1st International Conference on Radioanalytical and Nuclear Chemistry
April 10–15, 2016 / Budapest, Hungary

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Analysis of ^{210}Pb in water samples with plastic scintillation resins.

Lluch, E.[1]; Barreda, J.[1]; **Tarancón, A.[1]**; Bagán, H.[2]; García, J .F.[1]

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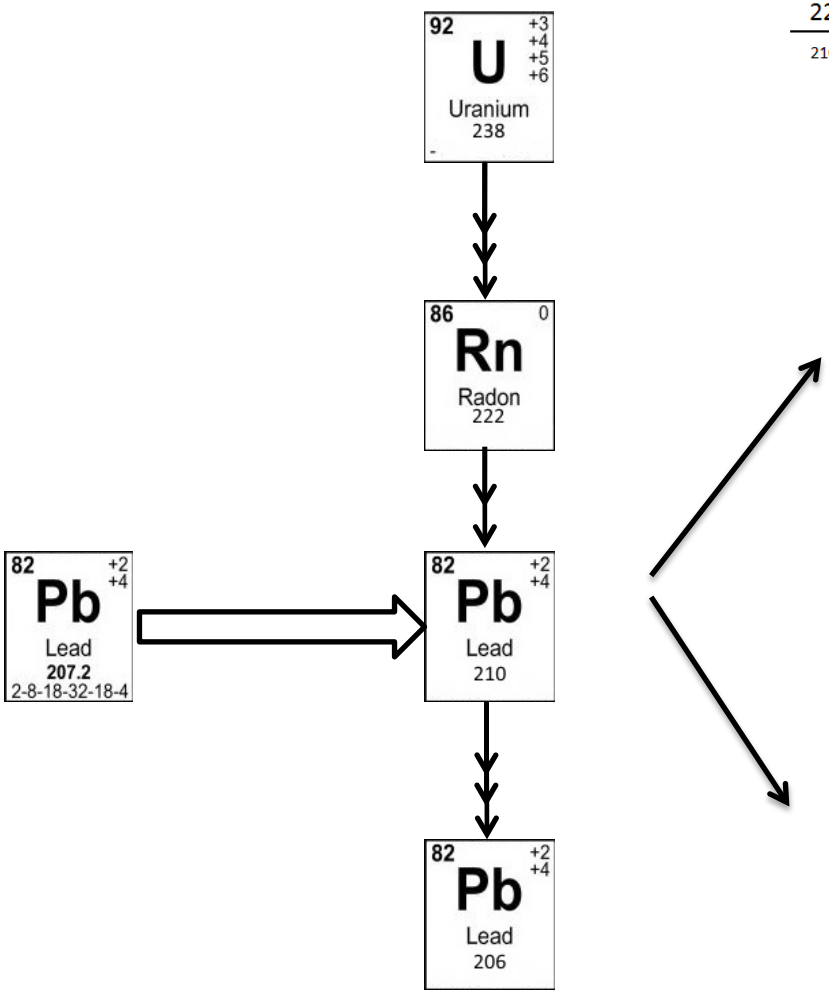

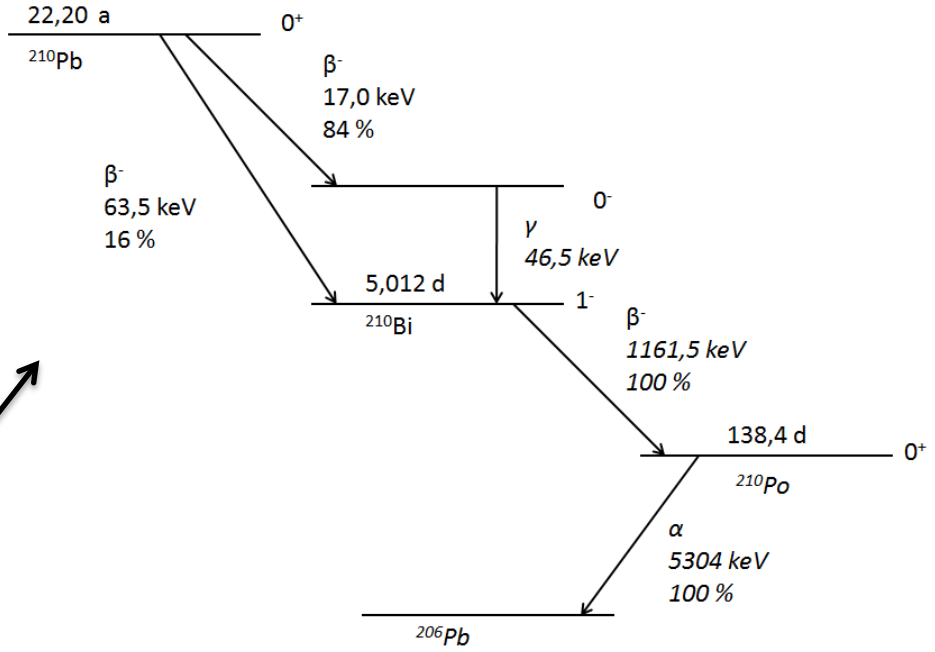

- Radiation Protection
- Chronology

Analysis of ^{210}Pb in water samples with plastic scintillation resins.

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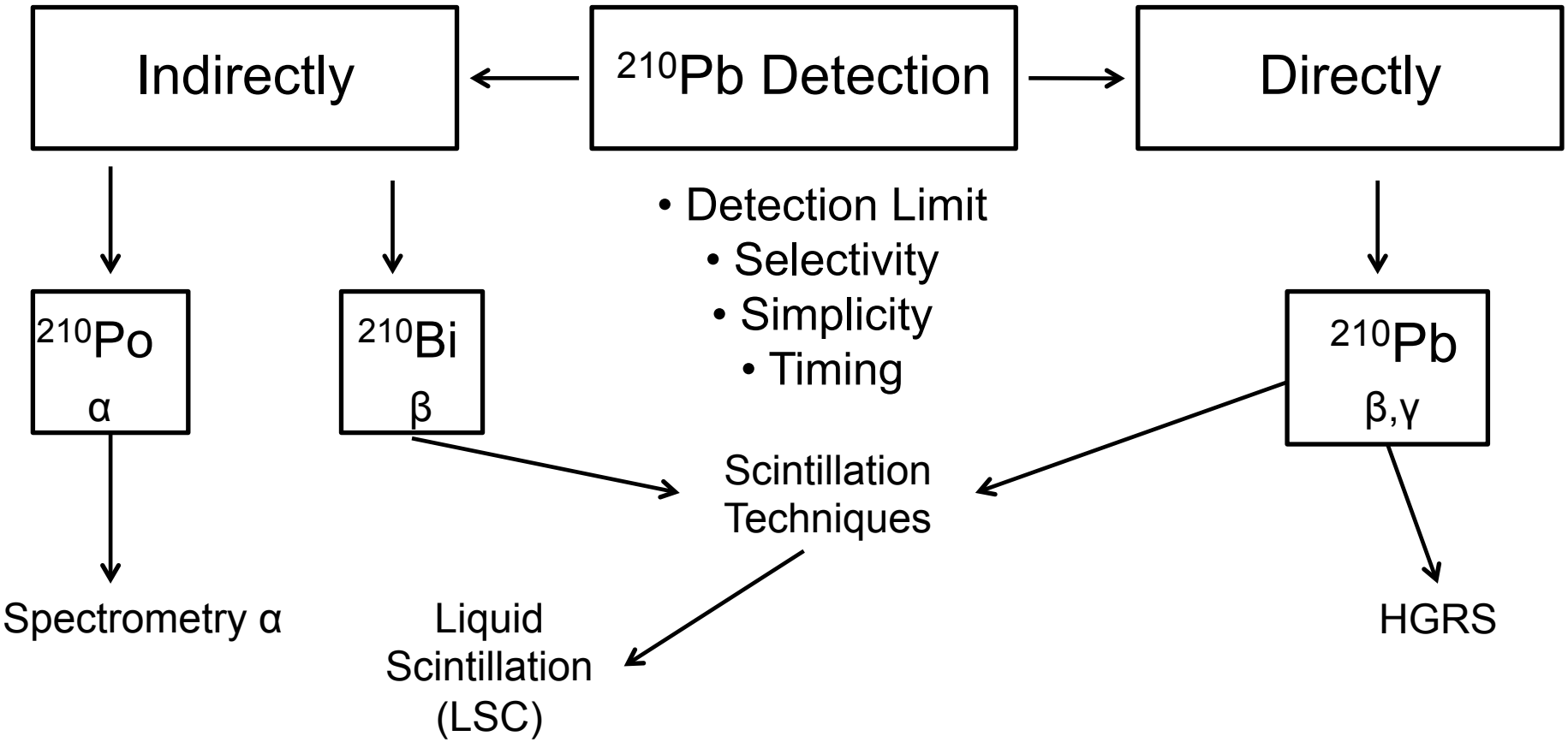
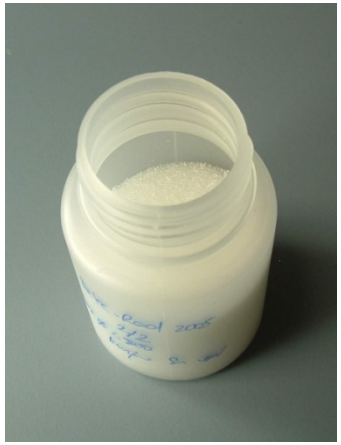
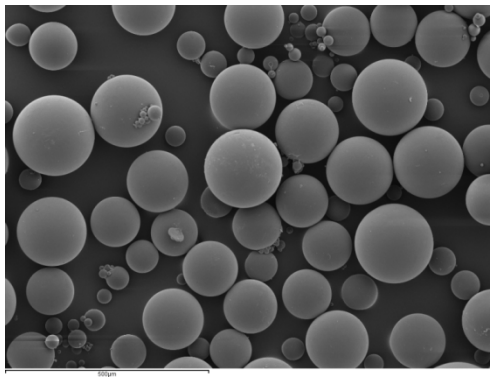


Need for reducing operations and time without loosing selectivity and sensitivity

Plastic Scintillation microspheres (PSm) and Plastic Scintillation resins (PSresin)



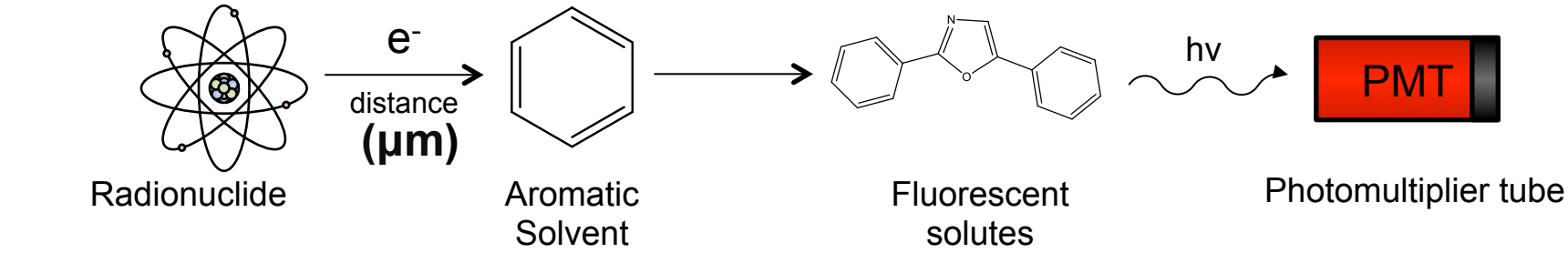
- Solid solution of fluorescence solutes (PPO, POPOP, pT, Bis-MSB,…) in a polymeric solvent (polyvinyltoluene or polystyrene) and other additives.
- Diameter: from tent to hundreds of micrometers

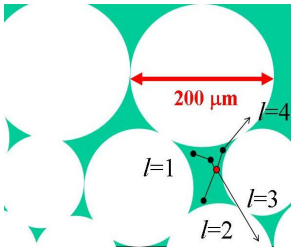


POLYMER

SOLID

Plastic Scintillation microspheres (PSm)





Particle Quenching

Ionization Quenching

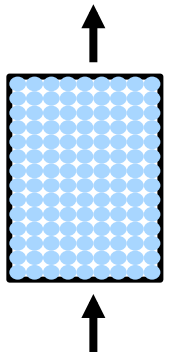
Chemical Quenching

Color Quenching

Optical Quenching

- Sample preparation similar to LS
- Low efficiency for low energy beta emitters
- Similar efficiency for high energy beta emitters

Uses and advantages of PSm



- No production of mixed waste.
- Reusability.
- Sample recovery after measurement.
- Salty samples can be used avoiding phase separation.
- Capability for real time and continuous monitoring.
- Absorption and measurement of ^{222}Rn

•Immobilization of selective extractans compounds onto the PSm surface.

PSresin

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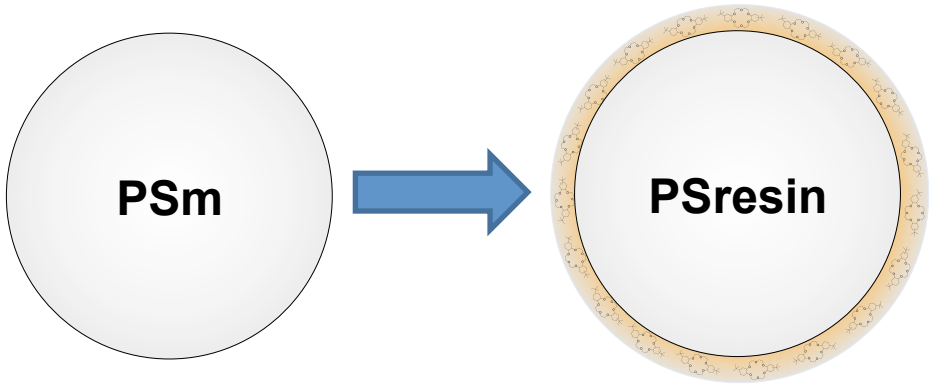
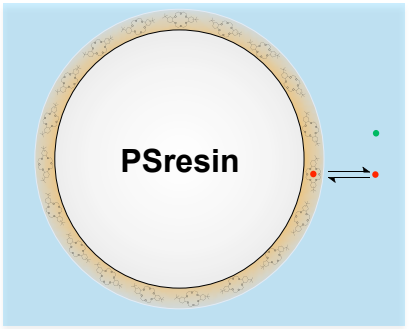
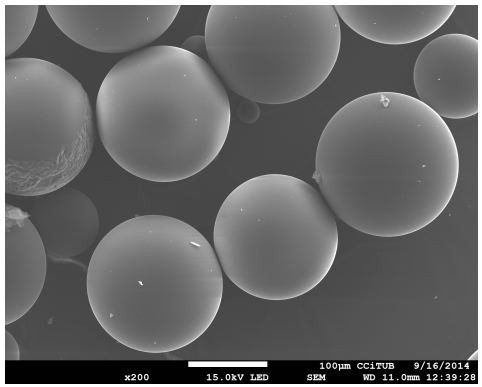
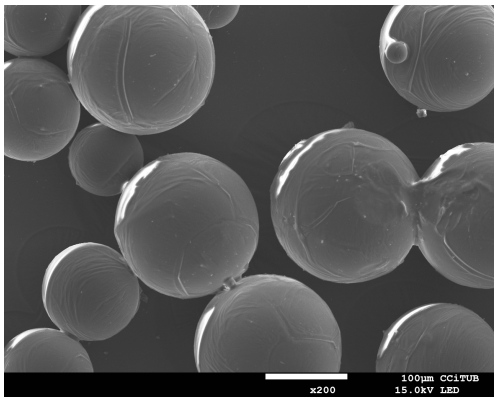
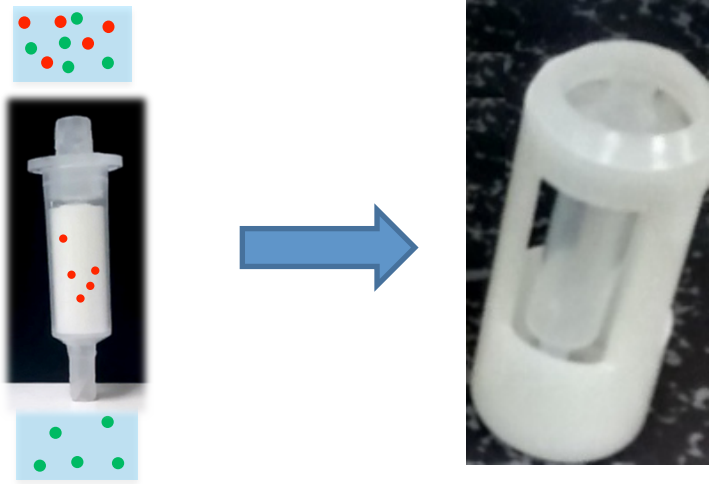
Plastic Scintillation resins (PSresin)











Solid Phase Extraction and Scintillation Detection

Analysis of ^{210}Pb in water samples with plastic scintillation resins.

Lluch, E.; Barreda, J.; **Tarancón, A.**; Bagán, H.; García, J .F.

Plastic Scintillation resins (PSresin)

- **$^{90}Sr^{2+}$:** 4,4'(5')-di-t butylcyclohexane 18-crown-6 in 1-octanol

Radiostrontium separation and measurement in a single step using plastic scintillators plus selective extractants. Application to aqueous sample analysis. Analytica Chimica Acta, 686, 1-2, 50-56. **2011.**

- **$S^{14}CN^-$:** Aliquat·336

Determination of oil reservoir radiotracer (S14CN$^-$) in a single step using a Plastic Scintillator extractive resin. Analytica Chimica Acta, 766, 30-35. **2012.**

- **$^{99}TcO^{4-}$:**
- **$^{126}Sn^{4+}$:**

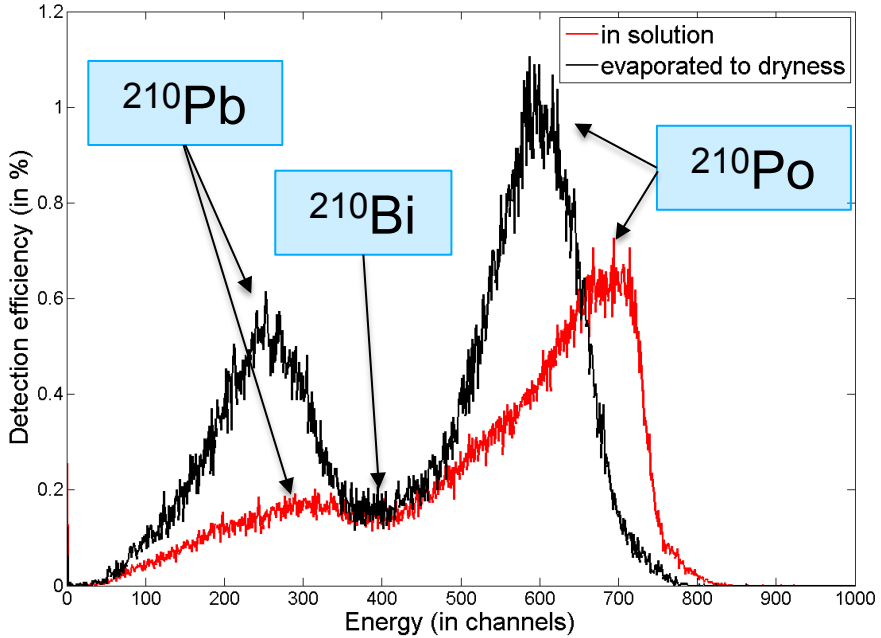
IN PROGRESS

OBJECTIVE

- Development of analysis method for ^{210}Pb using a PSresin.
  - Study of the measurement of ^{210}Pb/ ^{210}Bi/ ^{210}Po with PSm
  - Optimization of the coating of the PSresin.
  - Study of the separation medium.
  - Application to spiked river water samples.

Measurement of ^{210}Pb/ ^{210}Bi/ ^{210}Po with PSm

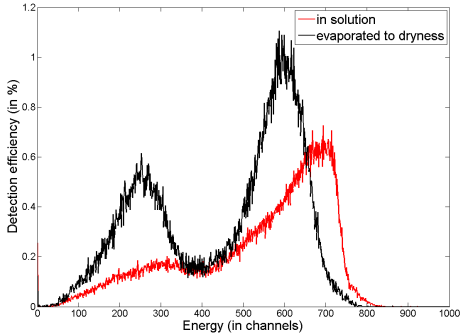
- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po
- Detector: Quantulus™
- Conditions:
  1. In solution
  2. Dryness Evaporation



- In solution: ^{210}Pb very small (High particle quenching for low-energy β)
- Evaporation to dryness: ^{210}Pb better defined (less particle quenching)

Measurement of ^{210}Pb/ ^{210}Bi/ ^{210}Po with PSm

- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po
- Detector: Quantulus™
- Conditions:
  1. In solution
  2. Dryness Evaporation

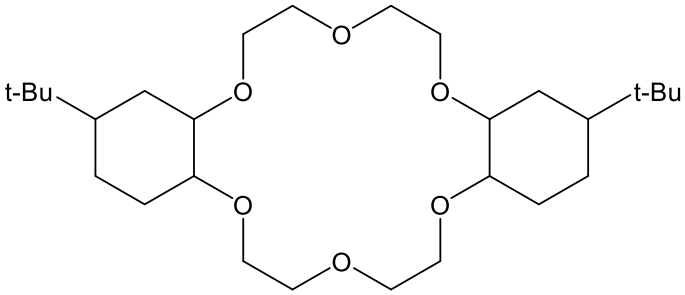


	Detection Efficiency
In solution	173(2) %
Dryness Evaporation	253(7) %

- In solution: Efficiency of ^{210}Pb very low (particle quenching).
- Evaporation to dryness: Efficiency of ^{210}Pb higher than 50%.
- **^{210}Pb can be measured with PSresin is solution is removed.**

Optimization of the coating of the Psresin.

- Extractant: 4',4''(5'')-di-tert-butildiciclohexà-18-corona-6 in 1-octanol



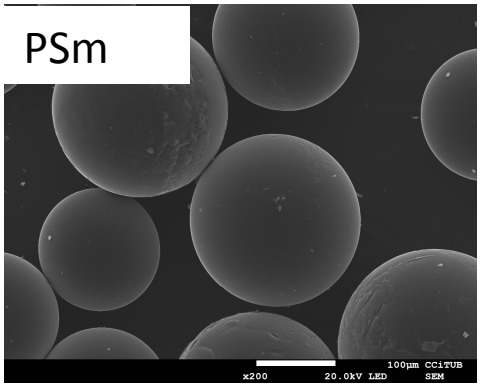
- Immobilization procedure: Impregnation through solvent evaporation at reduced pressure.
- Reference proportion: 1:1 (10 g of PSm for 6.26 g of Extractant)
- Proportions evaluated: 1:1/25, 1:1/12,5, 1:1/8, 1:1/4 and 1:3/4

Evaluation of PSresin

1. Morphology (Secondary Electron Microscopy Images)
2. Study of the retention of ^{210}Pb
3. Study of the elution of ^{210}Bi and ^{210}Po.

PSresin optimization. Morphology (SEM)

PSm



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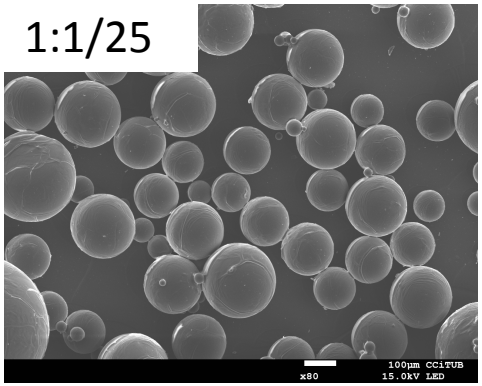

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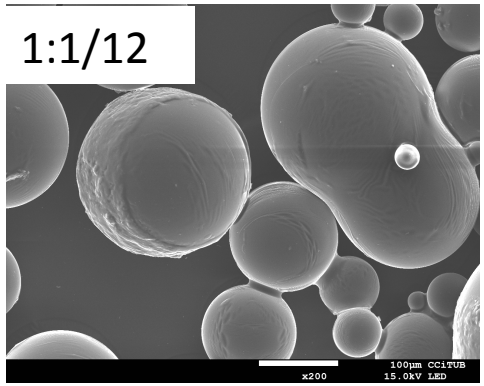

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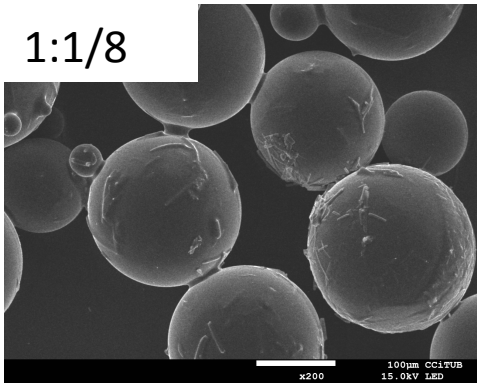
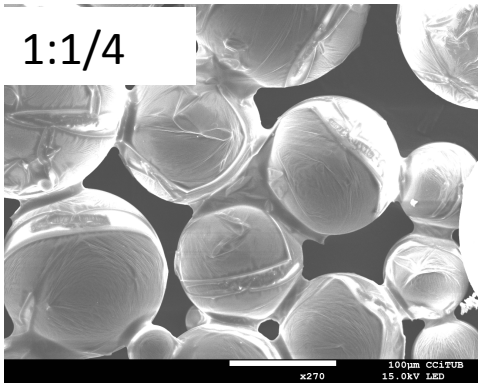
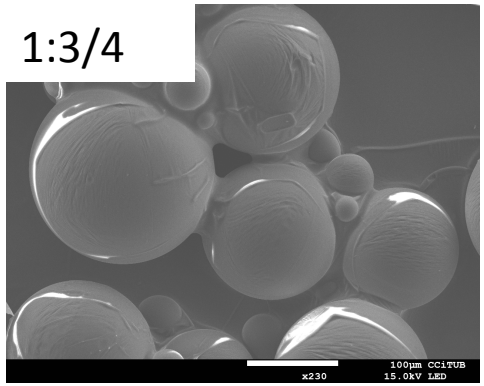


1:1/4



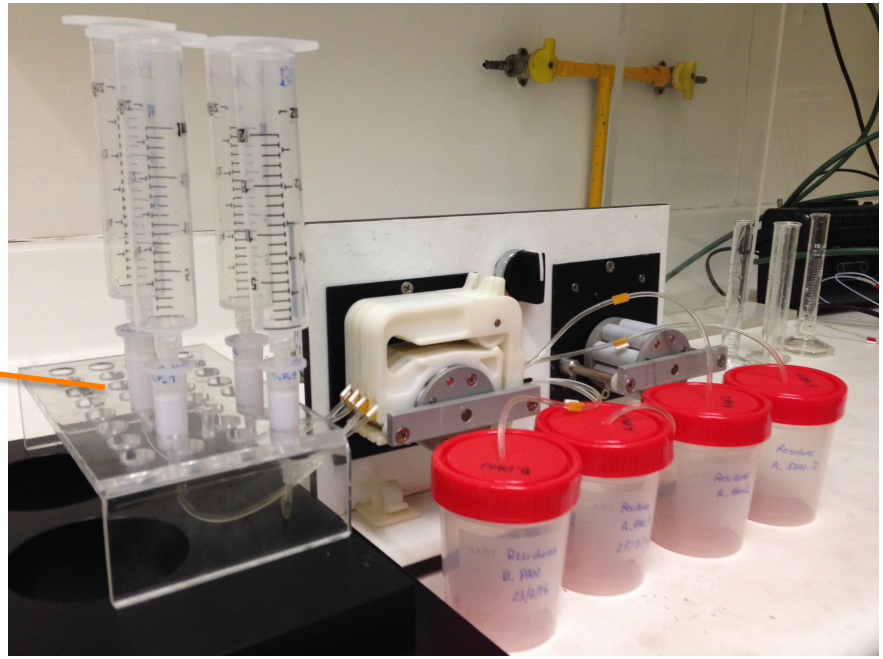
1:3/4



The amount of extractant increases when increasing the proportion

PSresin optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po in 0.17M HNO_3
- Column: 1 g of Psresin in a SPE cartridge
- Separation conditions: → Peristaltic pump. Flow: 0.5 mL/min
  1. Conditioning : 5 mL of 0.17M HNO_3.
  2. Sample loading: 2*5 mL of sample
  3. Cleaning: 4*5 mL of 0.17M HNO_3.
- Detector: Quantulus™



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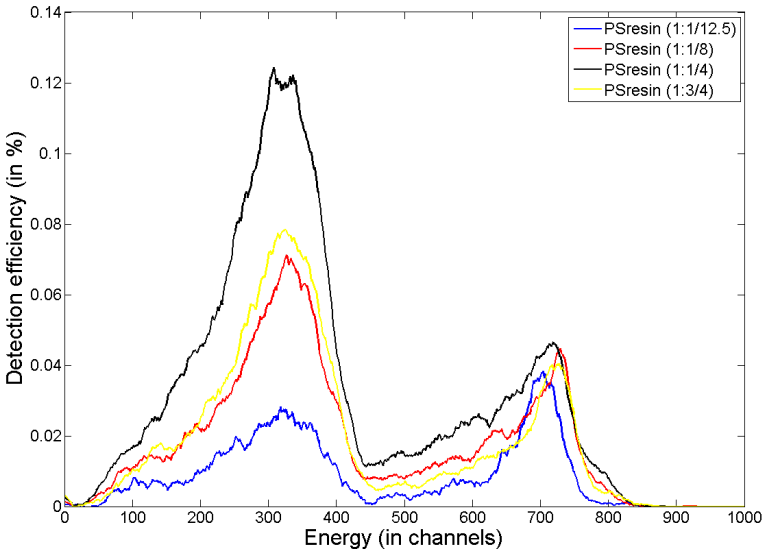
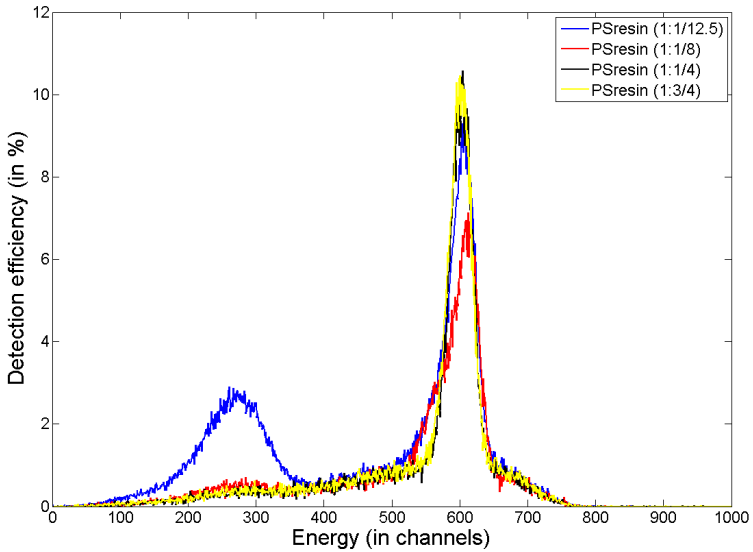
PSresin optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

SPE cartridge with PSresin



Elution from sample loading



- Higher retention of ^{210}Pb in the 1:1/4 proportion.
- ^{210}Bi (and ^{210}Po) are retained at this medium.
- ^{210}Pb is not eluted in the sample loading process

Analysis of ^{210}Pb in water samples with plastic scintillation resins.
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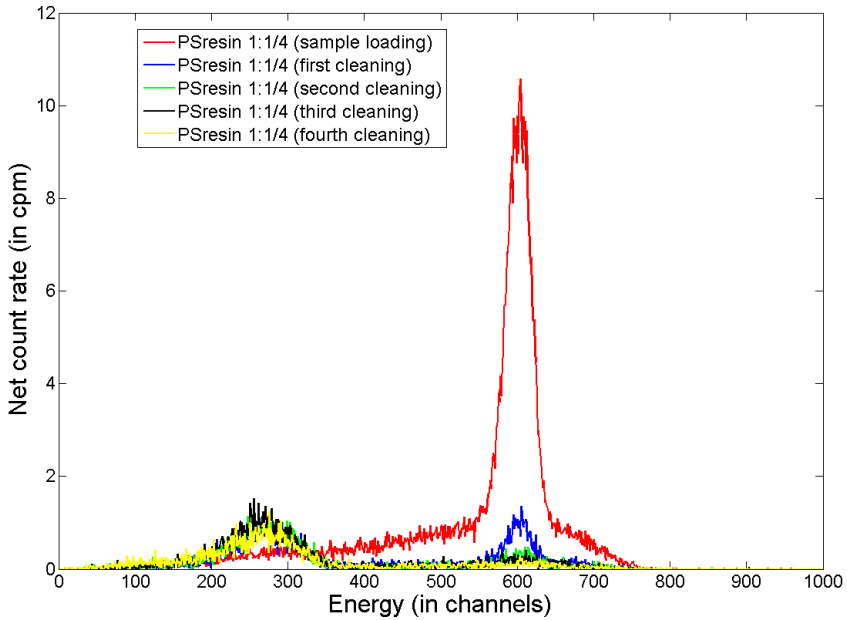
PSresin optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

Elution fractions from the 1:1/4 proportion

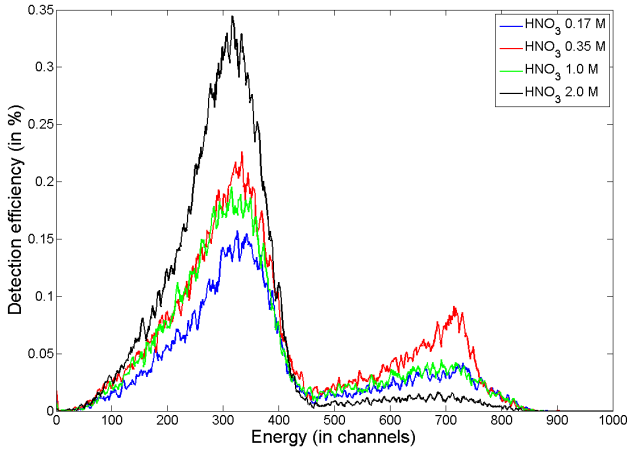


- ^{210}Bi and ^{210}Po are eluted in the loading process.
- ^{210}Pb is slightly eluted in the cleaning process.

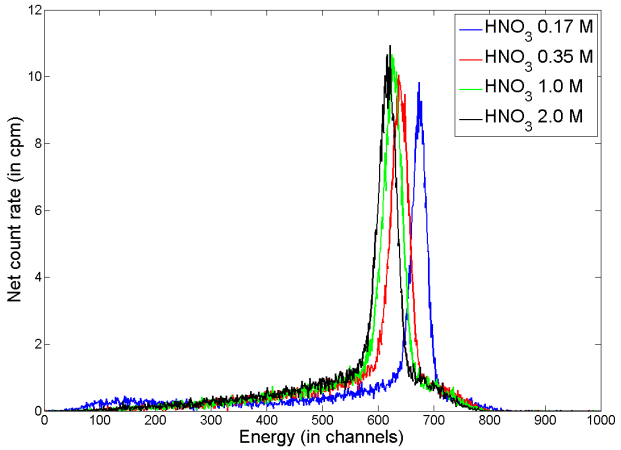
Medium optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

- **Mediums evaluated: HNO_3 at 0.04M, 0.17M, 0.35M, 1.0M and 2.0M.**
- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po
- Column, Separation conditions and Detection: As previous experiments.

SPE cartridge with PSresin



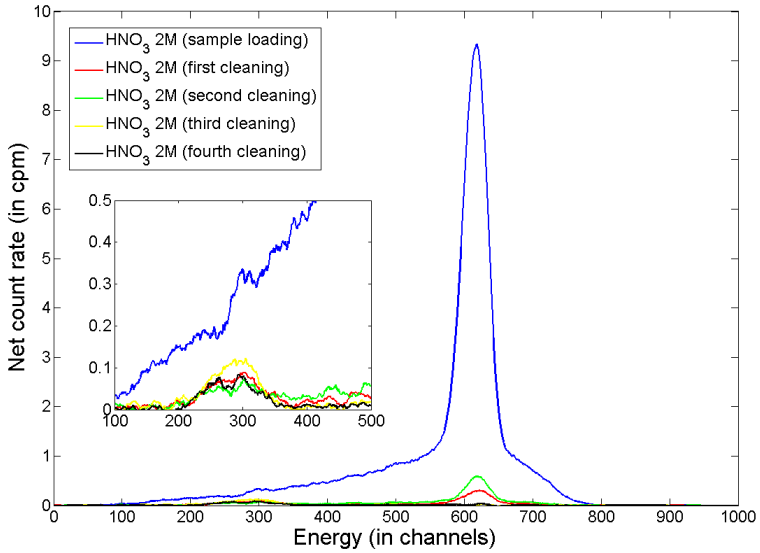
Elution from sample loading



- Maximum retention of ^{210}Pb observed at 2.0 M
- Minimum retention of the ^{210}Bi/^{210}Po at HNO_3 2.0 M

Medium optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

- **Mediums evaluated: HNO_3 at 0.04M, 0.17M, 0.35M, 1.0M and 2.0M.**



- Elution of ^{210}Pb at HNO_3 2.0 M is very small.

^{210}Pb analysis in spiked river waters.

- Sample: River water spiked with 200 Bq/L of ^{210}Pb, 80 mg/L of $Pb(NO_3)_2$ and 2 M HNO_3.
- Column: 1 g of Psresin in a SPE cartridge
- Separation conditions:
  1. Conditioning : 5 mL of 2 M HNO_3.
  2. Sample loading: 2*5 mL of sample
  3. Cleaning: 4*5 mL of 2 M HNO_3 with 80 mg/L of $Pb(NO_3)_2$.
- Detector: Quantulus™
- Detector: ICP-OES

$$\frac{Net\ Count\ Rate}{Activity} = Total_{efficiency} = Recovery_{efficiency} \cdot Counting_{efficiency}$$

ICP-OES

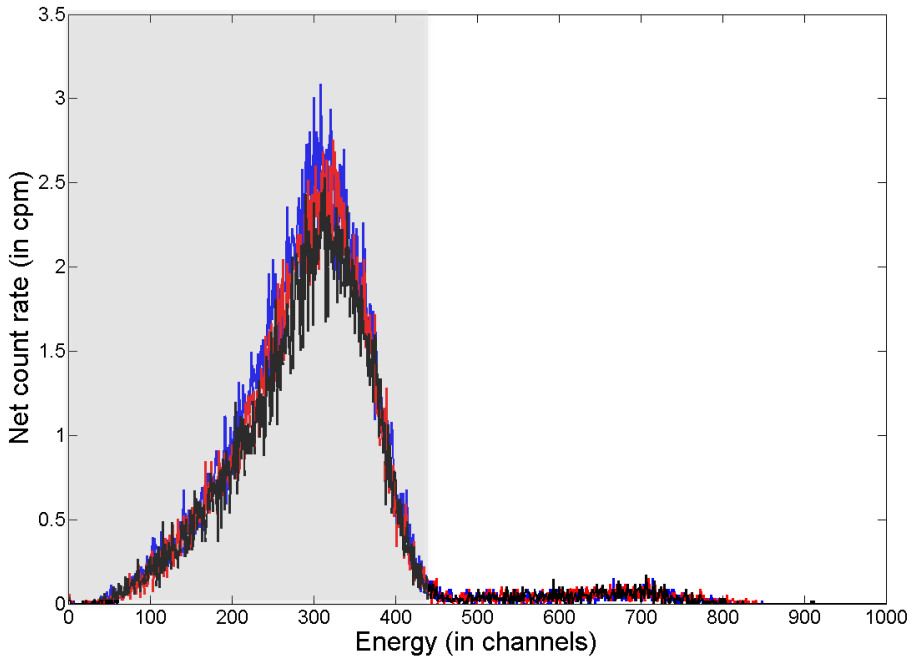
Calibration with Standards

^{210}Pb Calibration.

- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po, 80 mg/L of $Pb(NO_3)_2$ and 2 M HNO_3.
- Detector: Quantulus™
- Detector: ICP-OES

Counting Window

1-430



Background (cpm)	1.05
SQP(E)	800

^{210}Pb Calibration.

- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po, 80 mg/L of $Pb(NO_3)_2$ and 2 M HNO_3.
- N=3
- Detector: LS counter (Quantulus™)
- Detector: ICP-OES

	Mean (std dev.)	RSD (%)
Total Efficiency (%)	40 (3) %	7
Recovery (%)	91 (3) %	4

↓

	Mean (std dev.)	RSD (%)
Counting Efficiency (%)	44 (3) %	8

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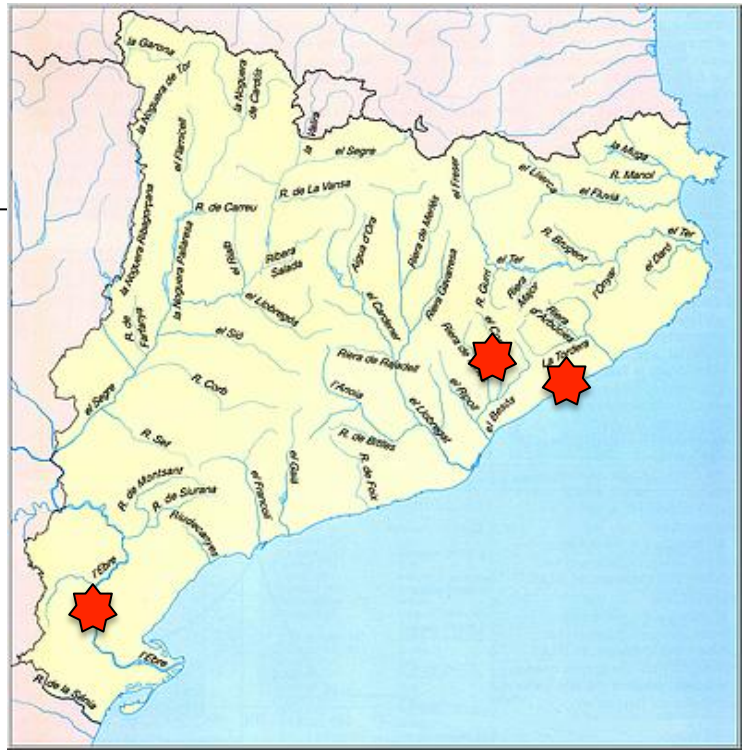
^{210}Pb analysis in spiked river water



- Samples:
  - River water: Ebro (Flix)
  - Subterranean water: Llavaneras
  - River water: Congost (Granollers)

- Spike:
  - Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po
  - 80 mg/L of $Pb(NO_3)_2$
  - 2 M HNO_3.

- Separation conditions:
  1. Conditioning : 5 mL of 2 M HNO_3.
  2. Sample loading: 10 mL of sample
  3. Cleaning: 20 mL (2M HNO_3/80 mg/L $Pb(NO_3)_2$)

^{210}Pb analysis in spiked river water

- Samples:
  - River water: Ebro (Flix)
  - Subterranean water: Llavaneras
  - River water: Congost (Granollers)

Recovery (in%)	
Sample	Mean (std dev.)
Calibration (n=3)	91 (3)
Ribarroja (n=2)	89 (1)
Llavaneras (n=6)	90 (3)
Congost (n=4)	89 (4)

- Recovery of 90%
- Good reproducibility

^{210}Pb analysis in spiked river water

Activity Quantification			
Sample	Activity Spiked (dpm/mL)	Acivity Measured (dpm/mL)	Deviation (%)
Ribarroja	10,1	10,8	-7,0
Ribarroja	10,1	11,0	-9,2
Ribarroja	10,1	9,8	3,6
Llavaneras	10,9	11,7	-7,0
Llavaneras	11,4	11,4	0,2
Llavaneras	11,4	11,8	-4,1
Congost	11,0	11,4	-4,2
Congost	10,4	10,7	-3,3
Congost	11,4	11,3	0,7

Errors lower than 10%

Conclusions

- SPE cartridges can be used as a support for separation and direct measurement in scintillation counter.
- We have developed a PSresin selective for ^{210}Pb with regards its daughter radionuclides (^{210}Bi and ^{210}Po) in a HNO_3 2M.
- Detection efficiency is around 40% in the ^{210}Pb window (1-430)
- Recovery of Pb^{2+} is around 90 % and reproducible (RSD <5%)
- ^{210}Pb activity determination in river water is performed with errors lower that 10%.

Acknowledgement

- Spanish *Ministerio de Economia y Competitividad* (MINECO) for financial support under CTM2014-02020

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Thank you for your attention

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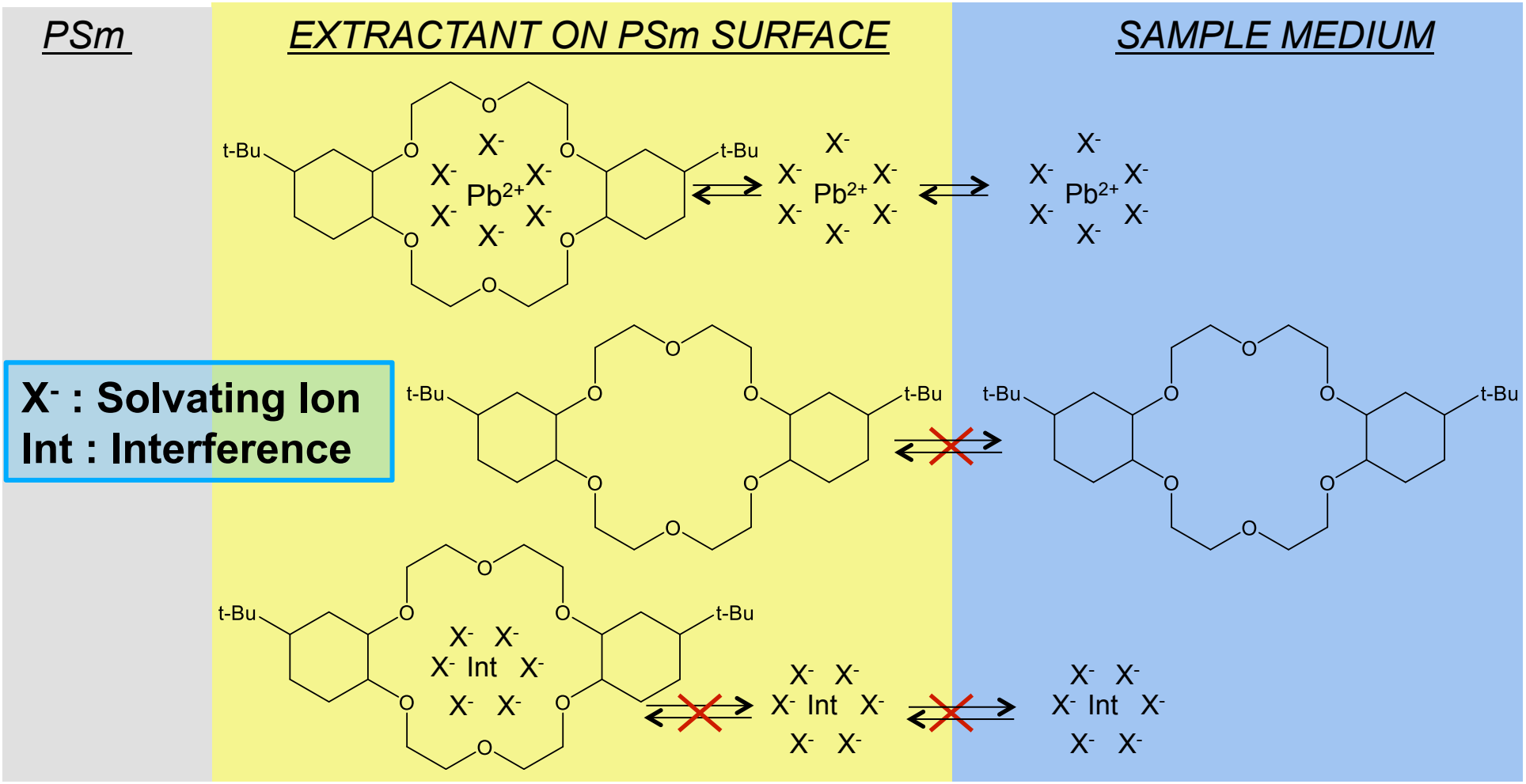
[1]Department of Analytical Chemistry, University of Barcelona, Martí i Franqués, 1-11, E-08028, Barcelona, Spain
[2] Department of Pure and Applied Biochemistry, Lund University, Getingevägen 60, hus II, 22100 SE, Lund, Sweden

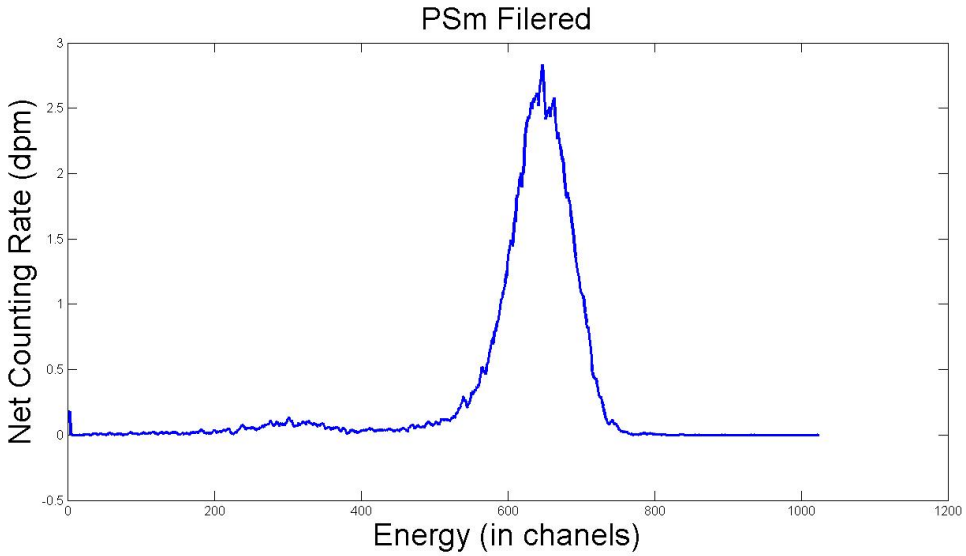
Selective Extraction of ^{210}Pb

Selectivity reported to the Crown-ether: 4',4''(5'')-di-tert-butildiciclohexà-18-corona-6



Measurement of ^{210}Pb/ ^{210}Bi/ ^{210}Po with cleaned PSm

- Sample: Standard Solution of ^{210}Pb/^{210}Bi/^{210}Po
- Detector: Quantulus™
- Conditions: Cleaned and filtered with water



	Detection Efficiency
filtered	36 (2) %

- ^{210}Po is signficantly retained in the PSm when are treated with water.

Medium optimization. ^{210}Pb retention and ^{210}Bi/^{210}Po elution

- Mediums evaluated: HNO_3 at 0.04M, 0.17M, 0.35M, 1.0M and 2.0M.
- **Batch Experiments: 0.15 g of PSresins, 0.45 mL of a 0.8 g/L $Pb(NO_3)_2$ 3.15 mL of HNO_3 medium in 6 mL vials. 24 Hours of agitation**
- Measurement: ICP-OES

HNO_3 Concentration	Pb^{2+} retention efficiency
0.04 M	79 (5) %
0.17 M	91 (5) %
0. 35 M	96 (6) %
1.0 M	98 (5) %
2.0 M	98 (5) %

- Retention of ^{210}Pb increases with the HNO_3 concentration.
- Retention of ^{210}Pb at HNO_3 2.0M is quantitative.

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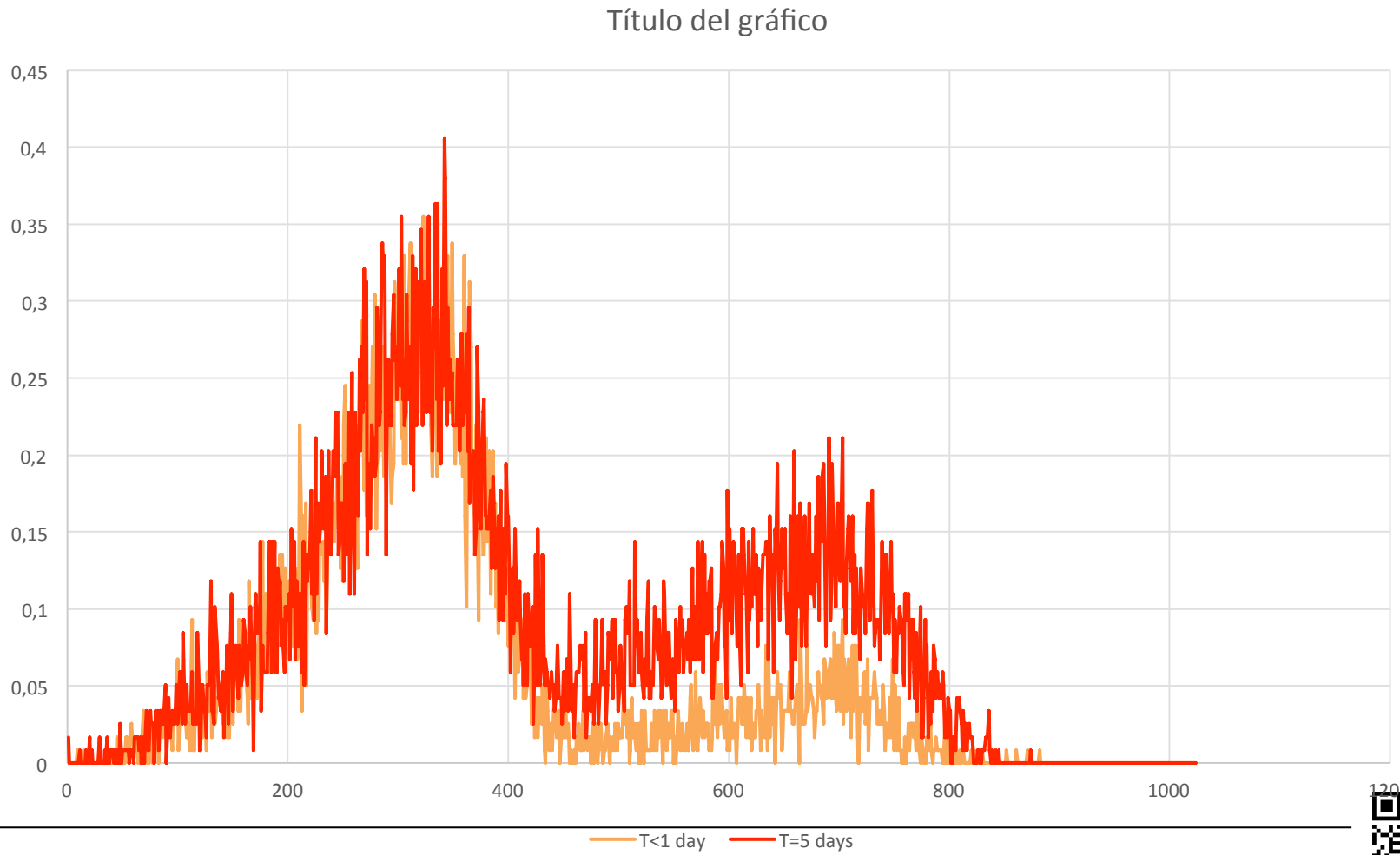
^{210}Pb analysis in spiked river waters.

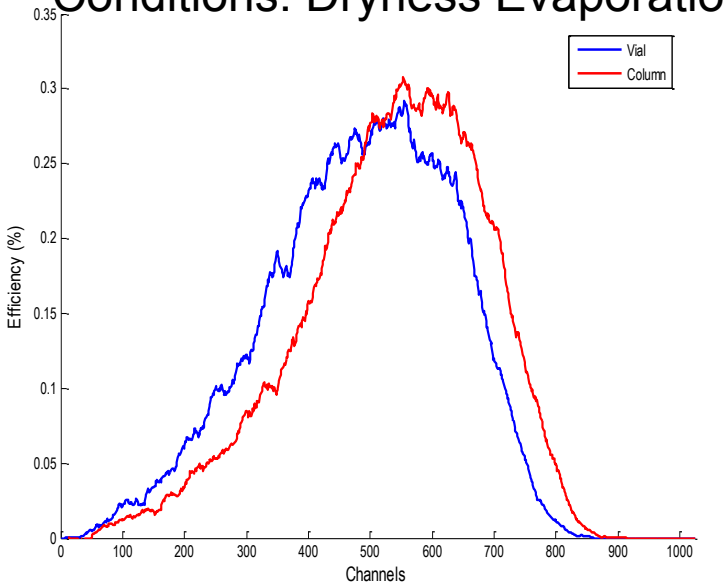


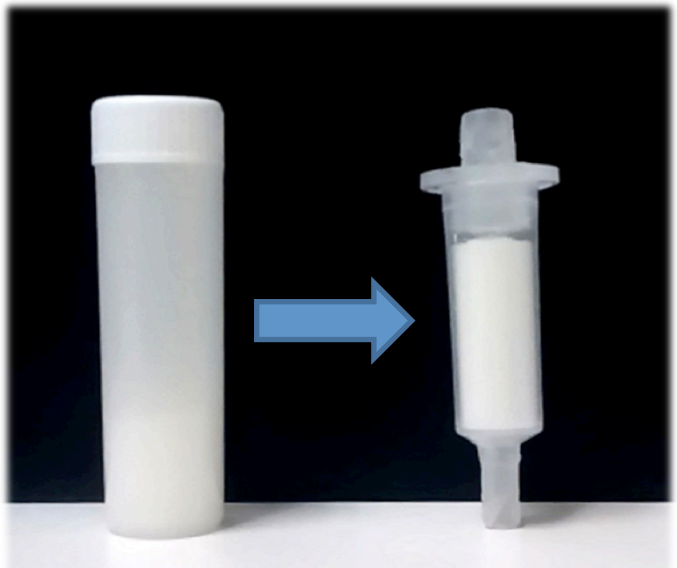
Analysis of ^{210}Pb in water samples with plastic scintillation resins.

Lluch, E.; Barreda, J.; **Tarancón, A.**; Bagán, H.; García, J .F.

Measurement with SPE cartridges

- Sample: Standard Solution of $^{90}Sr/^{90}Y$
- Detector: Quantulus™
- Conditions: Dryness Evaporation





	$^{90}Sr/^{90}Y$ Detection Efficiency
Vial	109 (2) %
Column	109 (7) %

Measurement with SPE is comparable to that done with classical vials.