1	Heating 1	-	-	1 min 30 s
2	Heating 2	25°	100%	8 min
3	Heating 3	30°	5%	1 min 30 s
3-4	Pouring	130°	100%	-
4	Cooling 1	120°	50%	1 min
5	Cooling 2	90°	30%	5 min

20 min

^{41}Ca in concrete samples

Fusion

Unsieved concrete	Sieved concrete
✓ 0,1 g	✓ 0,5 g
✓ 0,2 g	✓ 0,7 g
✓ 0,3 g	✓ 0,9 g
✗ 0,4 g	✗ 1 g
✗ 0,5 g	

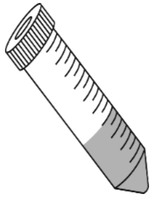
^{41}Ca in concrete samples

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

Chemical separation

T – 1000°C

~20 min

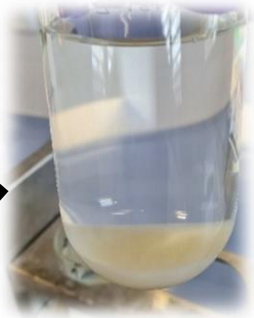


100 µL sample
for **ICP-AES**

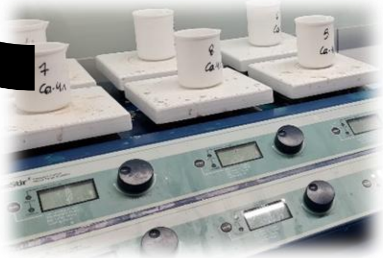
polyethylene glycol (PEG)
PRECIPITATION

↑ pH 8 (NH_4OH)

~2h



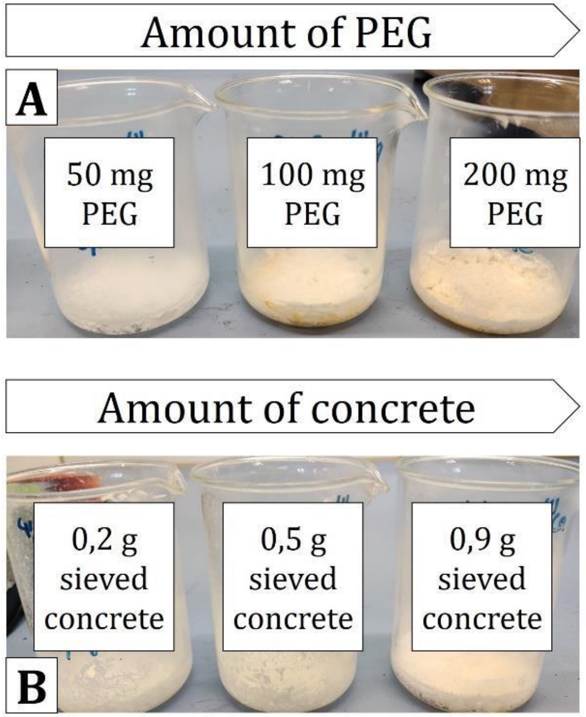
$Fe(OH)_3$



overnight

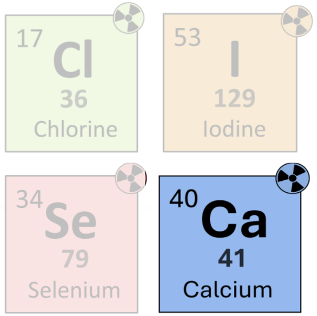
Evaporate to dryness

Jelly-like phase



Based on Russell et al., 2021

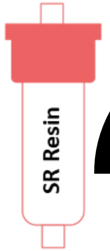
^{41}Ca in concrete samples



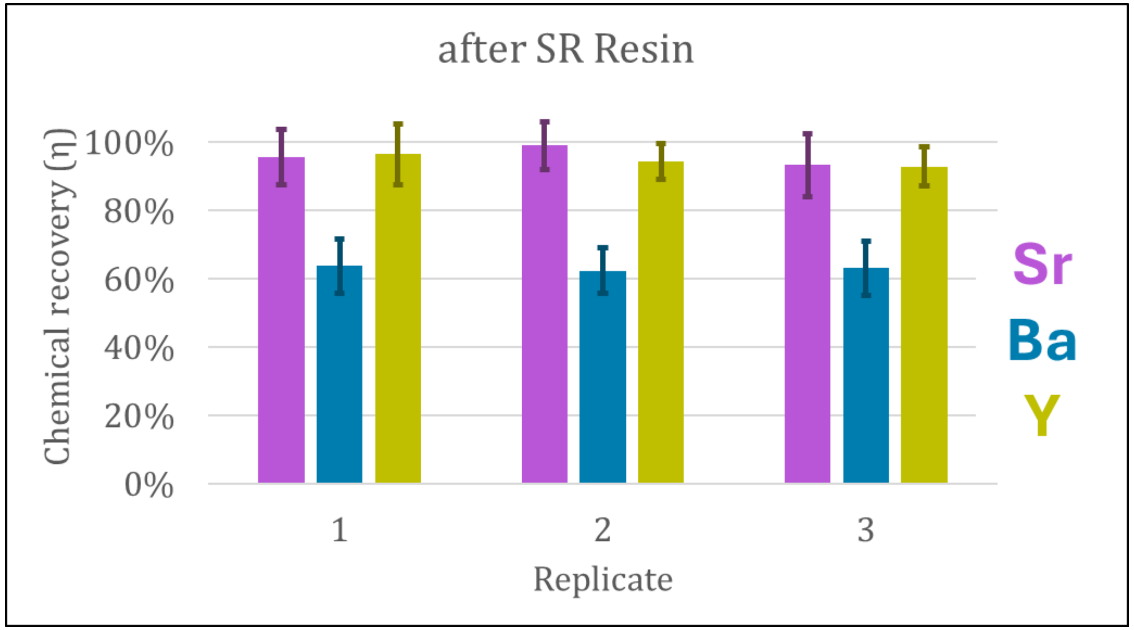
Chemical separation

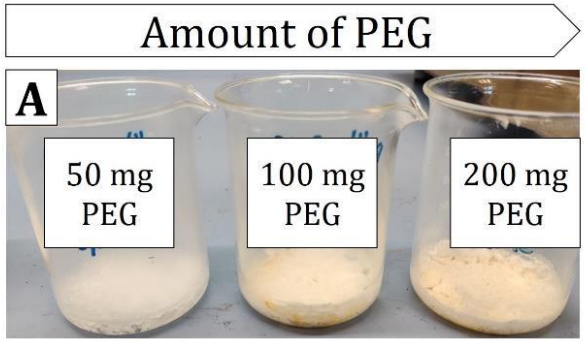
For Sr separation from Ba and Y

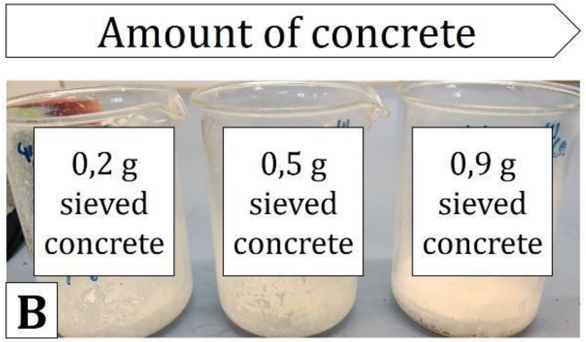
No Sr retention



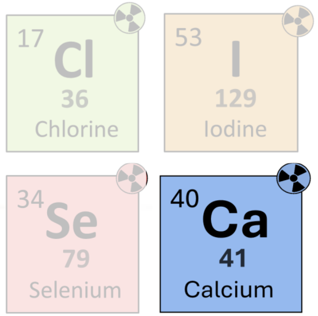
Evaporate to dryness







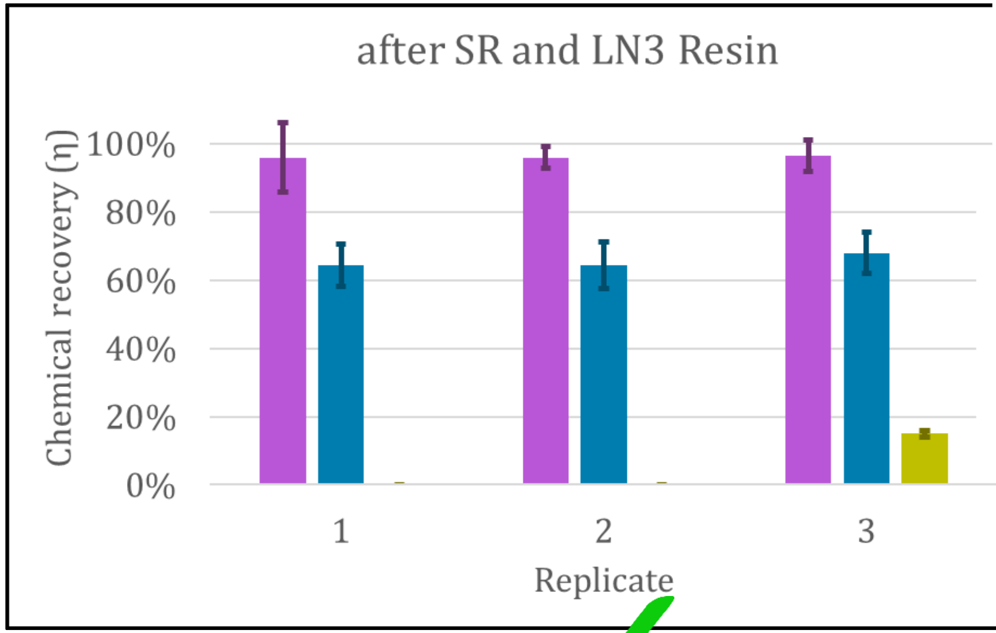
^{41}Ca in concrete samples



Chemical separation

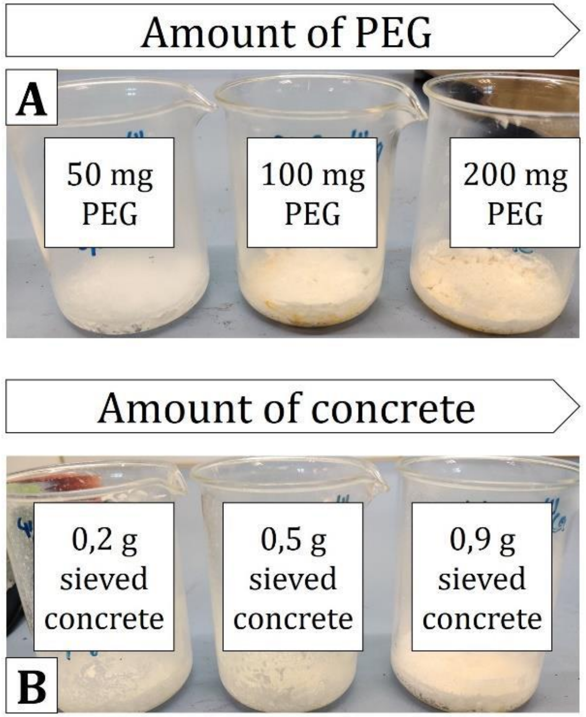
For Sr separation from Ba and Y

No Sr retention



Y retention

Evaporate to dryness



^{41}Ca in concrete samples

17 Cl 36 Chlorine

53 I 129 Iodine

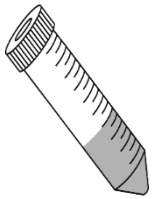
34 Se 79 Selenium

40 Ca 41 Calcium

Chemical separation

T – 1000°C

~20 min

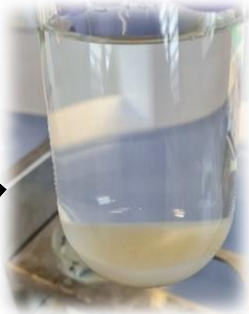


100 µL sample
for **ICP-AES**

polyethylene glycol (PEG)
PRECIPITATION

↑ pH 8 (NH_4OH)

~2h

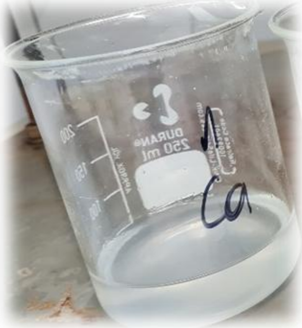


$Fe(OH)_3$

overnight

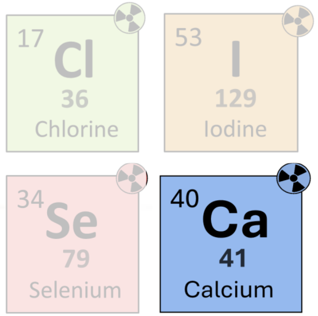
Oxalic acid
↑ pH 5 (NH_4OH)
PRECIPITATION

~1h

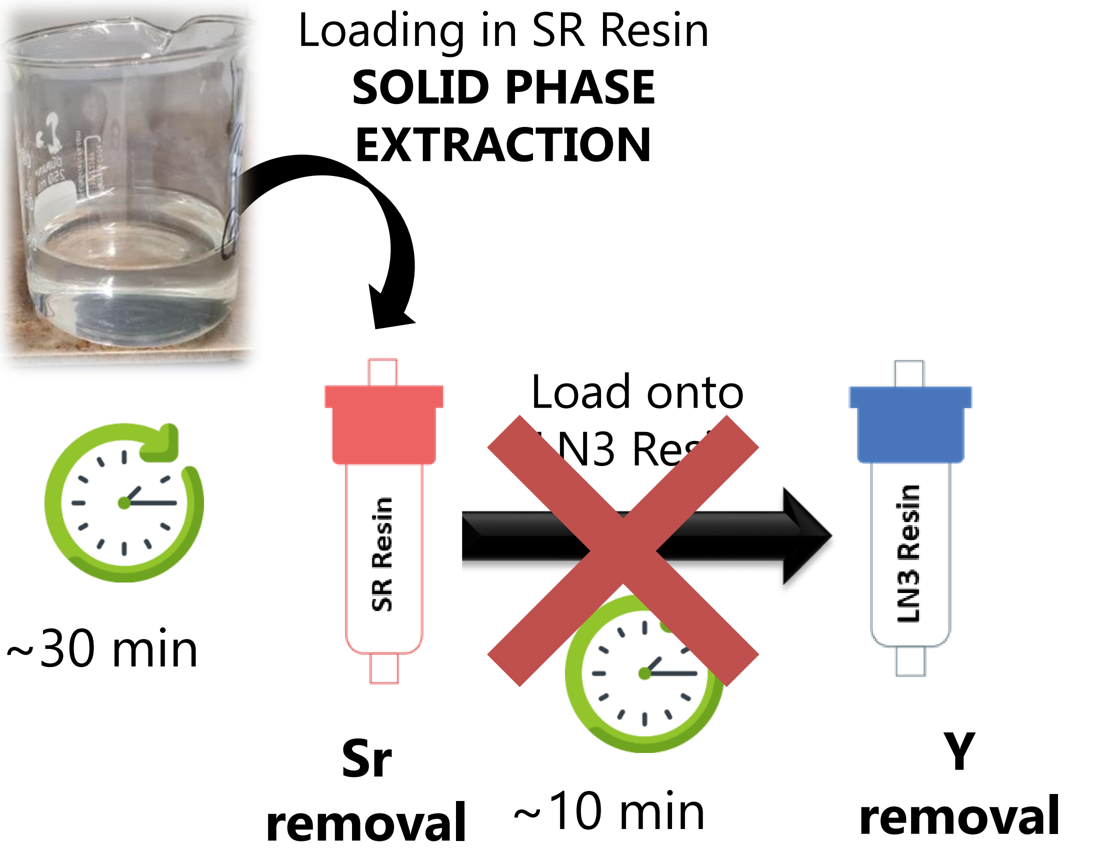


Dissolve precipitate in 1 M HNO_3

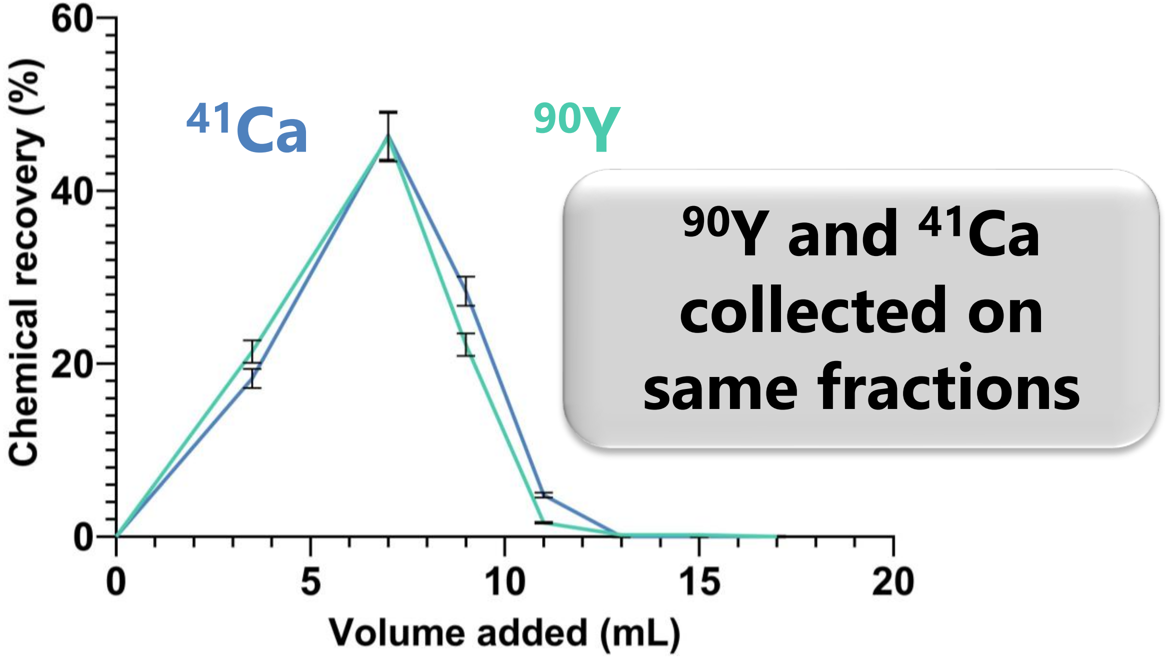
^{41}Ca in concrete samples



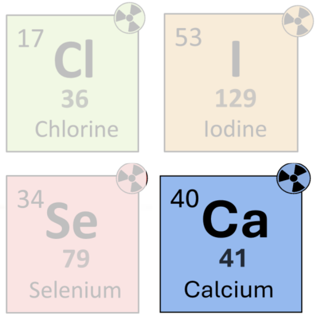
Chemical separation



Possible ^{41}Ca quantification when ^{90}Y is present?

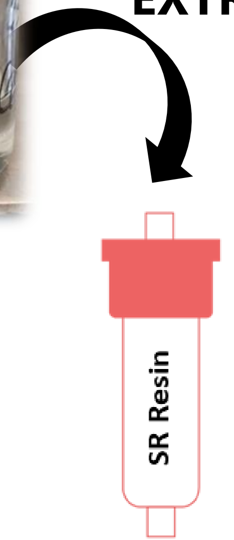


^{41}Ca in concrete samples



Chemical separation

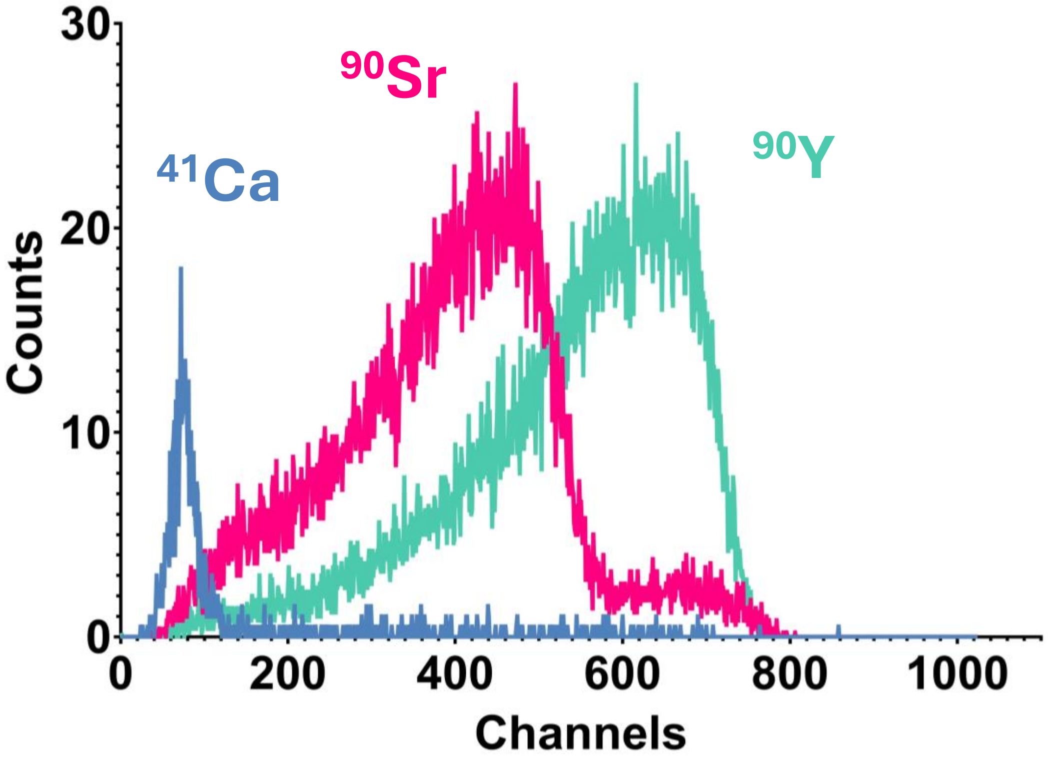
Loading in SR Resin
SOLID PHASE EXTRACTION



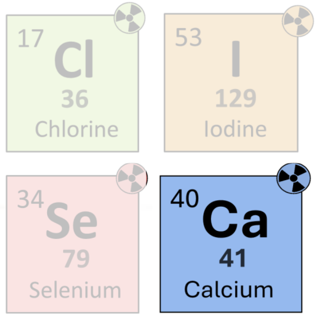
~30 min

Sr removal

Single LSC spectra



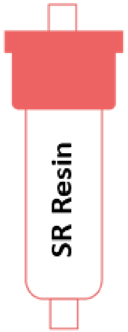
^{41}Ca in concrete samples



Chemical separation

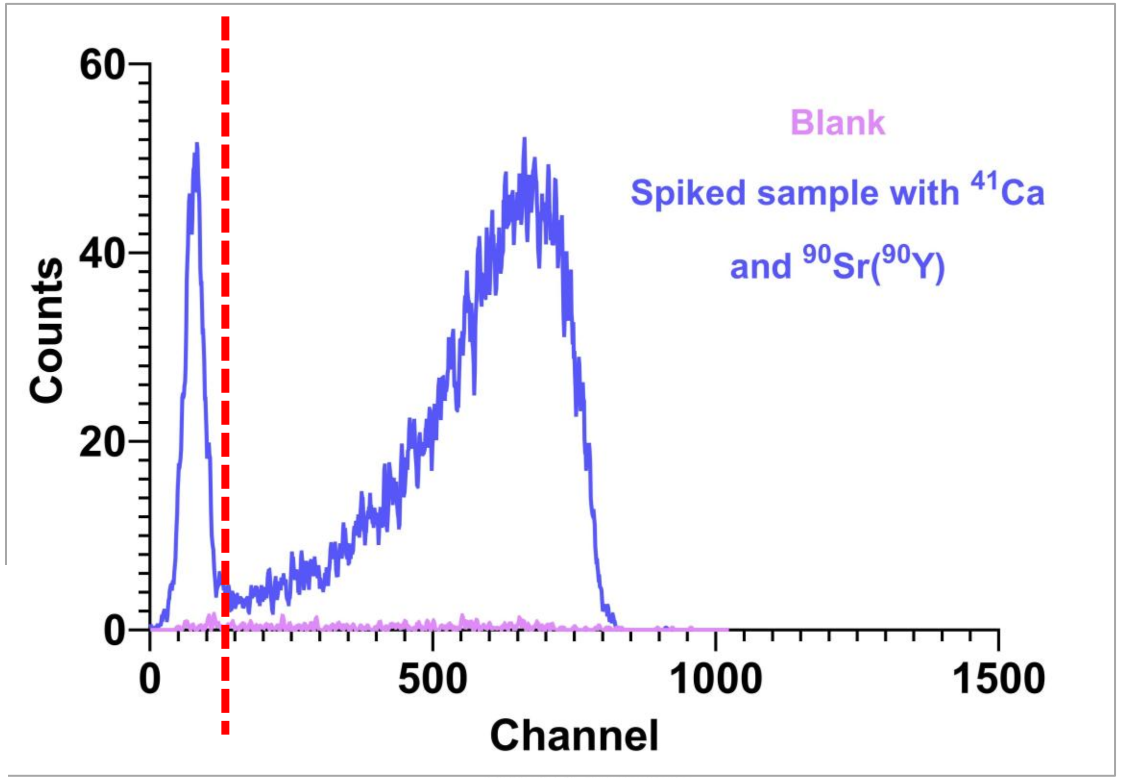
Loading in SR Resin
SOLID PHASE EXTRACTION

~30 min



Sr removal

Possible ^{41}Ca quantification by cutting the spectra



^{41}Ca in concrete samples

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

Chemical separation

Loading in SR Resin
SOLID PHASE EXTRACTION

~30 min

Evaporate to ½ volume

~1 h 30 min

½ sample for **LSC**

60 min

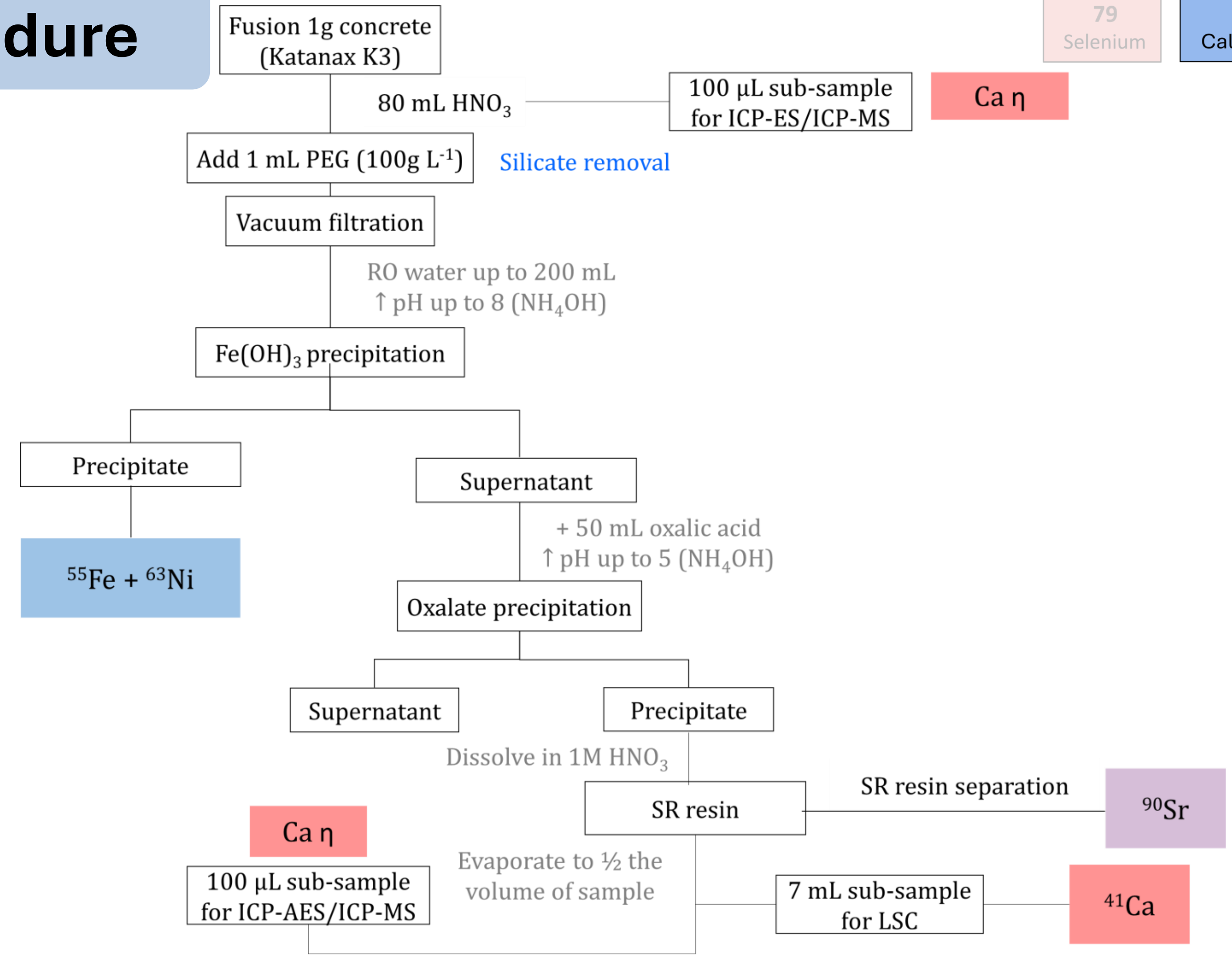
~80% ^{41}Ca chemical recovery

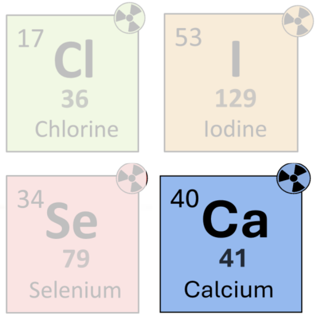
100 µL sample for **ICP-AES**

~64% calcium chemical recovery

^{41}Ca in concrete samples

Verification of the procedure



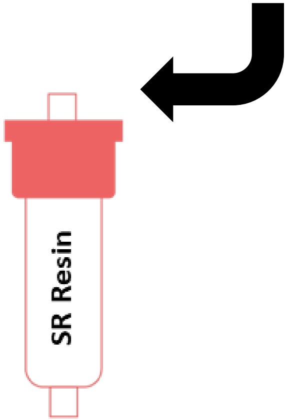


^{41}Ca in concrete samples

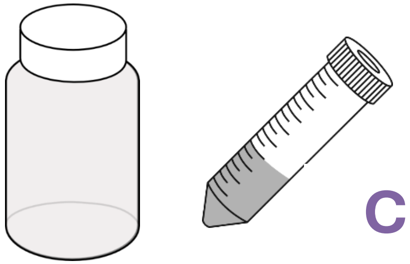
Verification of the procedure

polyethylene glycol (PEG) **PRECIPITATION** — ↑ pH 8 (NH_4OH) **$Fe(OH)_3$ PRECIPITATION** — Oxalic acid ↑ pH 5 (NH_4OH) **PRECIPITATION**

1 M HNO_3



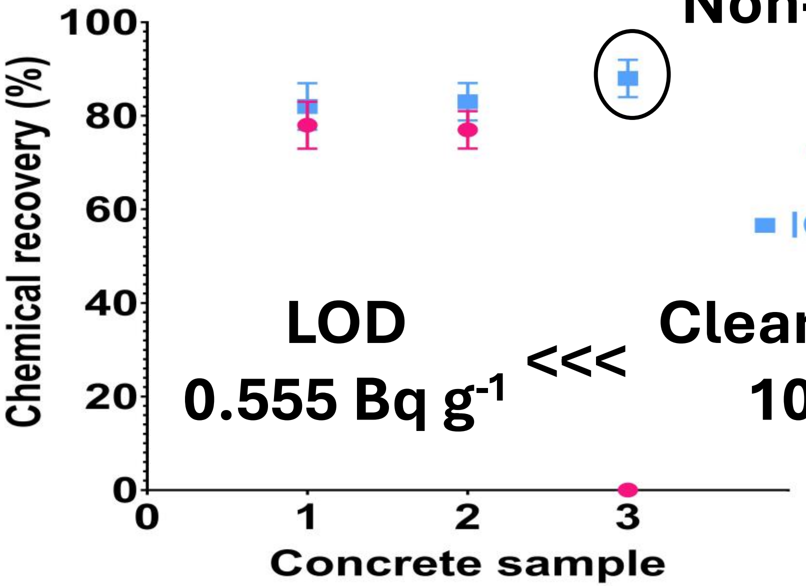
Evaporate to ½ volume



^{41}Ca activity

Ca η

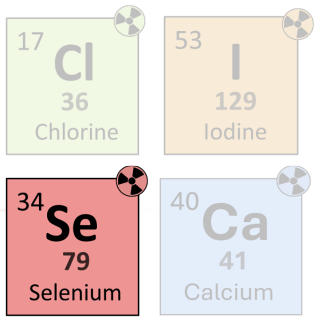
Ca η



Non-spiked

LOD 0.555 Bq g^{-1} <<< Clearance level 100 Bq g^{-1}

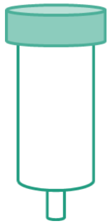
On the determination of ^{79}Se



? **Low specific activity and trace concentration**

Preconcentration steps

Selective separations



? **Lack of reference materials / standards solutions**

Use of ^{75}Se as a tracer for validation

CIEMAT/NIST for calibration

? **Complex chemistry (speciation)**

multiple oxidation states: −II, 0, +IV, +VI

REDOX during sample preparation

On the determination of ^{79}Se

17 Cl 36 Chlorine

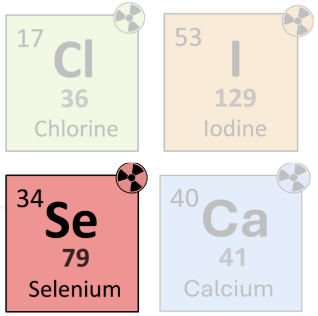
53 I 129 Iodine

34 Se 79 Selenium

40 Ca 41 Calcium

Development of a **new resin-based** material for the concentration and chemical separation of Se

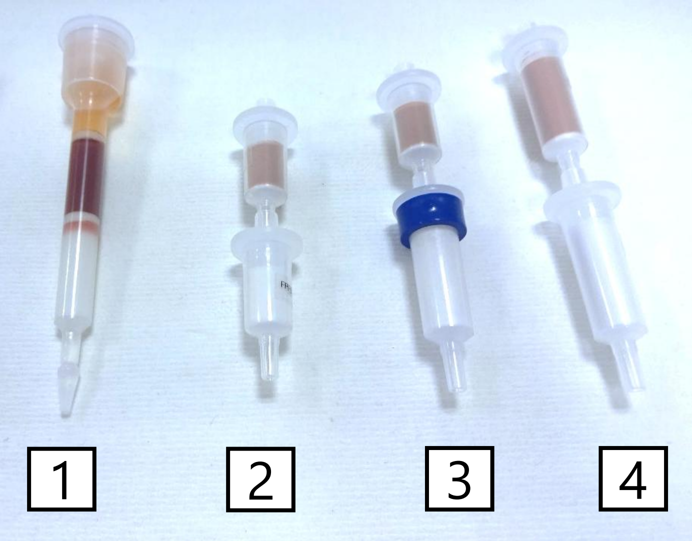
On the determination of ^{79}Se



SE Resin prototype

New SE Resin

TRISKEM



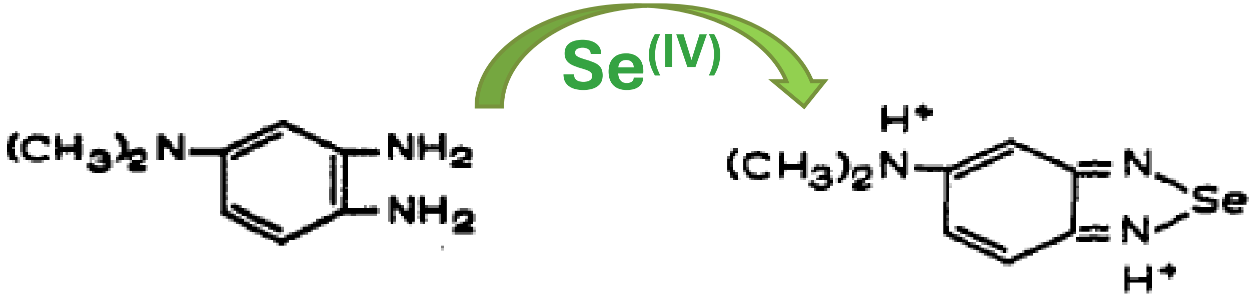
SE Resin

Se retention

Prefilter Resin

↓

extractant bleeding



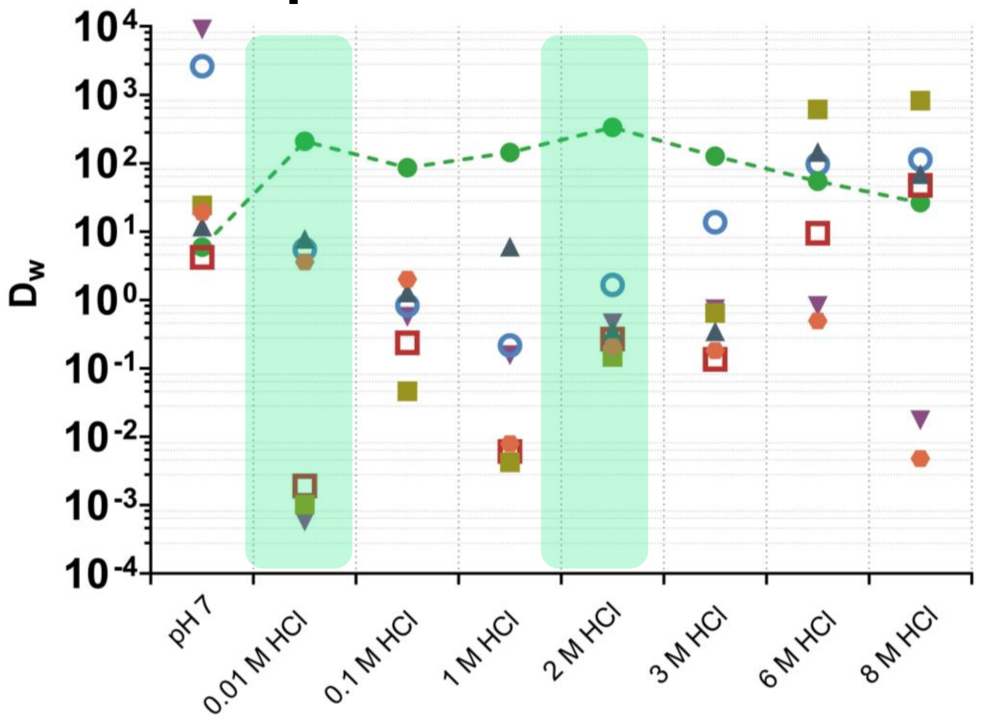
Acid media (H^+)

Influence Se oxidation state

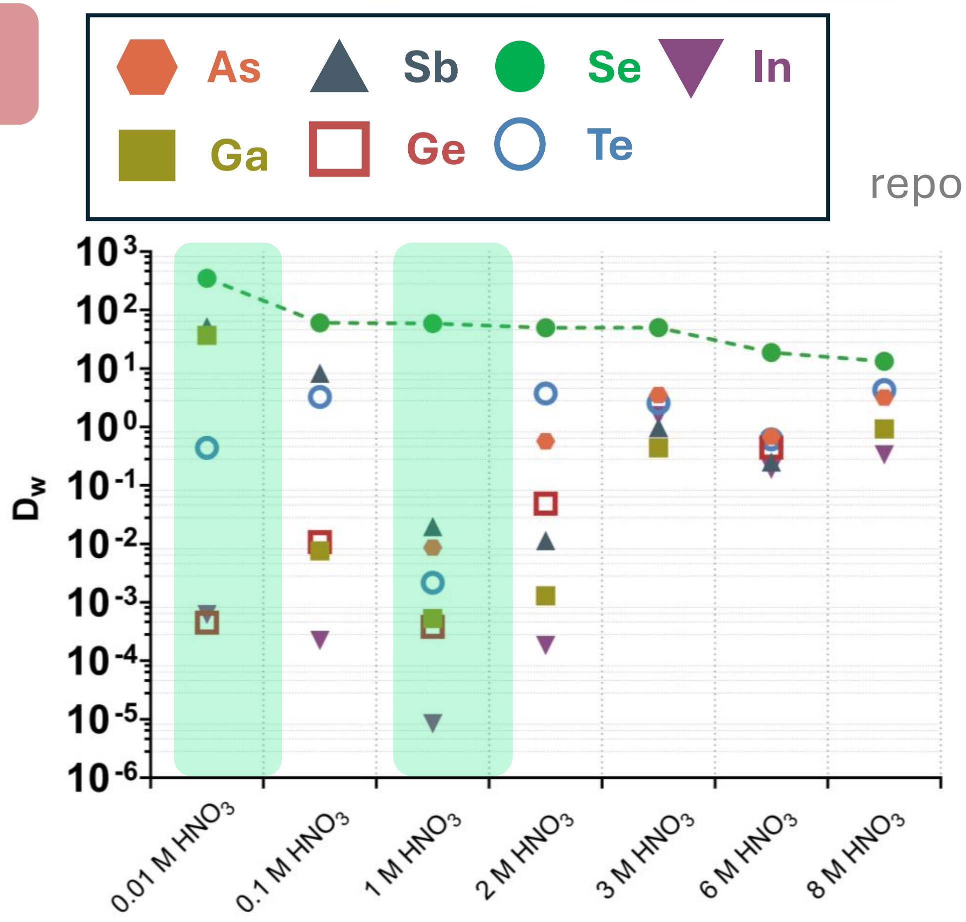
On the determination of ^{79}Se

SE Resin D_ws

Batch experiments



HCl



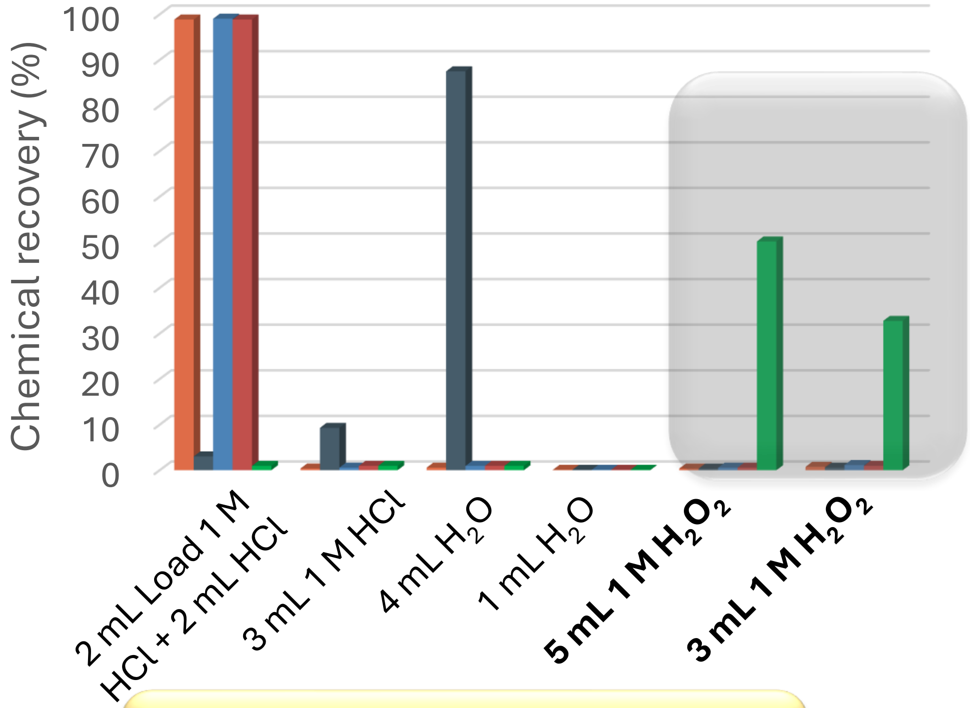
HNO_3

reported by Dirks et al. 2016

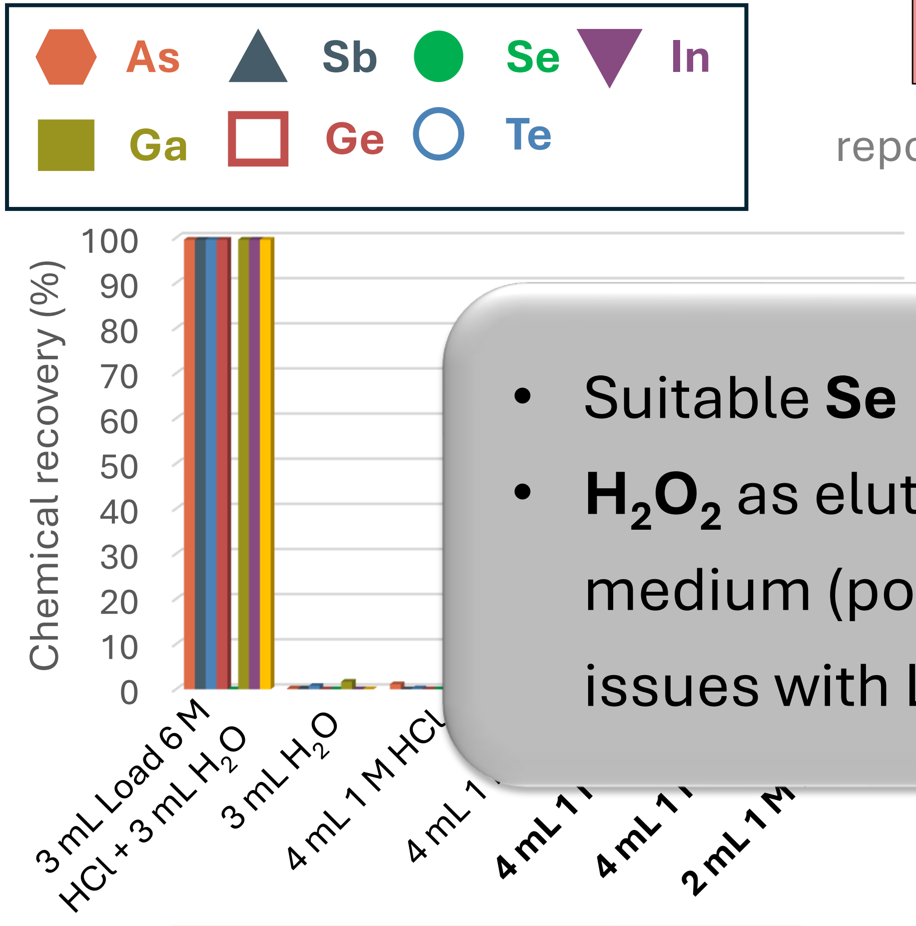
17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

On the determination of ^{79}Se

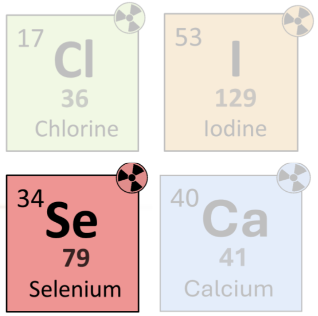
SE Resin elution profile



350 mg Se selective Resin **Load 1 M HCl**



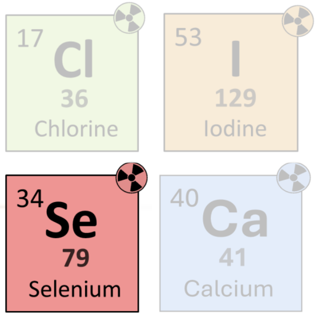
300 mg Se selective Resin **Load 6 M HCl**



reported by Dirks et al. 2016

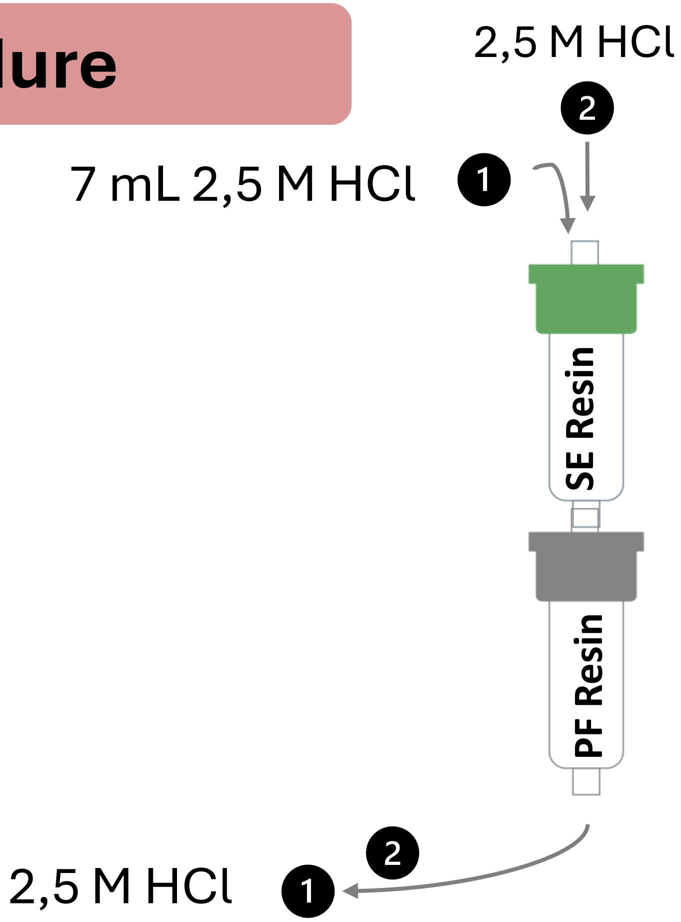
- Suitable **Se** retention
- **H_2O_2** as elution medium (potential issues with LSC)

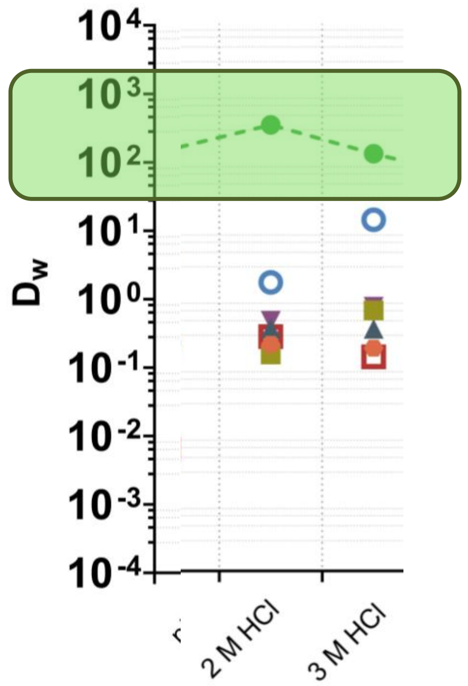
On the determination of ^{79}Se

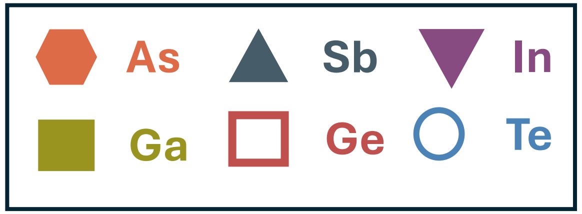


SE Resin procedure

"standard" procedure



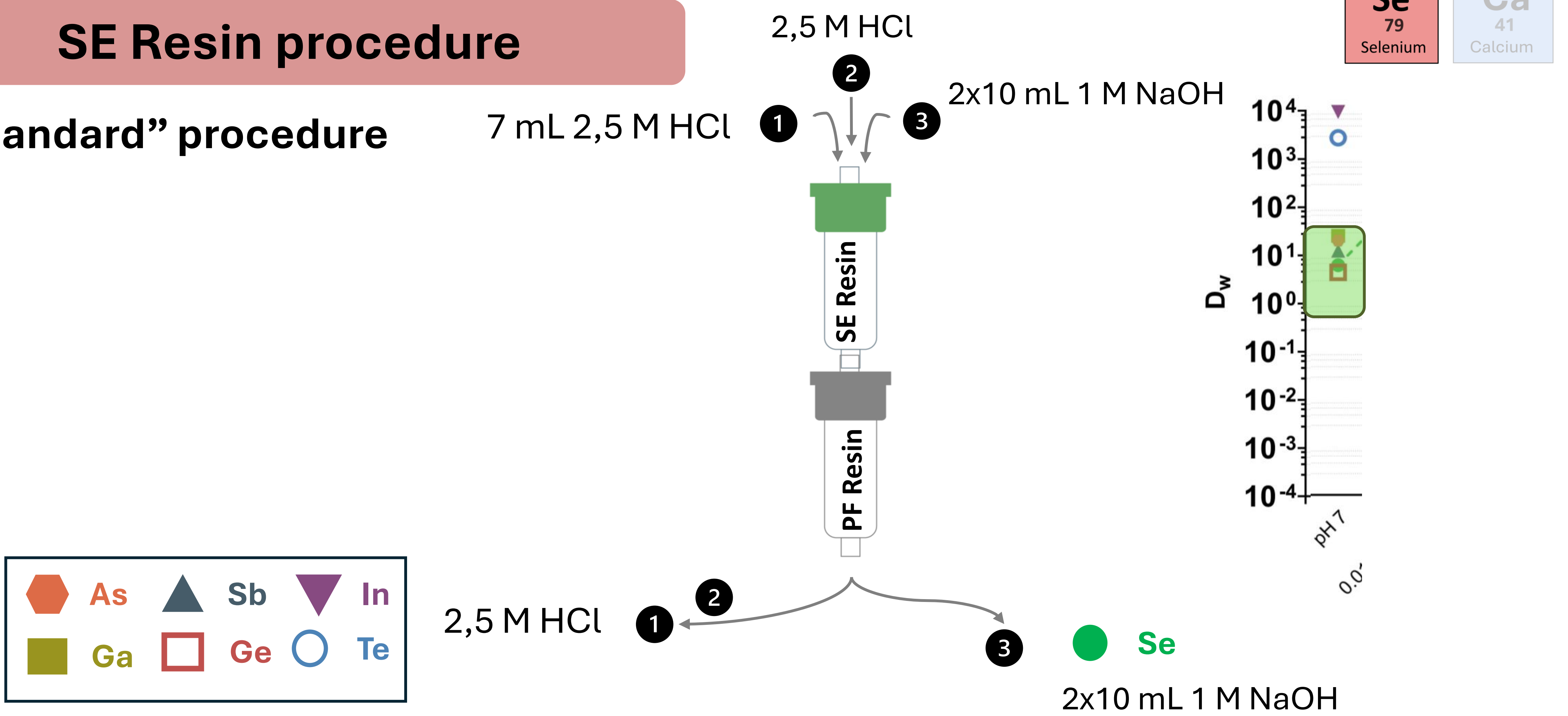




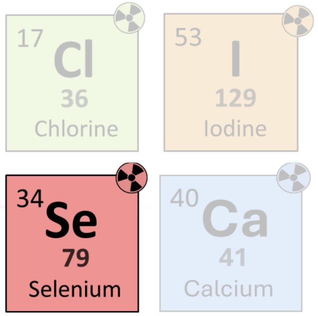
On the determination of ^{79}Se

SE Resin procedure

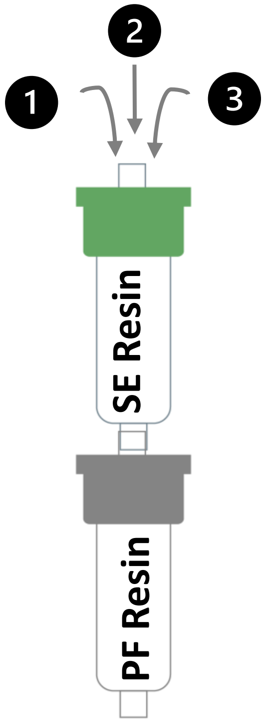
"standard" procedure



On the determination of ^{79}Se

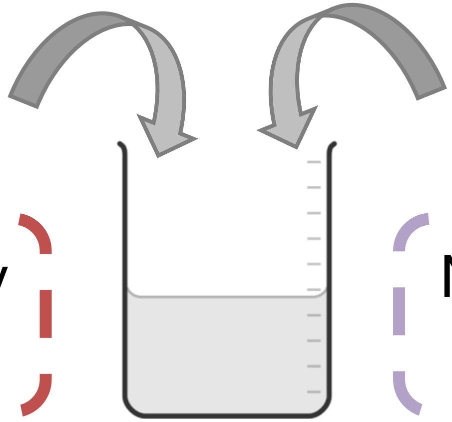


SE Resin procedure



1. 7 mL 2.5 M HCl
2. 10 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

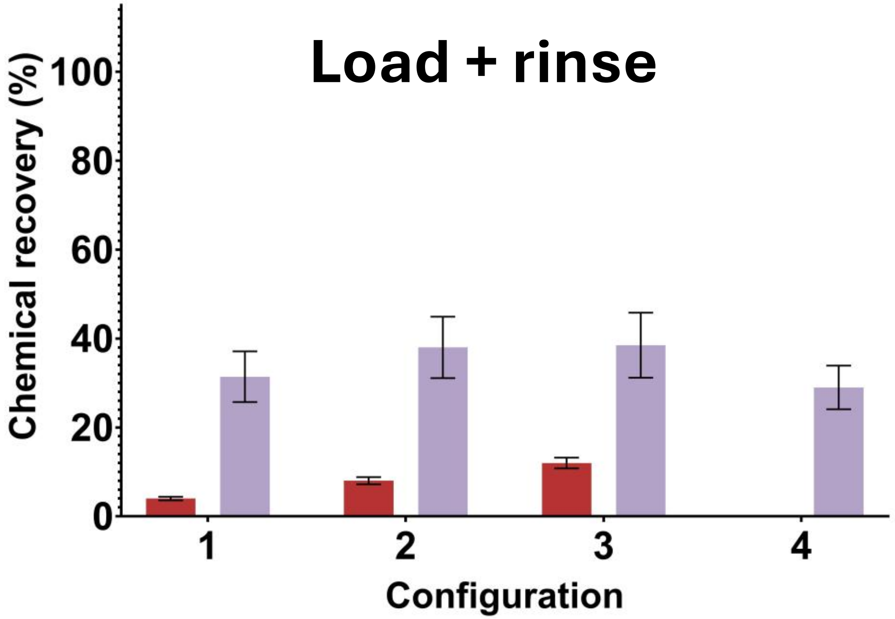
1 mg stable Se



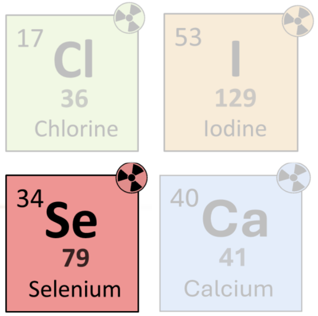
6 Bq ^{75}Se

Measured by SF-ICP-MS

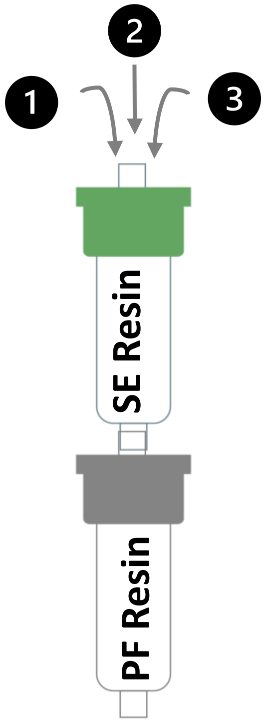
Measured by gamma-ray spectrometry



On the determination of ^{79}Se



SE Resin procedure



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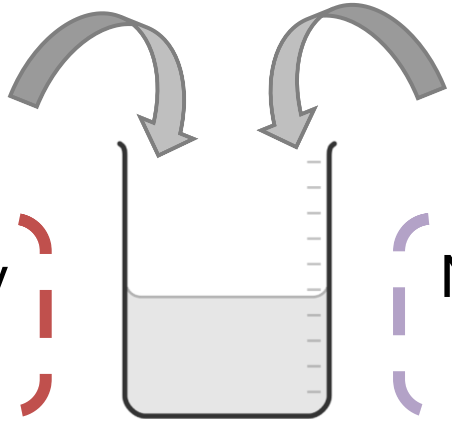
2. 10 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

1 mg stable Se



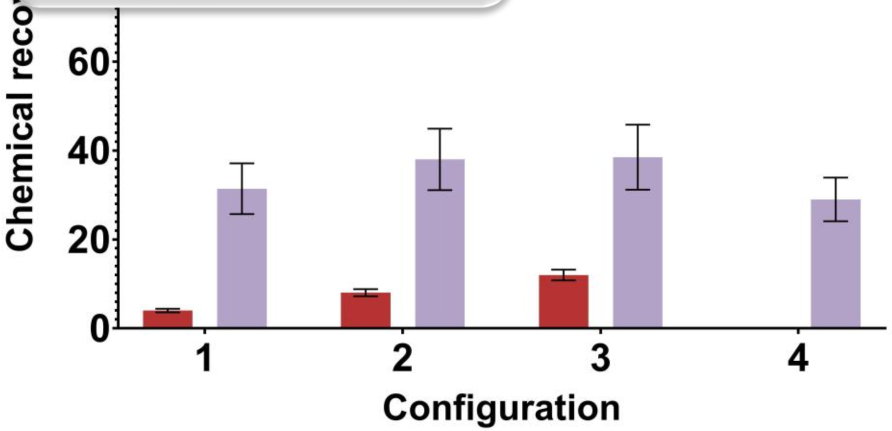
6 Bq ^{75}Se

Measured by SF-ICP-MS

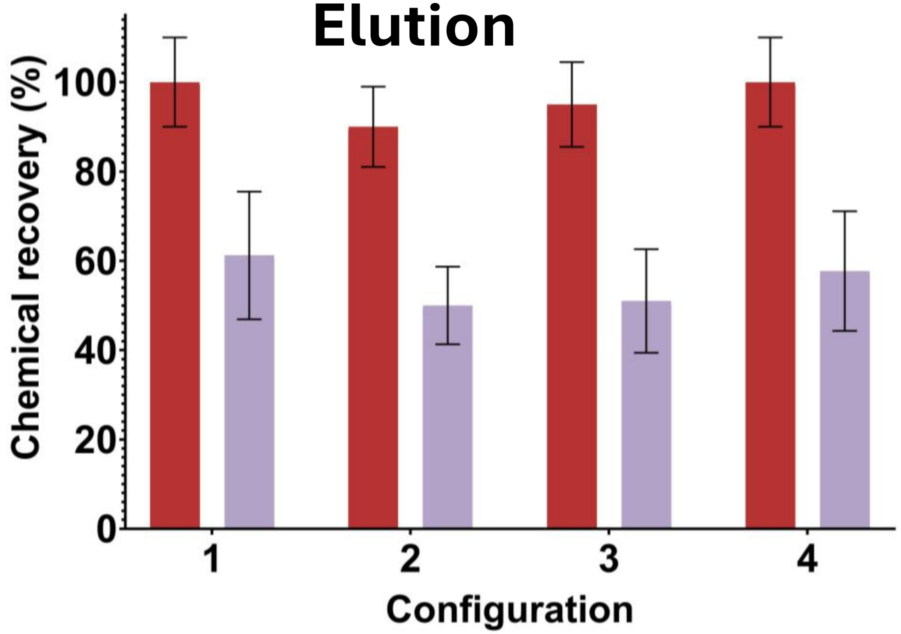
Measured by gamma-ray spectrometry

Difference in Se retentions

Load + rinse



Elution



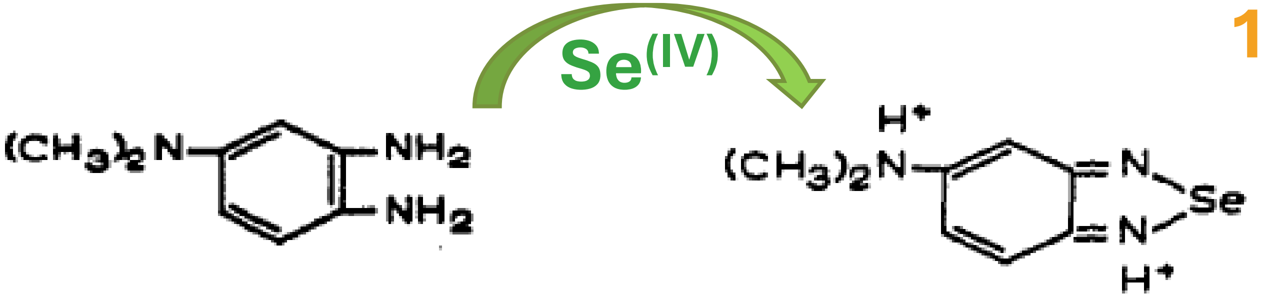
On the determination of ^{79}Se

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

SE Resin procedure

1. Incomplete mixing / spike conditioning
   Spike separately ^{75}Se and stable Se

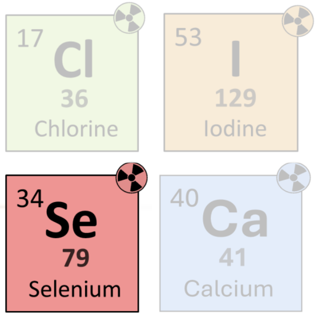
2. Different measurement techniques / calibration issues
   Total of 100 % with all fractions collected
3. Different chemical forms / speciation



100% | 1 mg stable Se | $Se^{(IV)}$

50% | 6 Bq ^{75}Se | $Se^{(IV/VI)}$

On the determination of ^{79}Se



SE Resin procedure

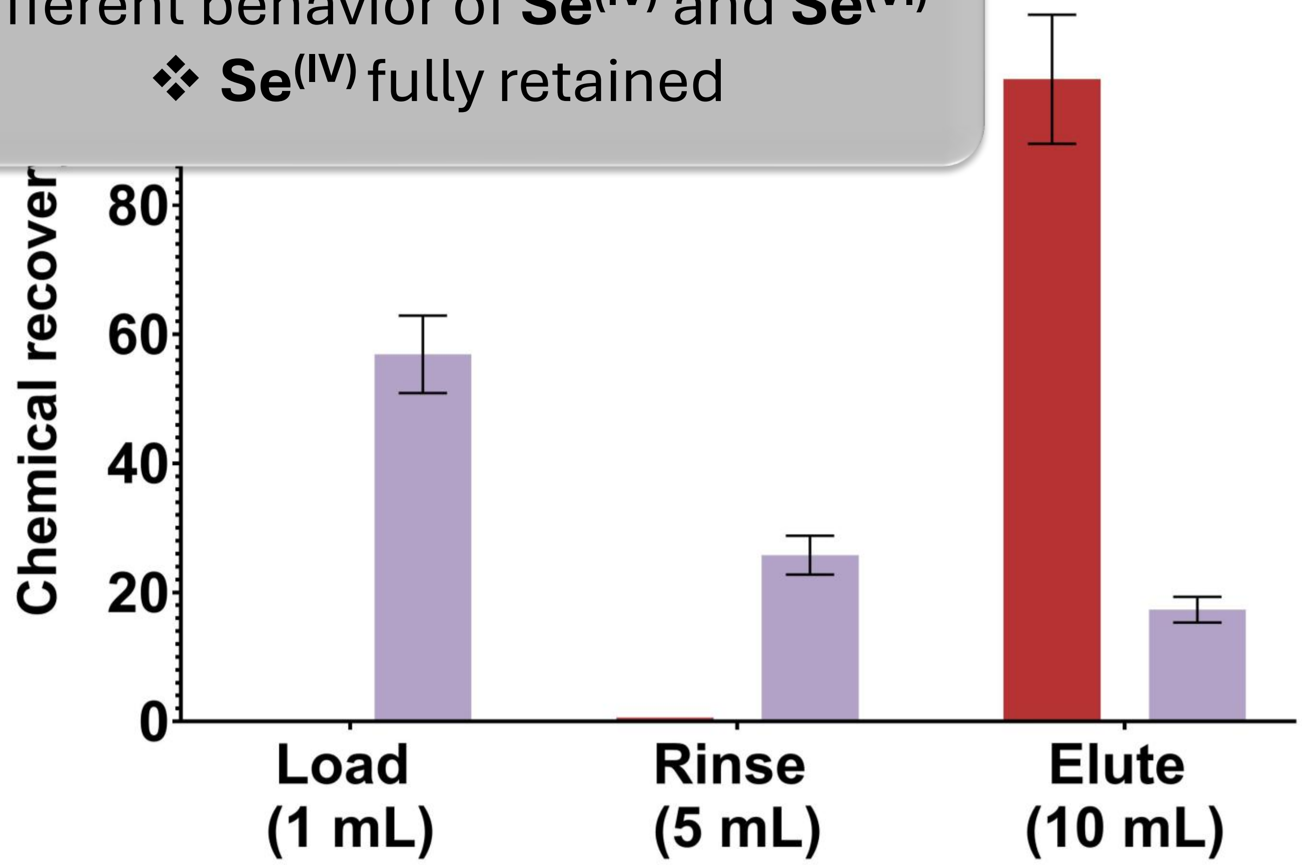
Se$^{(IV)}$ standard (2% HNO_3)

Se$^{(VI)}$ standard (in H_2O)

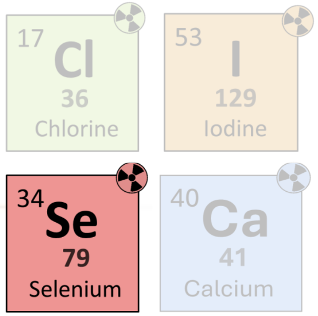
❶ 7 mL 2.5 M HCl
❷ 10 mL 2.5 M HCl
❸ 2x10 mL 1 M NaOH

Different behavior of **Se$^{(IV)}$** and **Se$^{(VI)}$**

❖ **Se$^{(IV)}$** fully retained



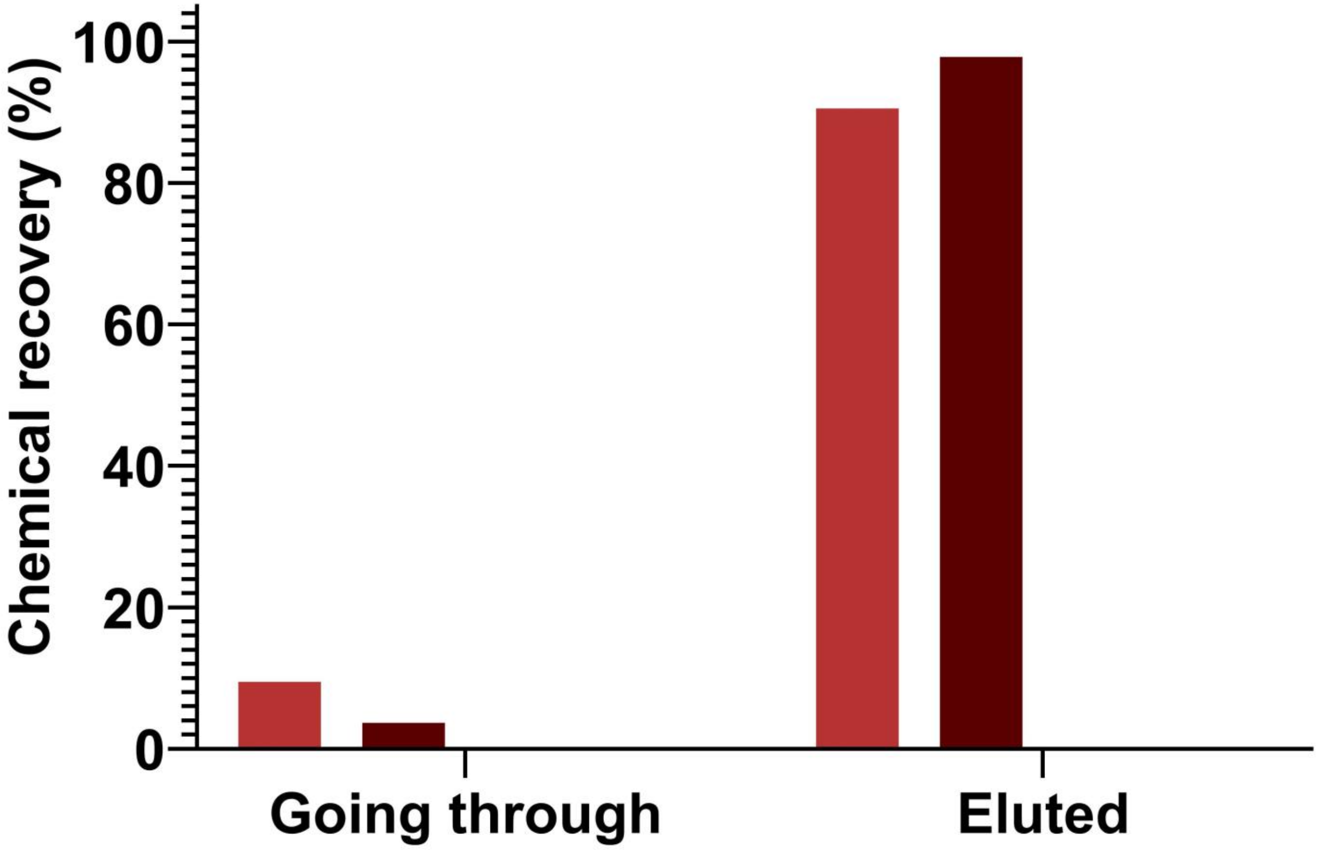
On the determination of ^{79}Se



Influence of pH on Se retention

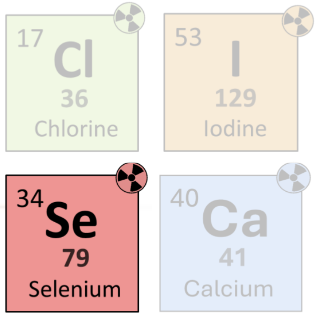
Bosca & Mot, 2021 ➔ relevance of pH on *piazselenol* formation (**pH~1.3**)

$Se^{(IV)}$ standard



1. 1 mL 2.5 M HCl
2. 5 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

On the determination of ^{79}Se



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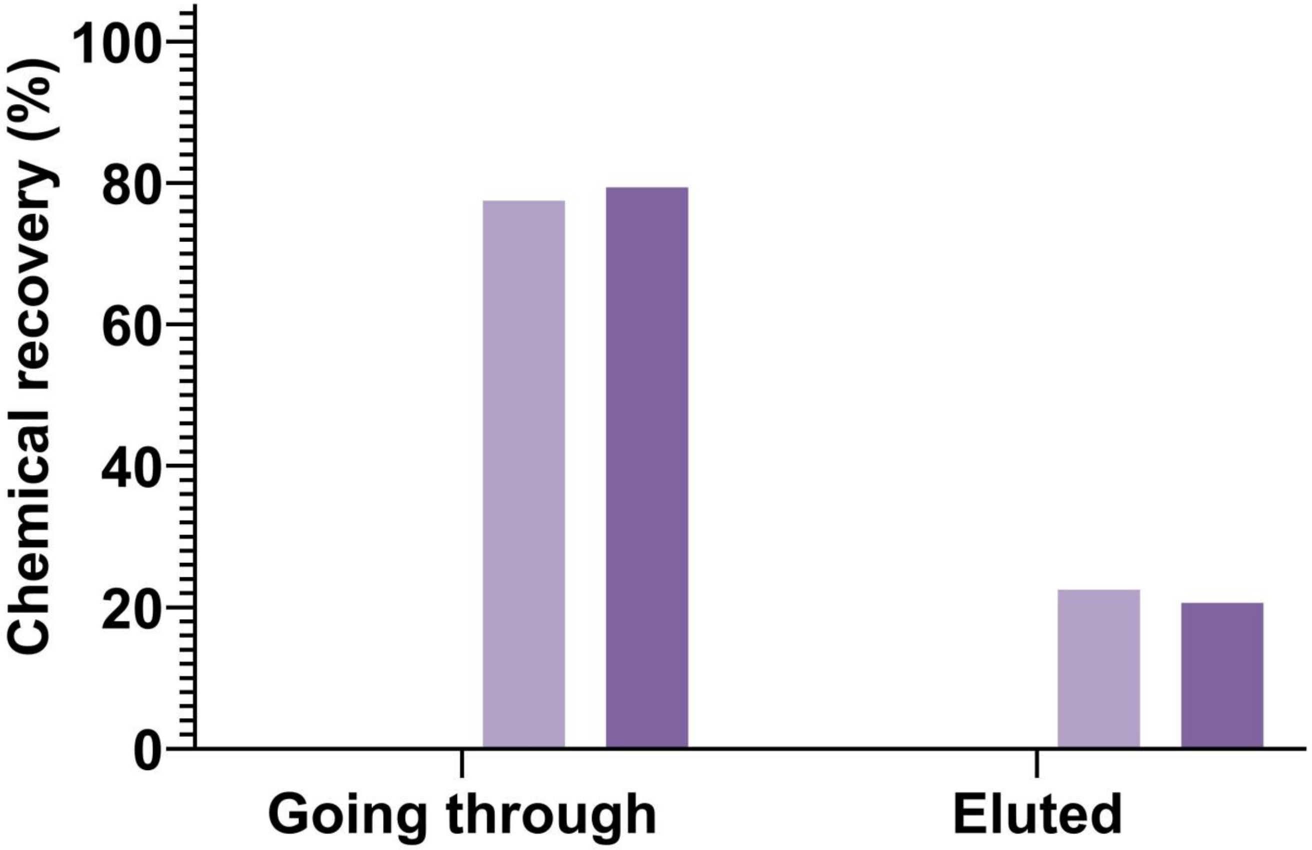
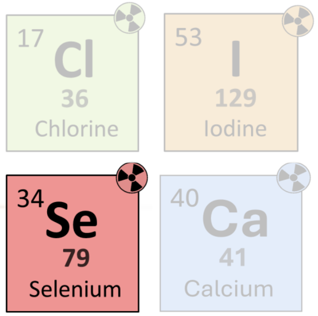
2. 5 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

$Se^{(IV)}$ standard

$Se^{(VI)}$ standard



On the determination of ^{79}Se



Influence of pH on Se retention

Bosca & Mot, 2021 → relevance of pH on *piazselenol* formation (**pH~1.3**)

1. 1 mL 2.5 M HCl
2. 5 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

$Se^{(IV)}$ standard

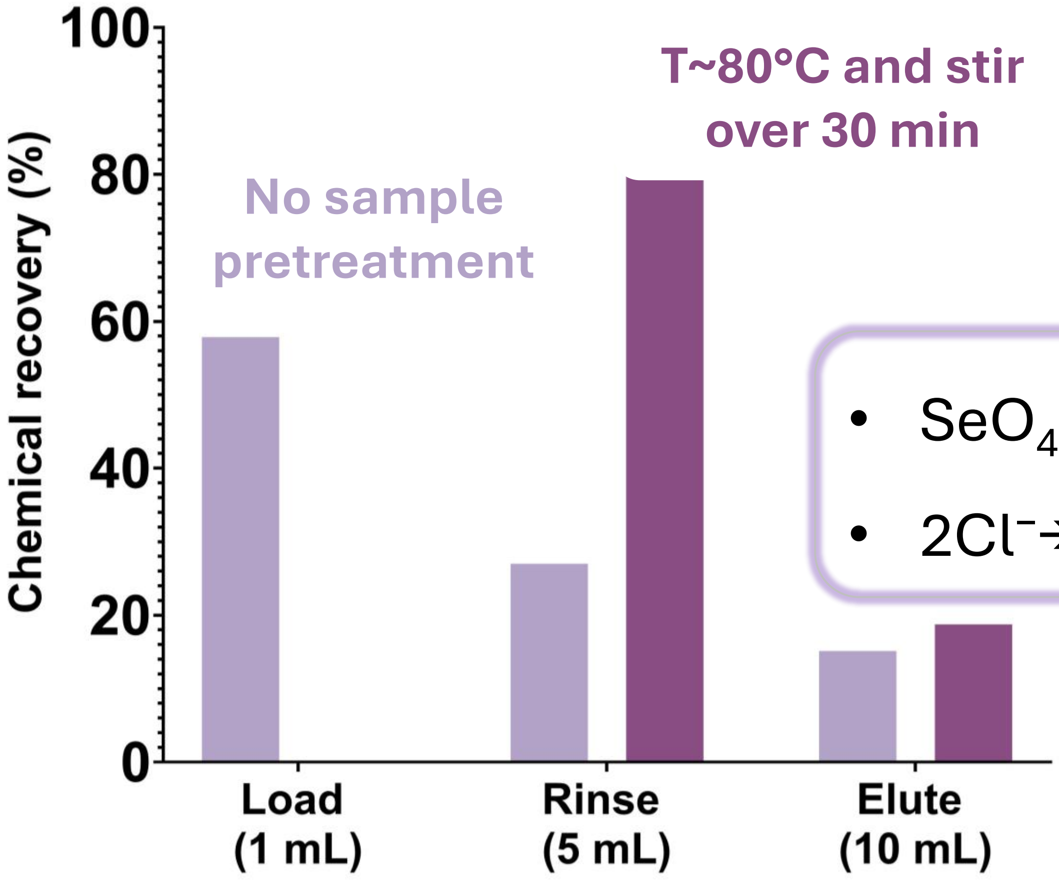
$Se^{(VI)}$ standard

Clear **differences** between Se^{+4} and Se^{+6} retention

Se **reduction cycle** still needed

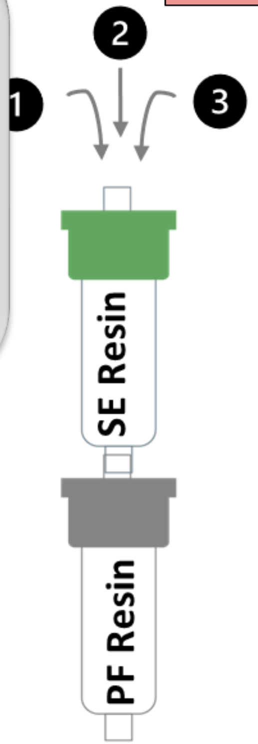
On the determination of ^{79}Se

Influence of T for Se reduction



Higher $Se^{(VI)}$ retention when warming the sample before loading

- $SeO_4^{2-} + 2H^+ + 2e^- \rightarrow SeO_3^{2-} + H_2O$
- $2Cl^- \rightarrow Cl_2 + 2e^-$



1. 1 mL 2.5 M HCl
2. 5 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH

On the determination of ^{79}Se

Influence of [HCl] for Se reduction

Ragnar Bye and Waiter Lund, 1988 ➔ relevance of HCl concentration, temperature and time on Se reduction

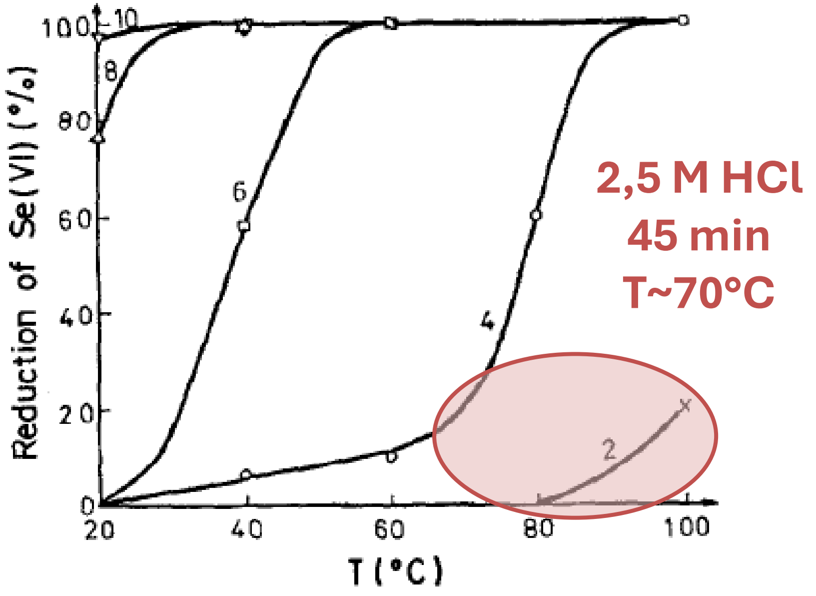


Fig. 2. Reduction of 1.6 μg Se(VI) with 10 ml HCl of various concentrations (mol/l) after 30 min in dependence on temperature

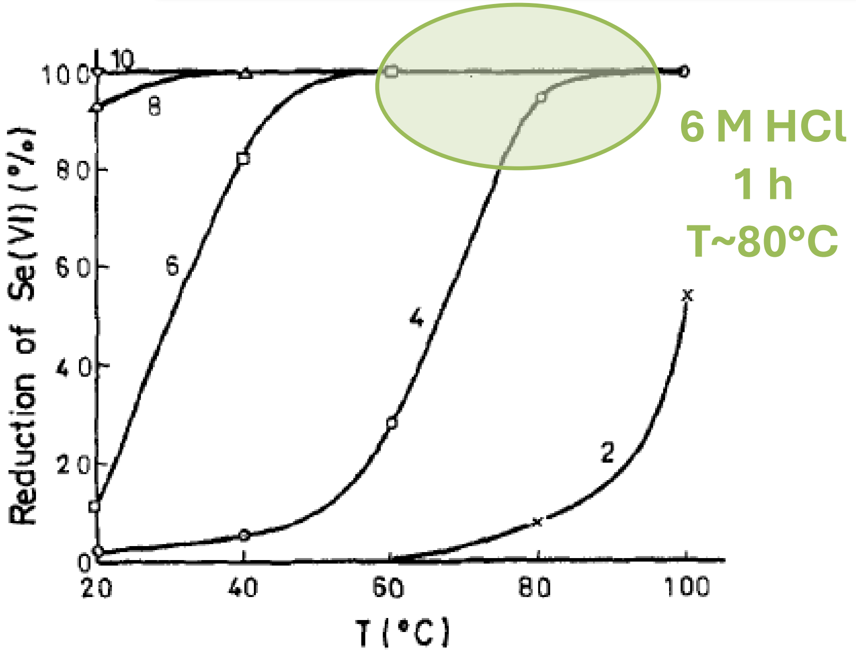
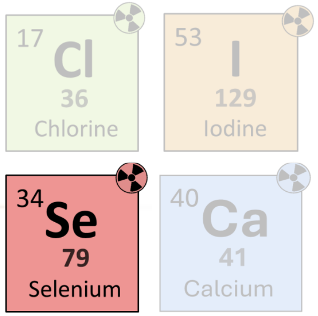
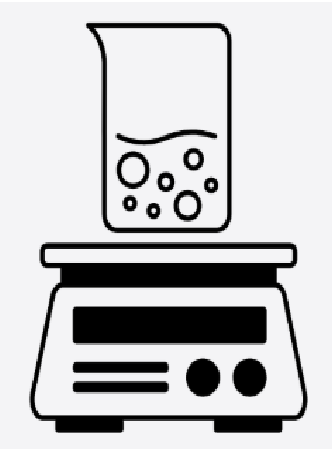


Fig. 4. Reduction of 1.6 μg Se(VI) with 10 ml HCl of various concentrations (mol/l) after 2 h in dependence on temperature

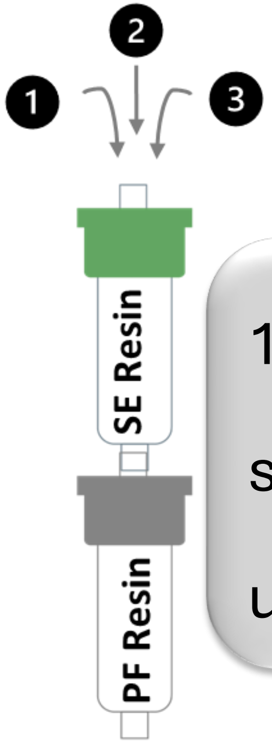
On the determination of ^{79}Se



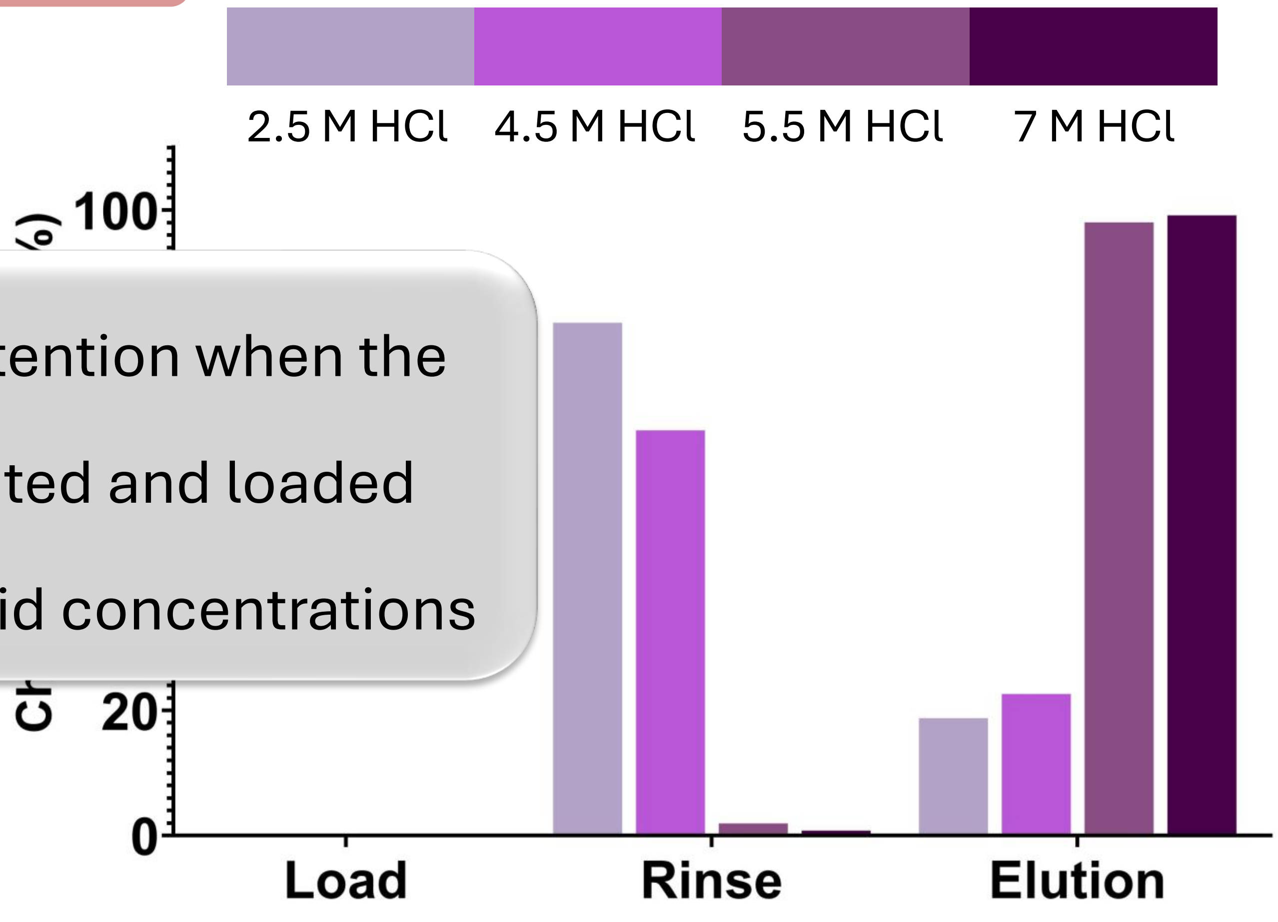
Influence of [HCl] for Se reduction



T=80°C
60 min



1. Load
2. 5 mL 2.5 M HCl
3. 2x10 mL 1 M NaOH



100% $Se^{(VI)}$ retention when the sample is heated and loaded under high acid concentrations

On the determination of ^{79}Se

Interferences removal

$Se^{(VI)}$
Fe
Ni
Co

T=80°C
60 min

Step	Fraction
Load	1 mL →
Rinse	5 mL 2,5 M HCl
Elution	5 mL 1 M NaOH
Elution	5 mL 1 M NaOH

5.5 M HCl
7 M HCl

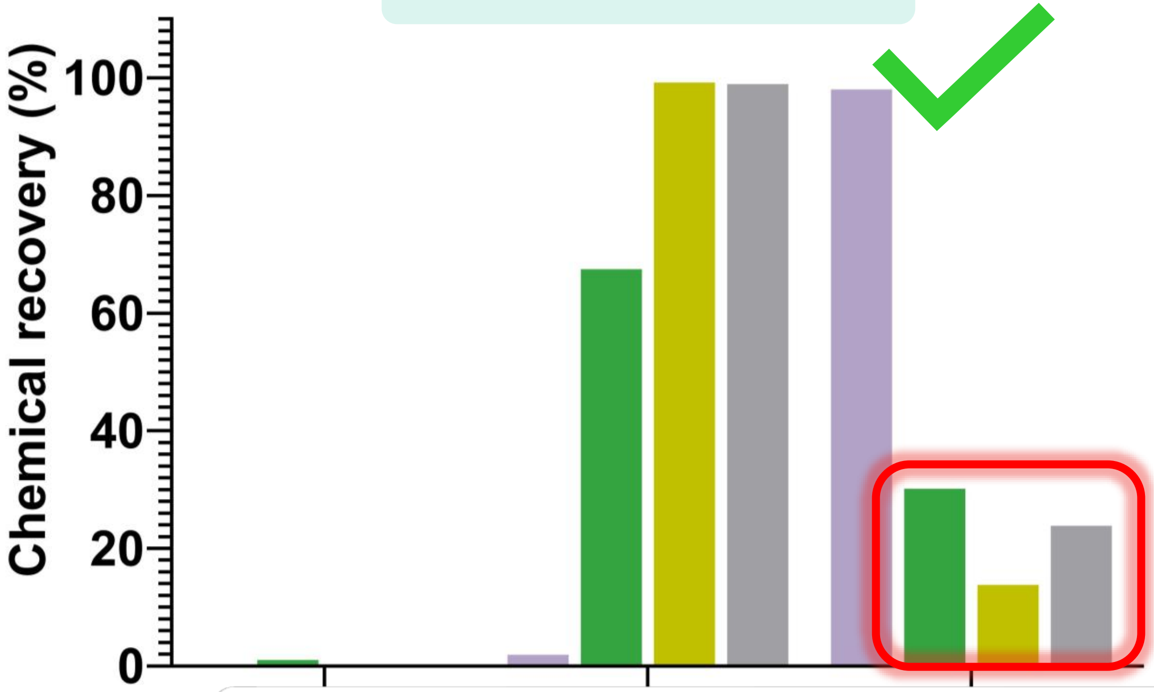
On the determination of ^{79}Se

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

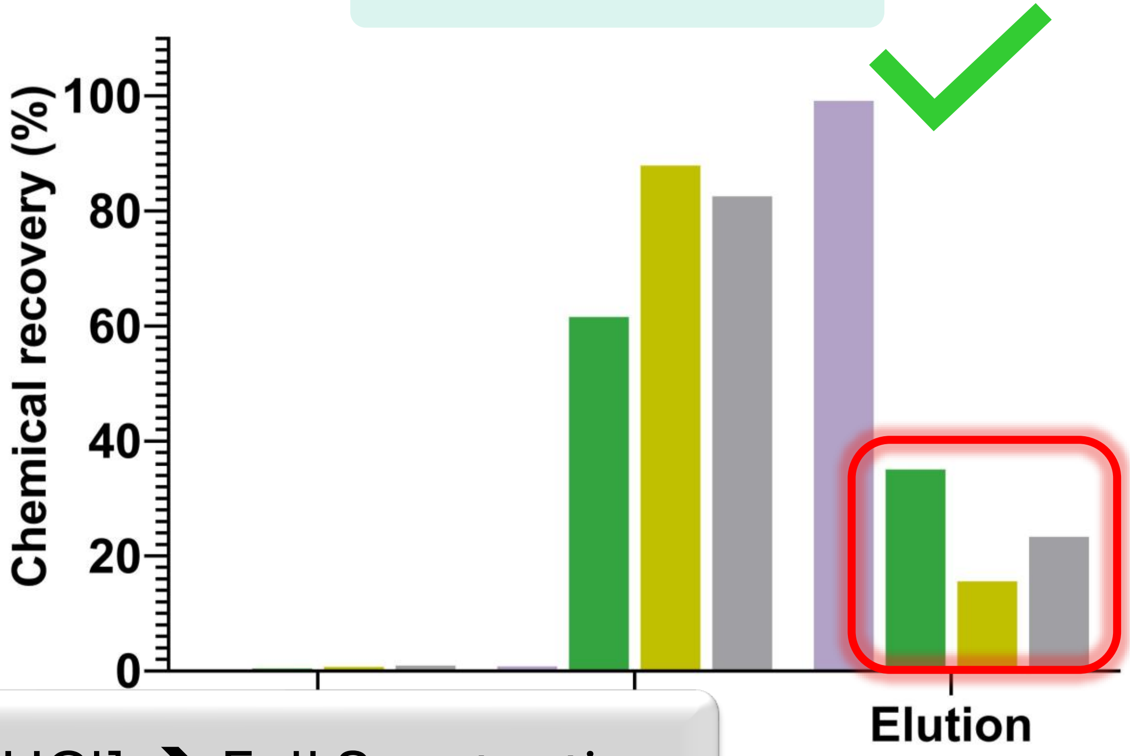
Interferences removal

Se(VI) Fe Ni Co

Load : 5.5 M HCl



Load : 7 M HCl



Se(VI) fully reduced when using ↑ [HCl] ➔ Full Se retention

On the determination of ^{79}Se

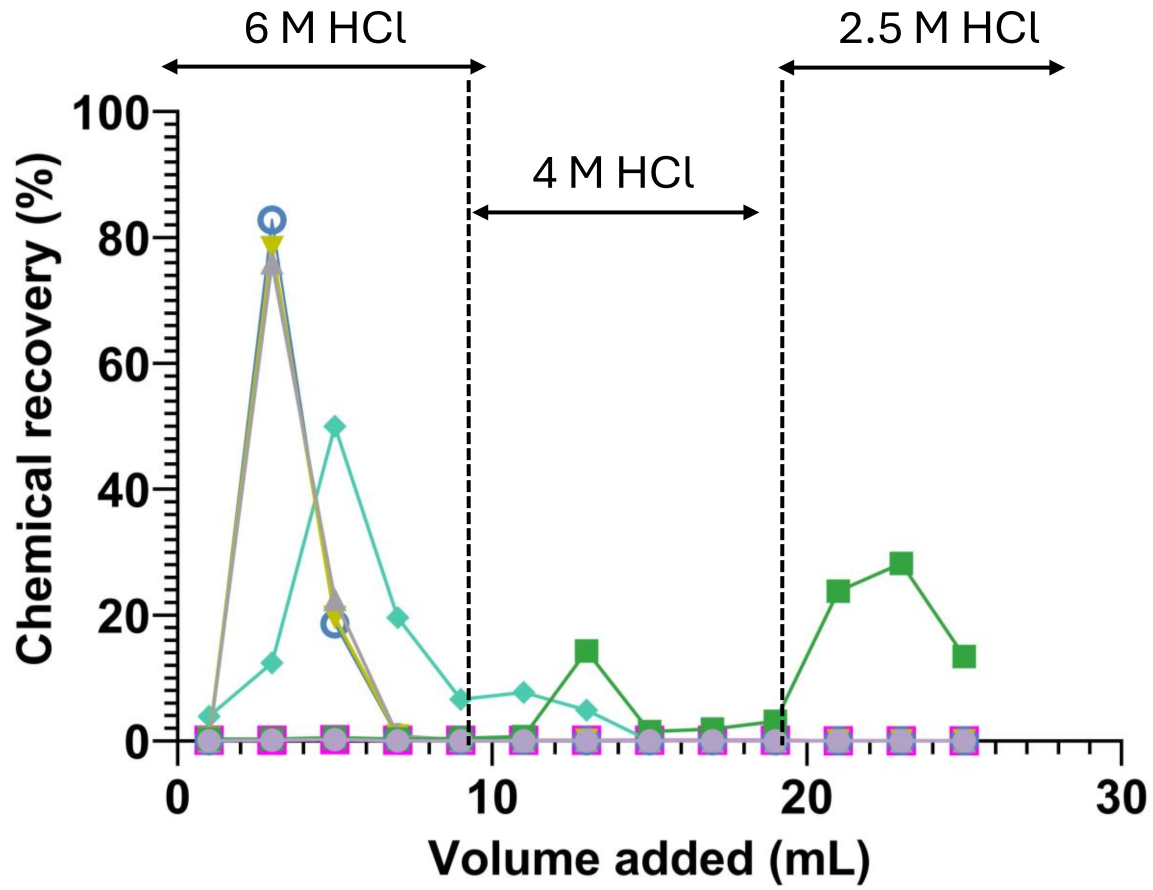
17 Cl 36 Chlorine

53 I 129 Iodine

34 Se 79 Selenium

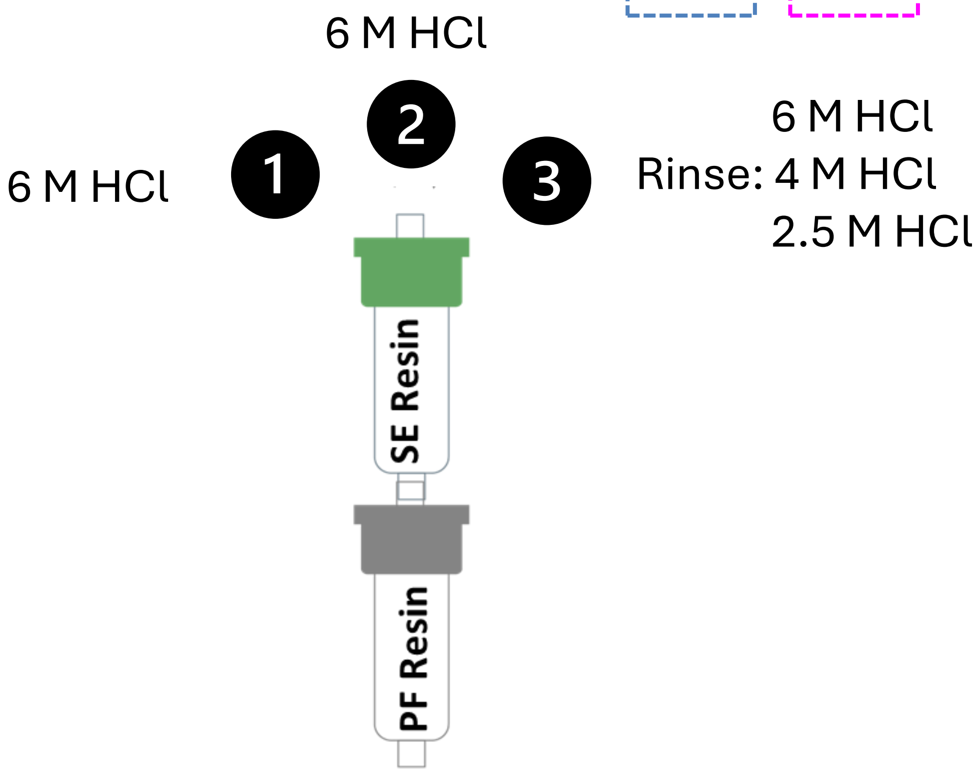
40 Ca 41 Calcium

Interferences removal

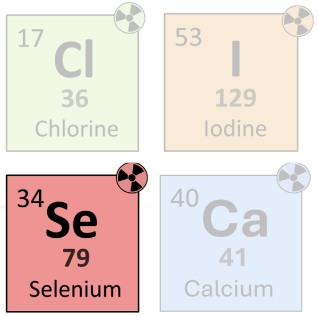


Se(VI) Fe Ni Co Cu

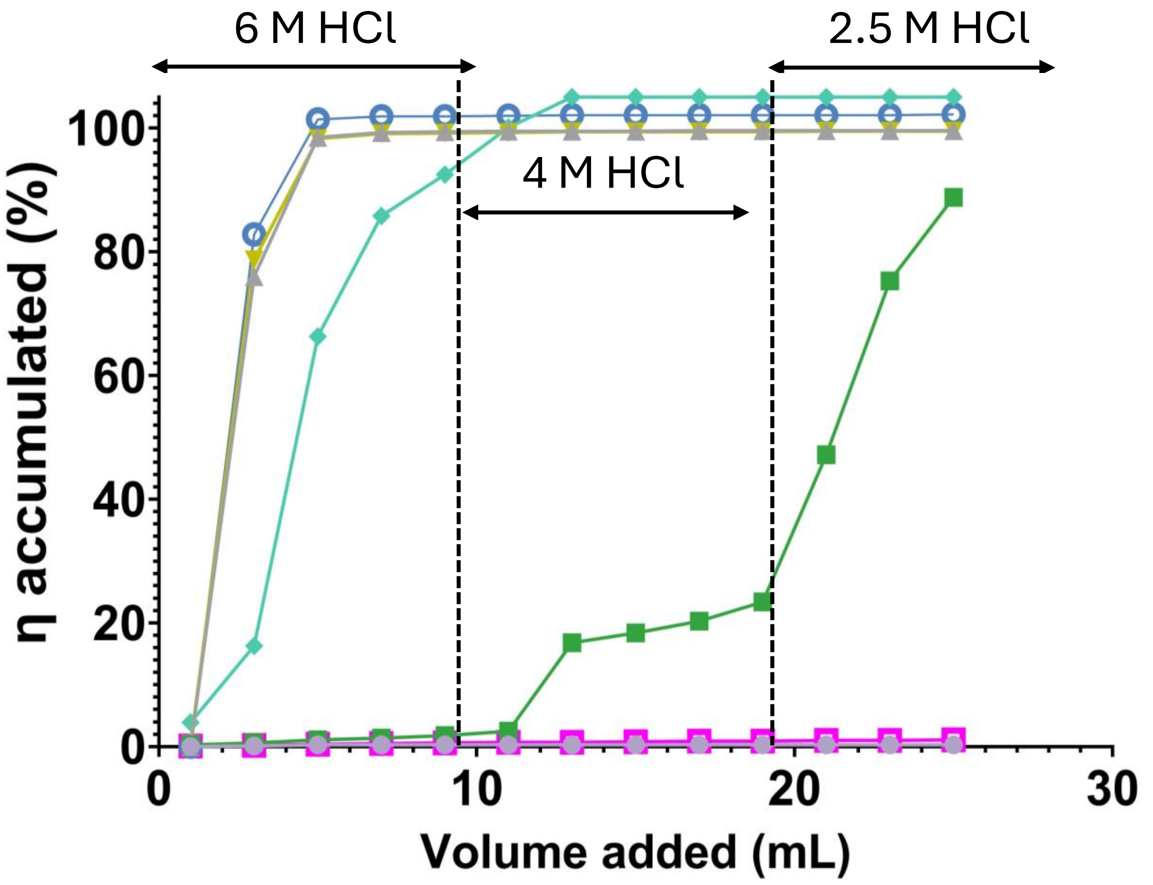
Ba Sn

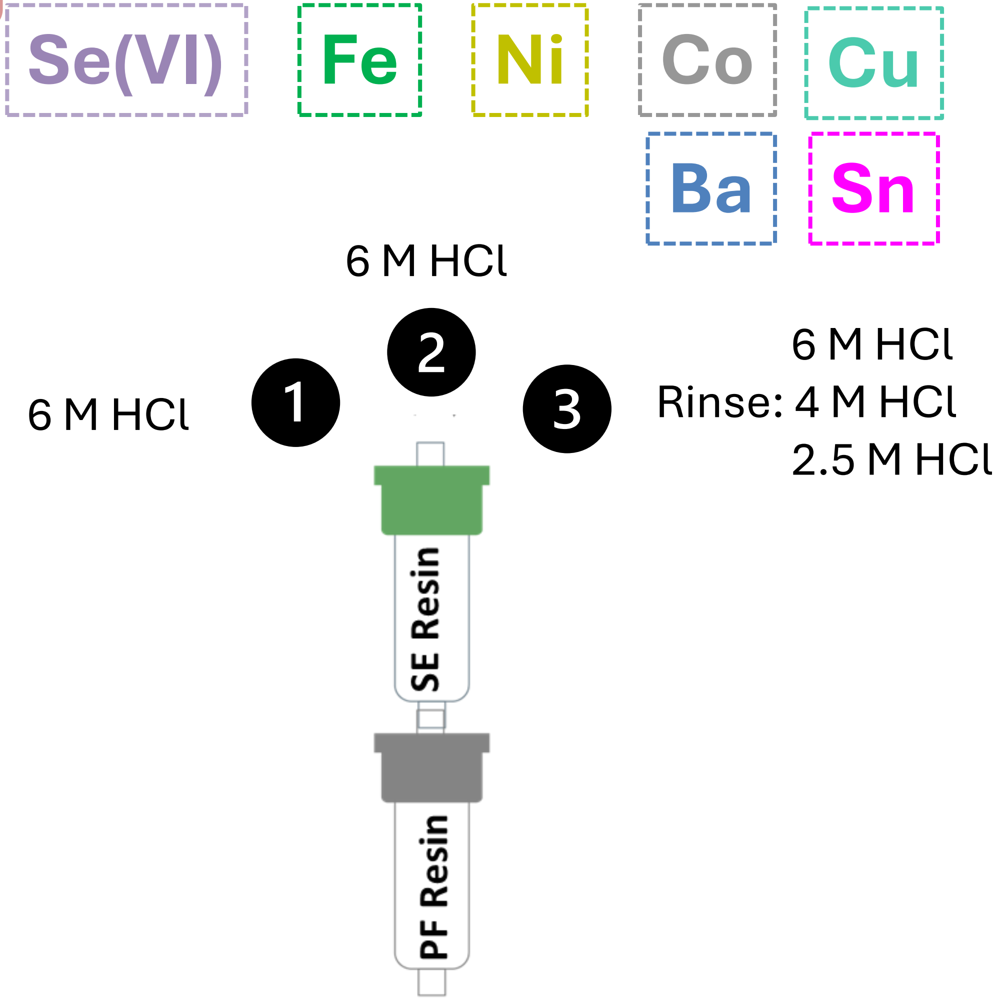


On the determination of ^{79}Se

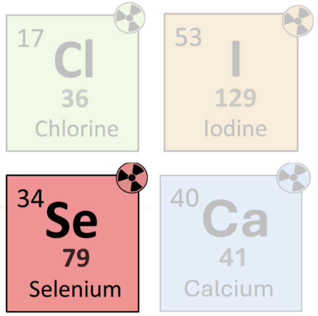


Interferences removal

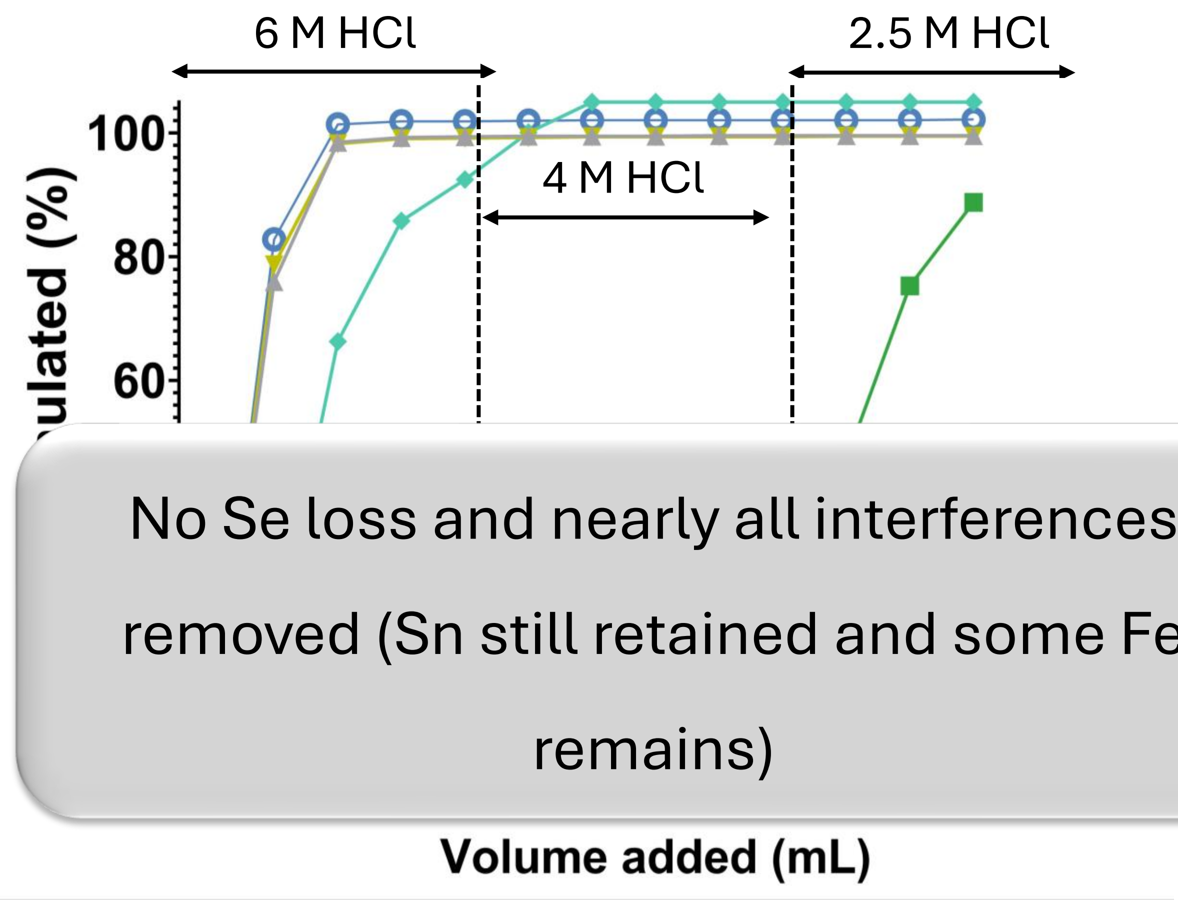




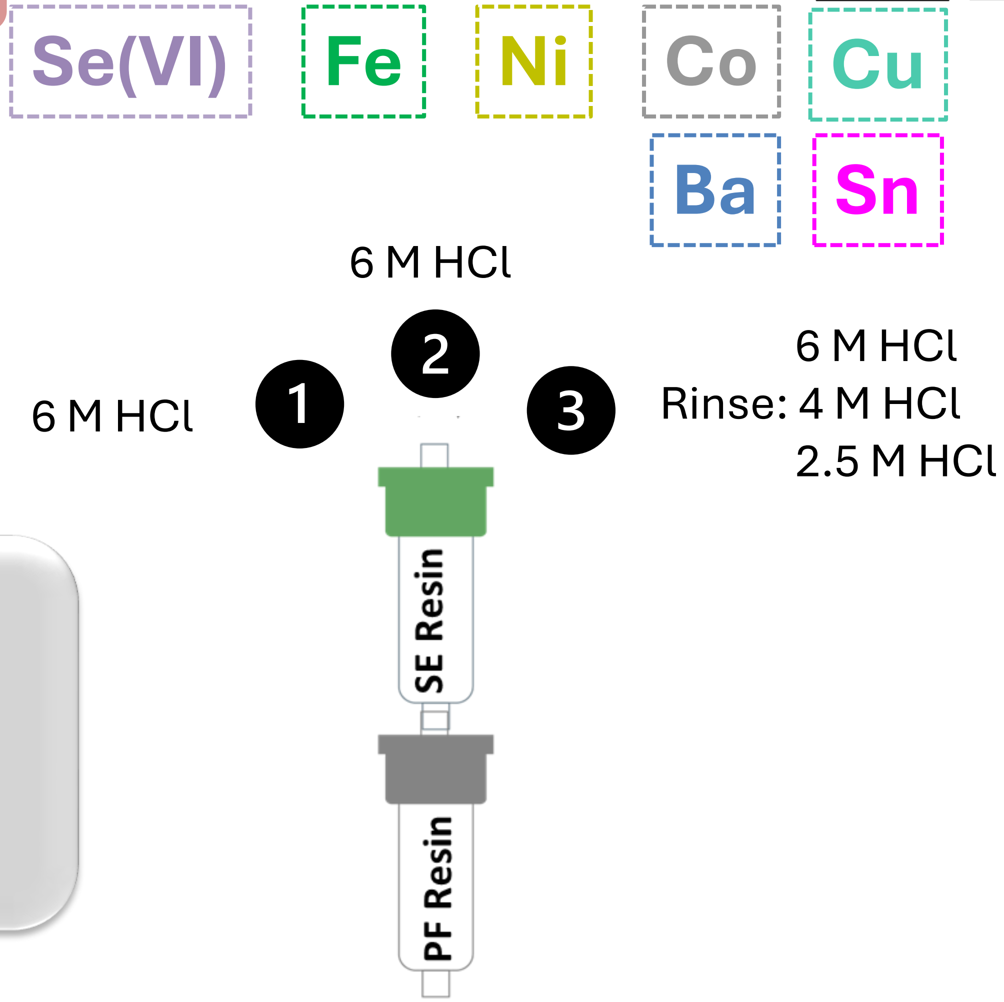
On the determination of ^{79}Se



Interferences removal



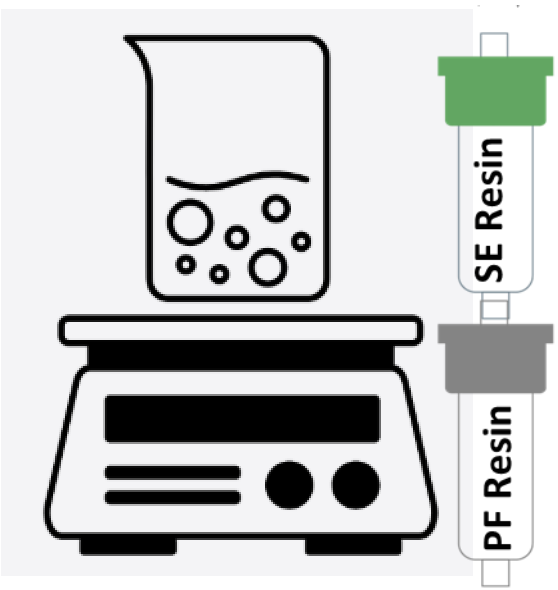
No Se loss and nearly all interferences removed (Sn still retained and some Fe remains)



On the determination of ^{79}Se

Final chemical separation procedure

Se(IV) Fe Ni
Co Sn



T=80°C
60 min

Se(VI) Fe Ni
Co Sn

T=80°C
60 min

1 Load

6 M HCl

2 Rinse

8 mL 6 M HCl

8 mL 2.5 M HCl

3 Elution

2 * 5 mL 1 M NaOH

On the determination of ^{79}Se

17 Cl 36 Chlorine

53 I 129 Iodine

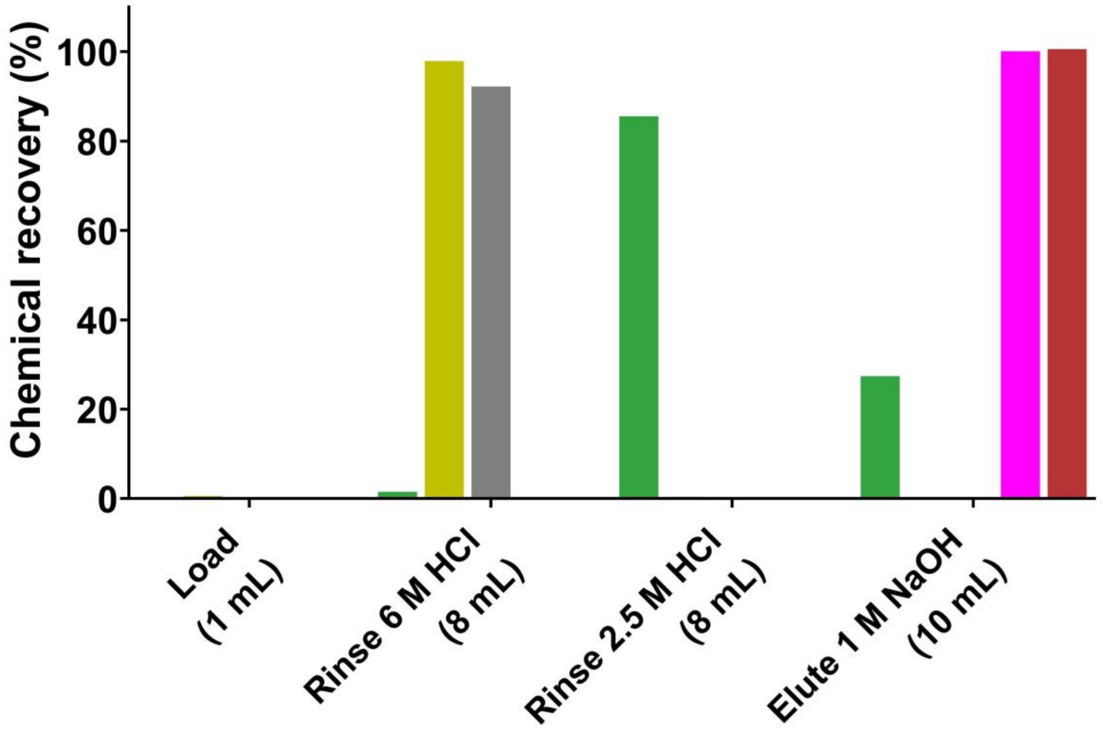
34 Se 79 Selenium

40 Ca 41 Calcium

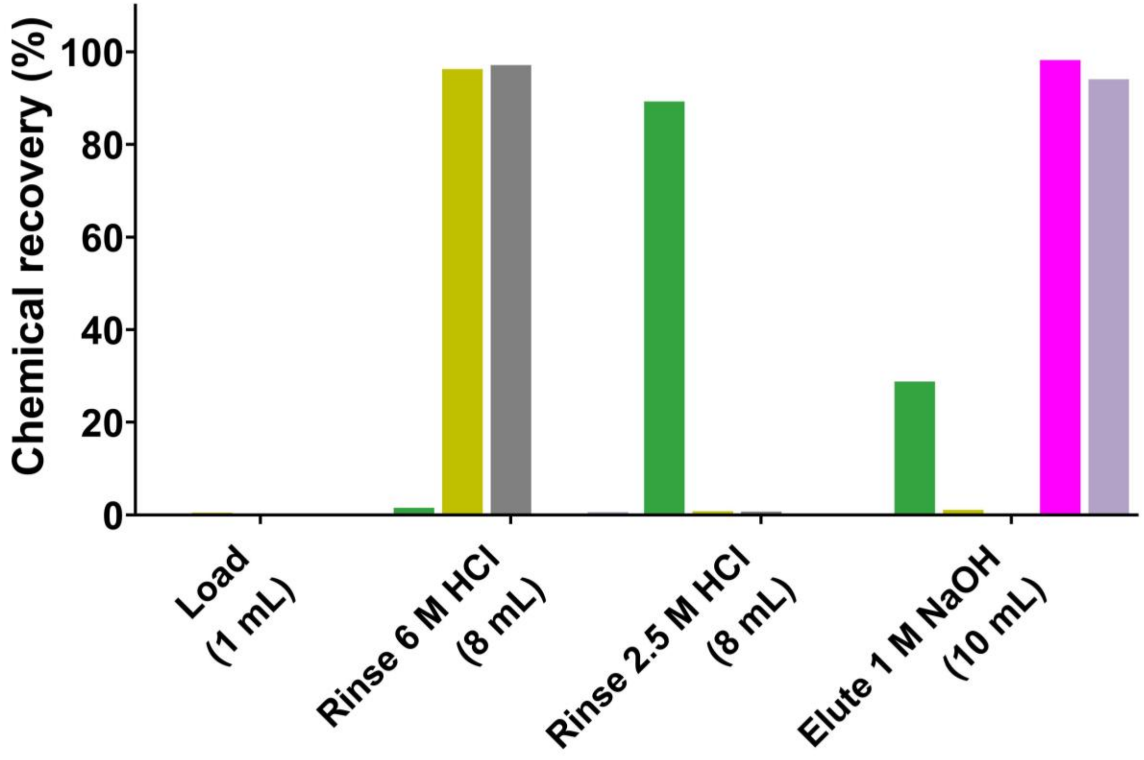
Final chemical separation procedure

Fe Ni Co Sn

Se(IV)



Se(VI)



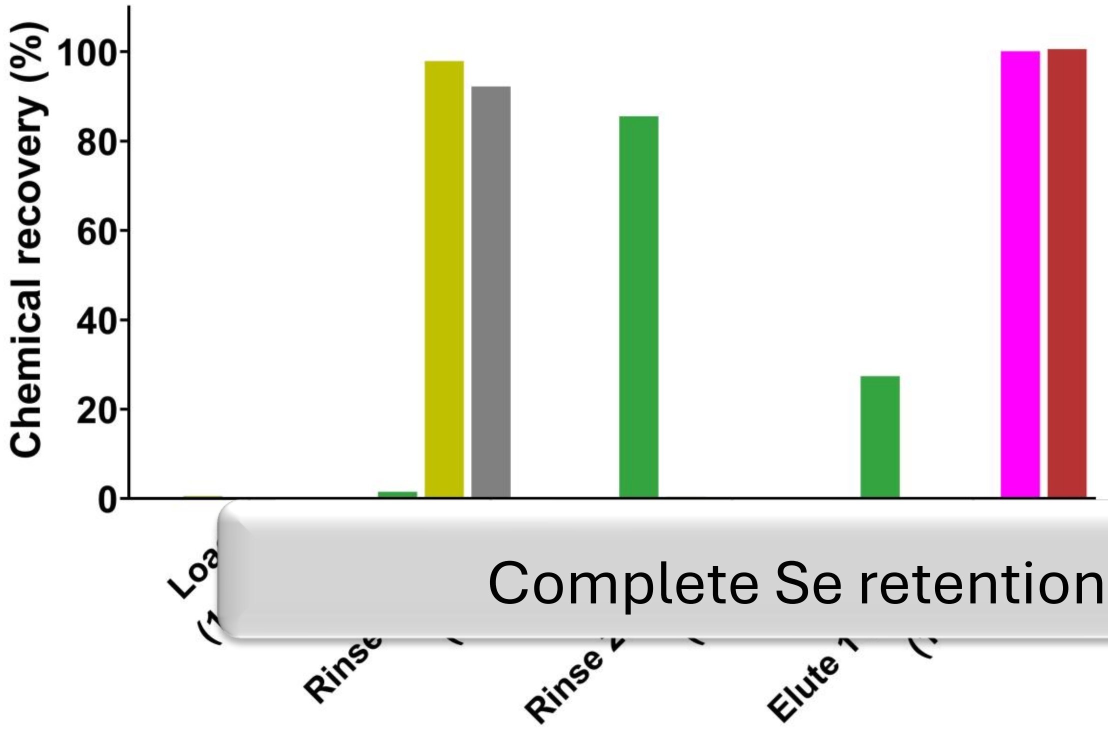
On the determination of ^{79}Se

17 Cl 36 Chlorine | 53 I 129 Iodine | 34 Se 79 Selenium | 40 Ca 41 Calcium

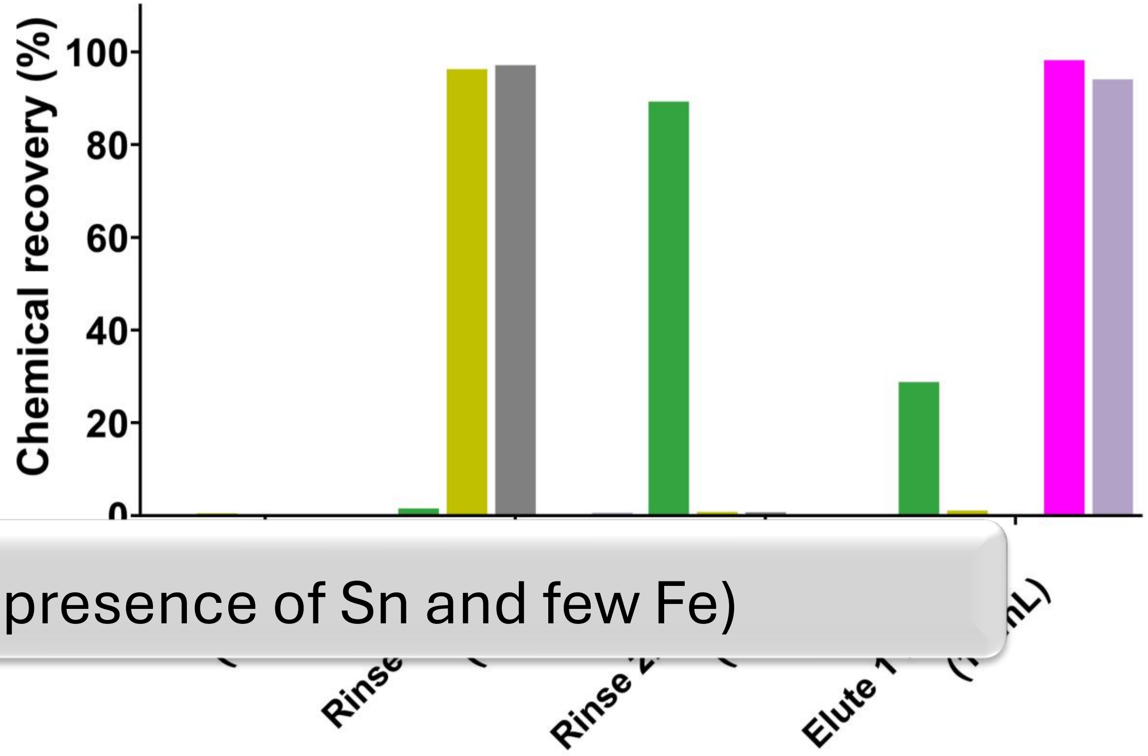
Final chemical separation procedure

Fe Ni Co Sn

Se(IV)

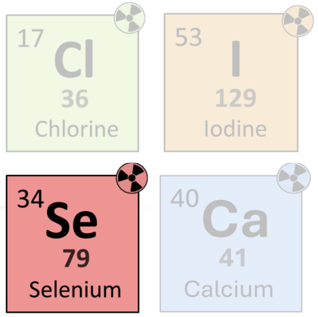


Se(VI)



Complete Se retention (presence of Sn and few Fe)

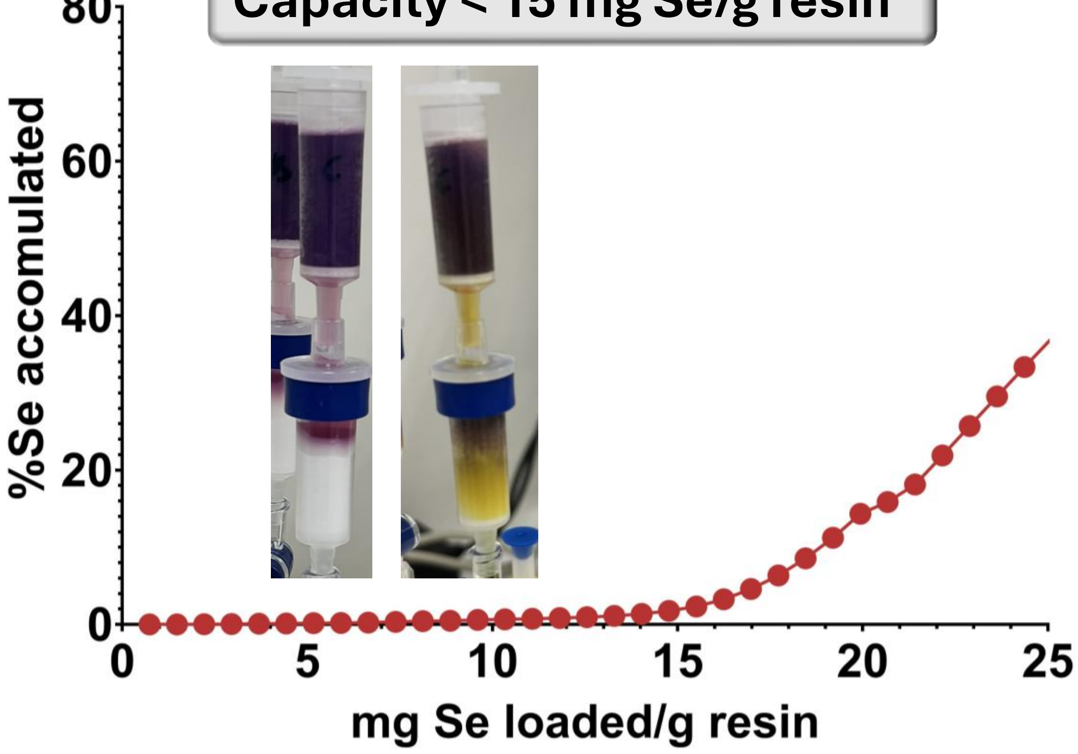
On the determination of ^{79}Se



Capacity and breakthrough volume

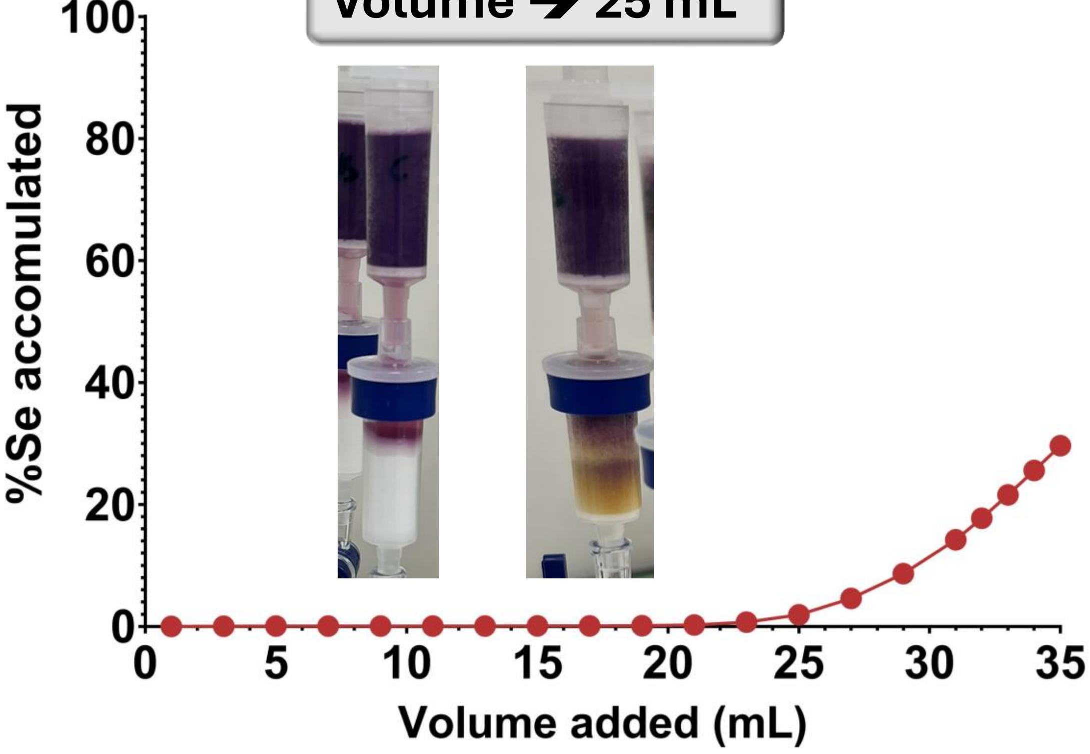
Capacity

Capacity < 15 mg Se/g resin



Breakthrough volume

Volume ➔ 25 mL

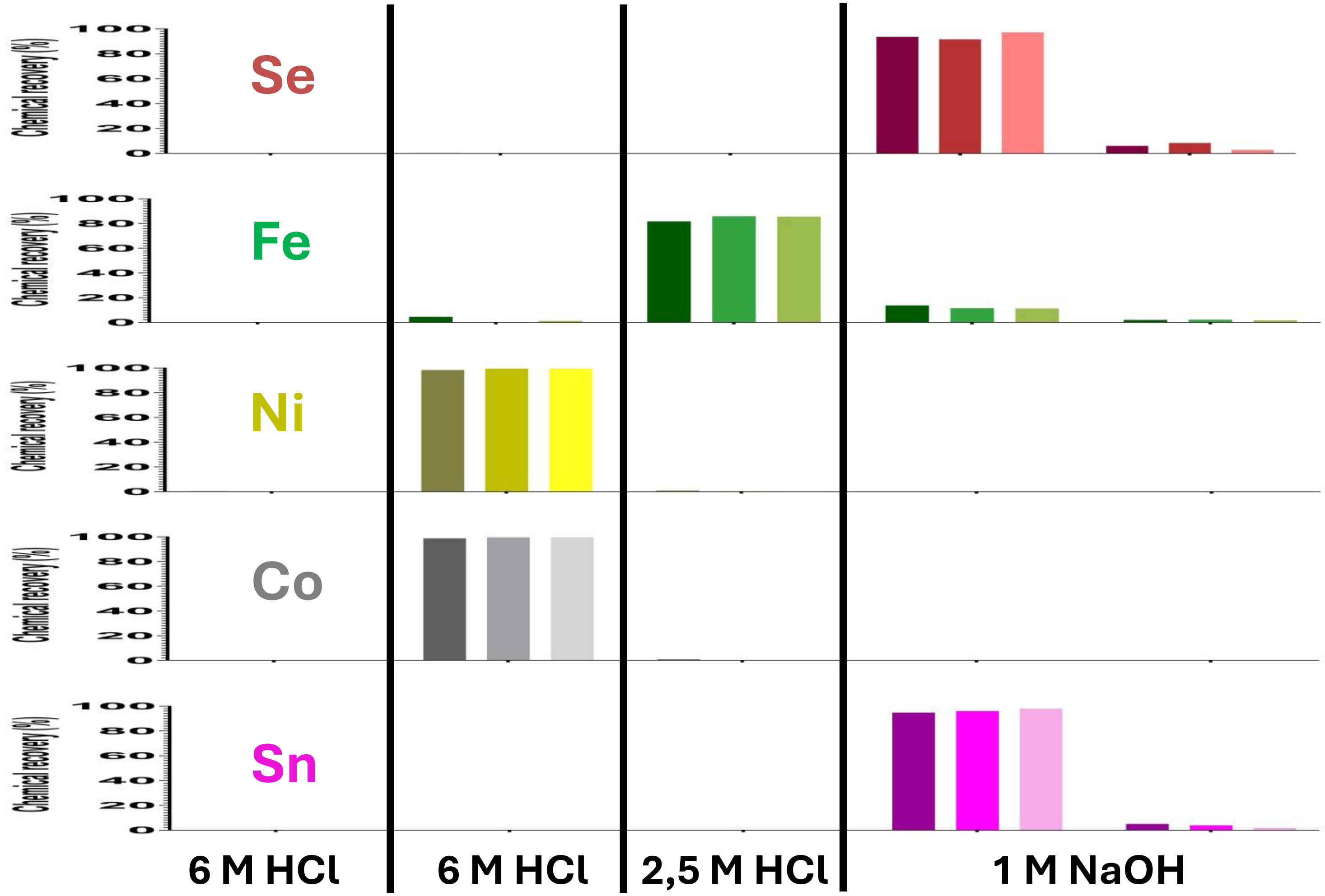


On the determination of ^{79}Se

17 Cl 36 Chlorine
53 I 129 Iodine
34 Se 79 Selenium
40 Ca 41 Calcium

Ageing of the resin

2022 → 2024 → 2025

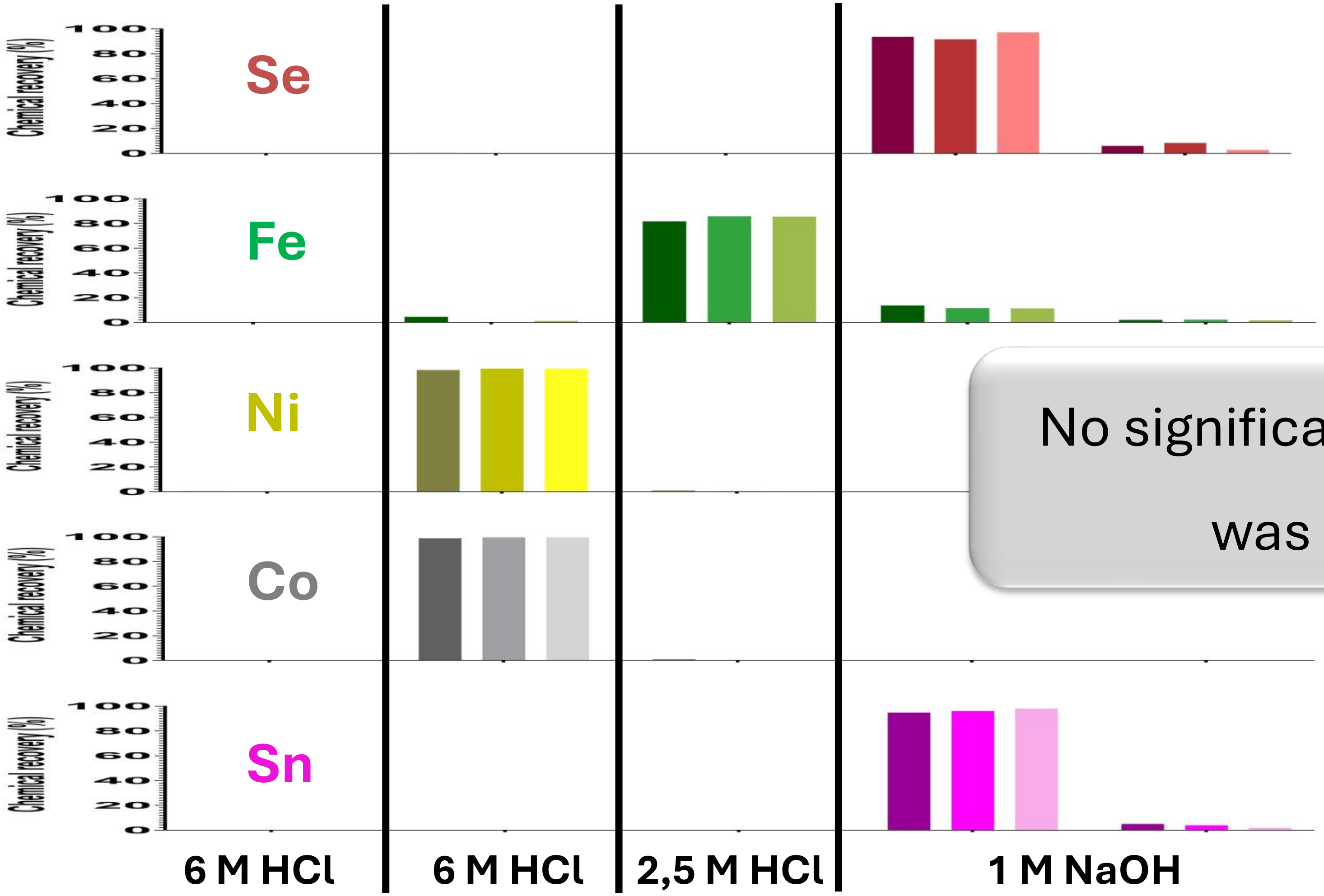


2022 2025

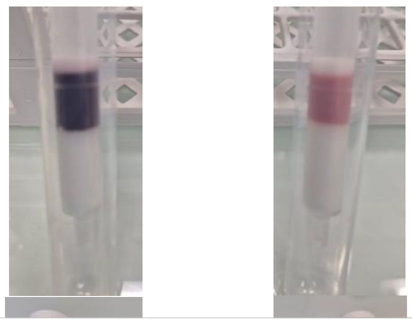
On the determination of ^{79}Se

Ageing of the resin

2022 > 2024 > 2025

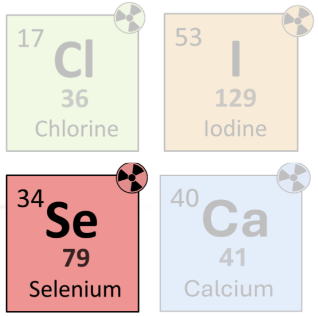


2022 2025

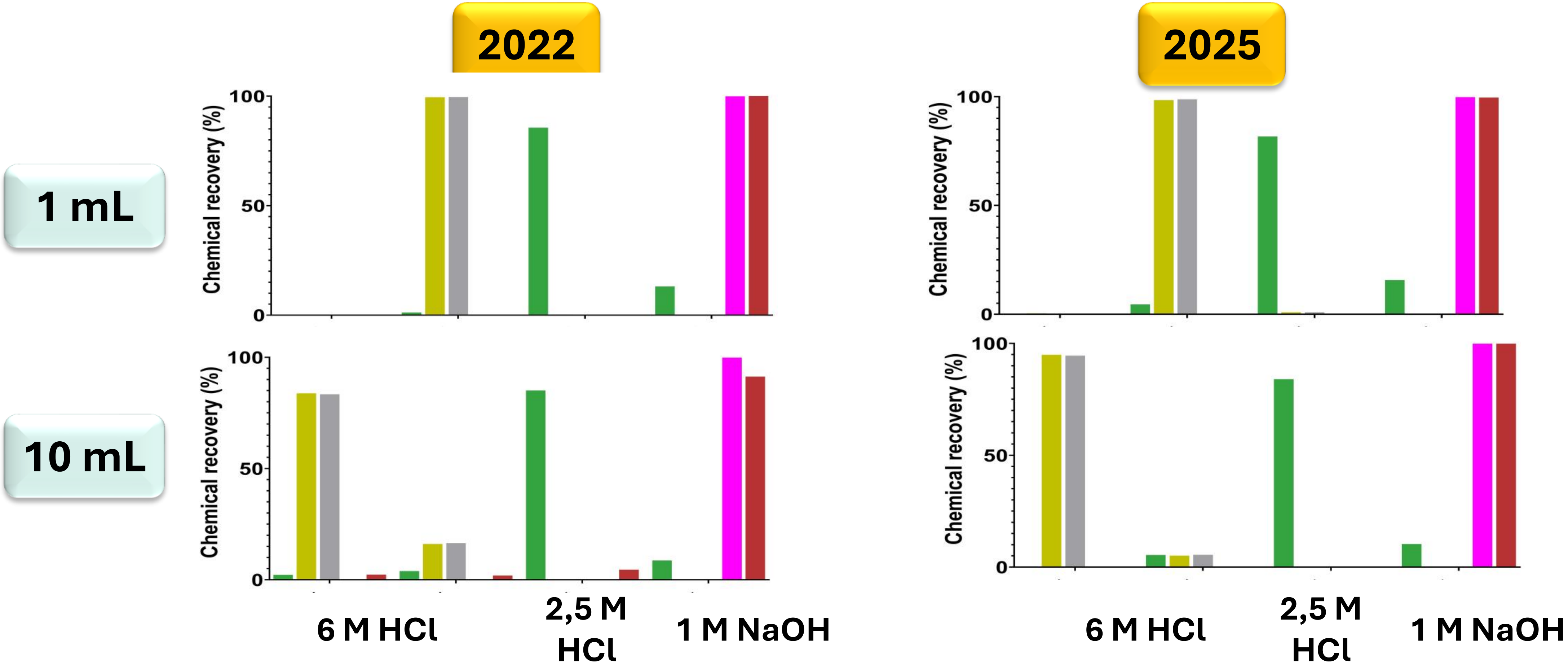


No significant difference in resin retention was observed for aged resin

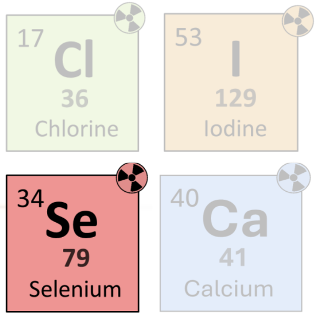
On the determination of ^{79}Se



Ageing of the resin



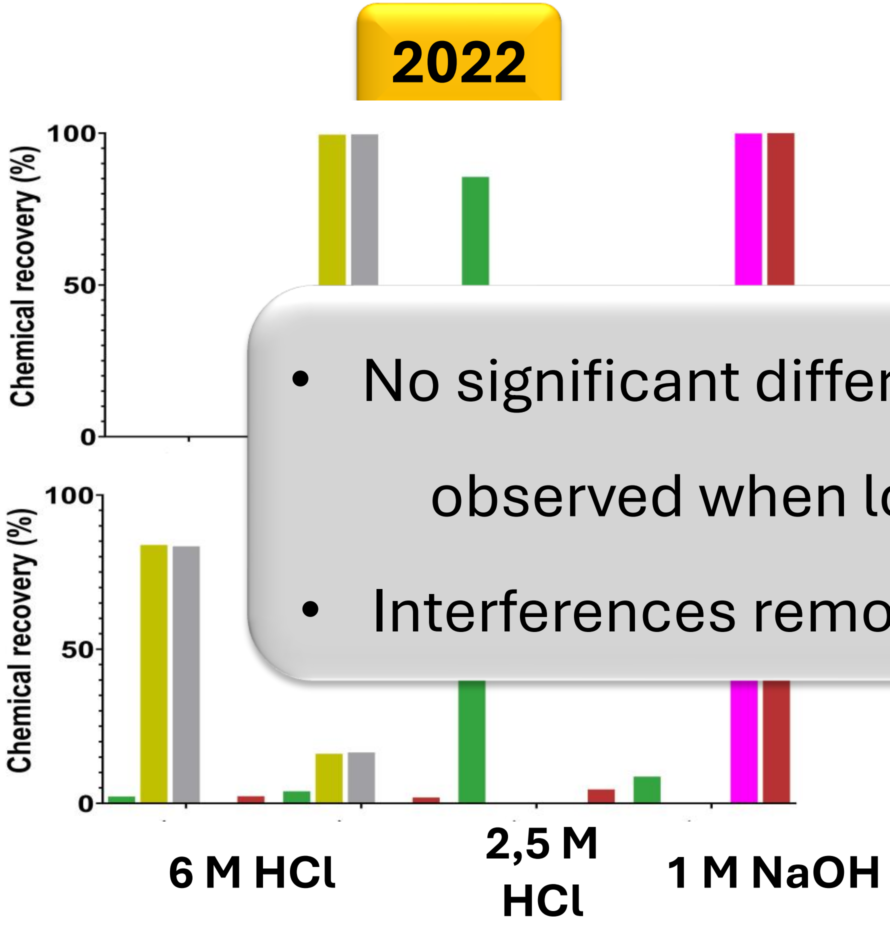
On the determination of ^{79}Se

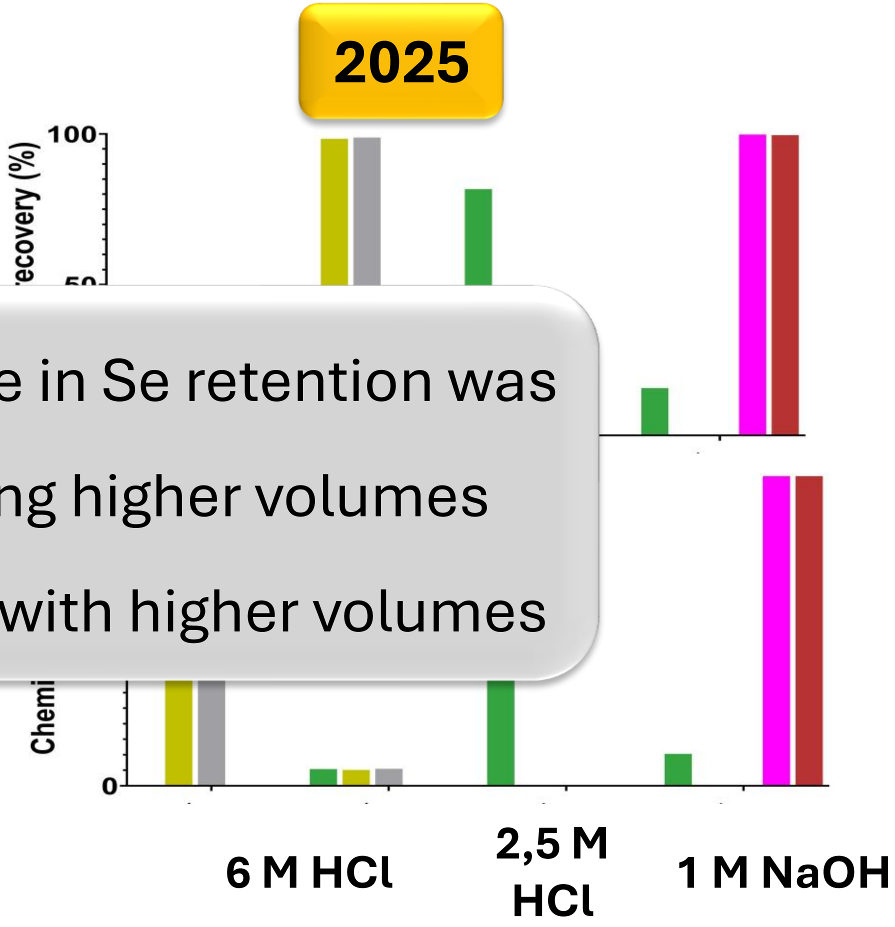


Ageing of the resin

1 mL

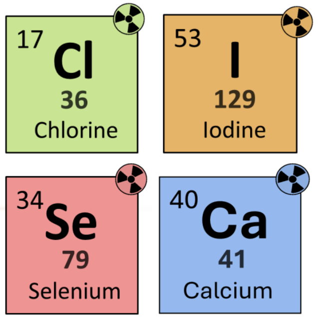
10 mL





- No significant difference in Se retention was observed when loading higher volumes
- Interferences removed with higher volumes

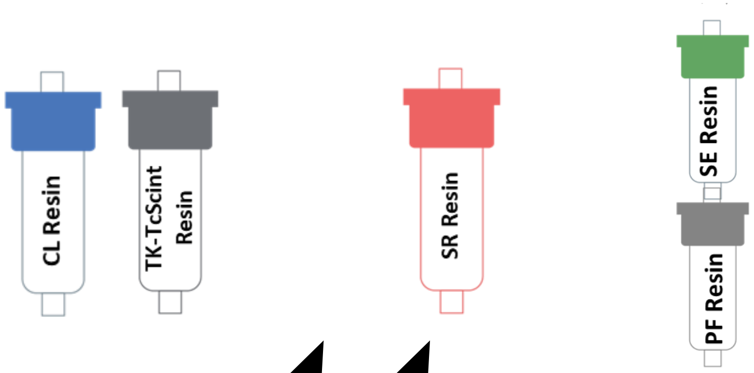
Overview



Develop and validate reliable analytical methods for the accurate determination of specific DTM radionuclides

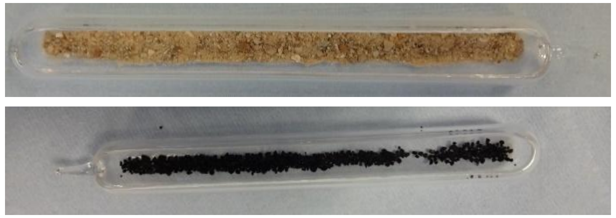
Challenges

Interferences



Low detection limit **(DL)** Clearance level

Variety of matrices



Turnaround time (TAT) Results in about 24-48 h

Thank you very much for your attention!

Inés Llopart, Steffen Happel, Alex Tarancón

03-11-2025