

# **Rapid and Simultaneous Determination of Np and Pu in Environmental Samples Using Sequential Injection Anion Exchange Chromatography and ICP-MS**

**PhD student: Jixin Qiao**

**Supervisor: Xiaolin Hou**

**Co-supervisor: Per Roos, Manuel Miró**

***Risø-DTU, Technical University of Denmark***

## BACKGROUND

**Plutonium isotopes ( $^{238,239,240,241}\text{Pu}$ ) and Neptunium ( $^{237}\text{Np}$ ) are highly hazardous radioactive pollutants in the environment due to:**

- 1) long radioactive half-lives;
- 2) high radiological toxicities;
- 3) long-term persistence in environment.

**Table 1. Nuclear Properties of Important Plutonium Isotopes**

<b>Isotope</b>	<b>Half-life</b>	<b>Specific activity (Bg/g)</b>	<b>Principal decay mode</b>	<b>Decay energy (MeV)</b>
$^{238}\text{Pu}$	<b>87.7yr</b>	$6.338 \times 10^{11}$	$\alpha$	$\alpha$ <b>5.499 (70.9%)</b>
$^{239}\text{Pu}$	<b><math>2.411 \times 10^4\text{yr}</math></b>	$2.296 \times 10^9$	$\alpha$	$\alpha$ <b>5.157 (70.77%)</b>
$^{240}\text{Pu}$	<b><math>6.561 \times 10^3\text{yr}</math></b>	$8.401 \times 10^9$	$\alpha$	$\alpha$ <b>5.168 (72.8%)</b>
$^{241}\text{Pu}$	<b>14.35yr</b>	$3.825 \times 10^{12}$	$\beta$ <b>-&gt;99.99%</b>	$\alpha$ <b>4.896 (83.2%)</b>
$^{237}\text{Np}$	<b><math>2.411 \times 10^6\text{yr}</math></b>	$2.603 \times 10^7$	$\alpha$	$\alpha$ <b>4.788 (51%)</b>

## BACKGROUND

**The determination of plutonium isotopes and Neptunium in the environment is important for:**

- 1) environmental risk assessment and monitoring of sites around nuclear facilities;
- 2) emergency preparedness;
- 3) surveys for the contaminated area resulting from nuclear weapon tests, nuclear accidents, and the discharge of nuclear waste.

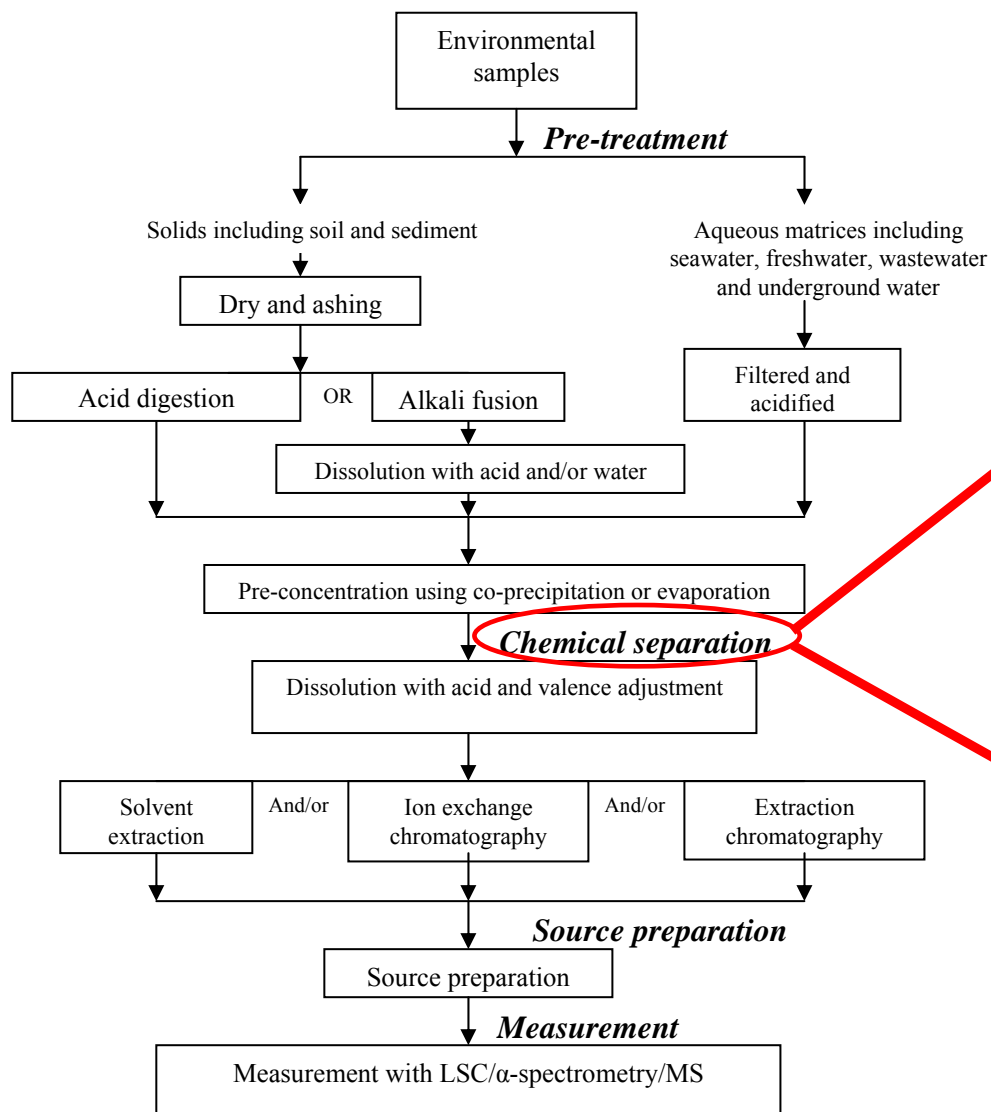
## BACKGROUND

- 1) The **levels of plutonium** isotopes and **neptunium** in the environment are very **low** and depending of the location.
- 2) **Plutonium** and **neptunium** often coexist with **matrix elements** (Ca, Mg, Al, V...) and **other radionuclides** (Th, U, Am, Cm...).

Table 2. Environmental level of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$ 

Sample	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$
Soil, Bq/kg	0.07	0.1-7
Herbaceous plants ,Bq/kg	$4.5 \times 10^{-4}$	0.3-2
Lichen, Bq/kg	-	4-10
Grain, vegetables, Bq/kg	$(0.2-14) \times 10^{-4}$	$(4-89) \times 10^{-4}$
Lake water ,Bq/L	-	$(0.1-29) \times 10^{-6}$
Sea water, Bq/L	-	$(0.7-52) \times 10^{-6}$

# BACKGROUND



- Advantages**
- ♣ sensitive;
  - ♣ precise;
  - ♣ accurate.

- Disadvantages**
- ♣ time-consuming;
  - ♣ labour intensive;
  - ♣ generate hazardous liquid and solid waste.

Fig. 1 Analytical procedure for the determination of Pu and Np in environmental samples

## OBJECTIVE

### Objective:

To develop a **new** analytical method for determination of plutonium isotopes and neptunium in environmental samples.

**Main Points:** 1)Automatic  
2)Rapid  
3)Simultaneous

## MAIN CHALLENGES

- **Small column size**
- **Same behavior of Pu and Np on the column**
- **Valence adjustment**
- **High chemical yields**
- **Good decontamination factors (U, Th, Pb)**

STRATEGY

Ion-exchange  
Chromatography



Sequential  
Injection (SI)

ICP-MS

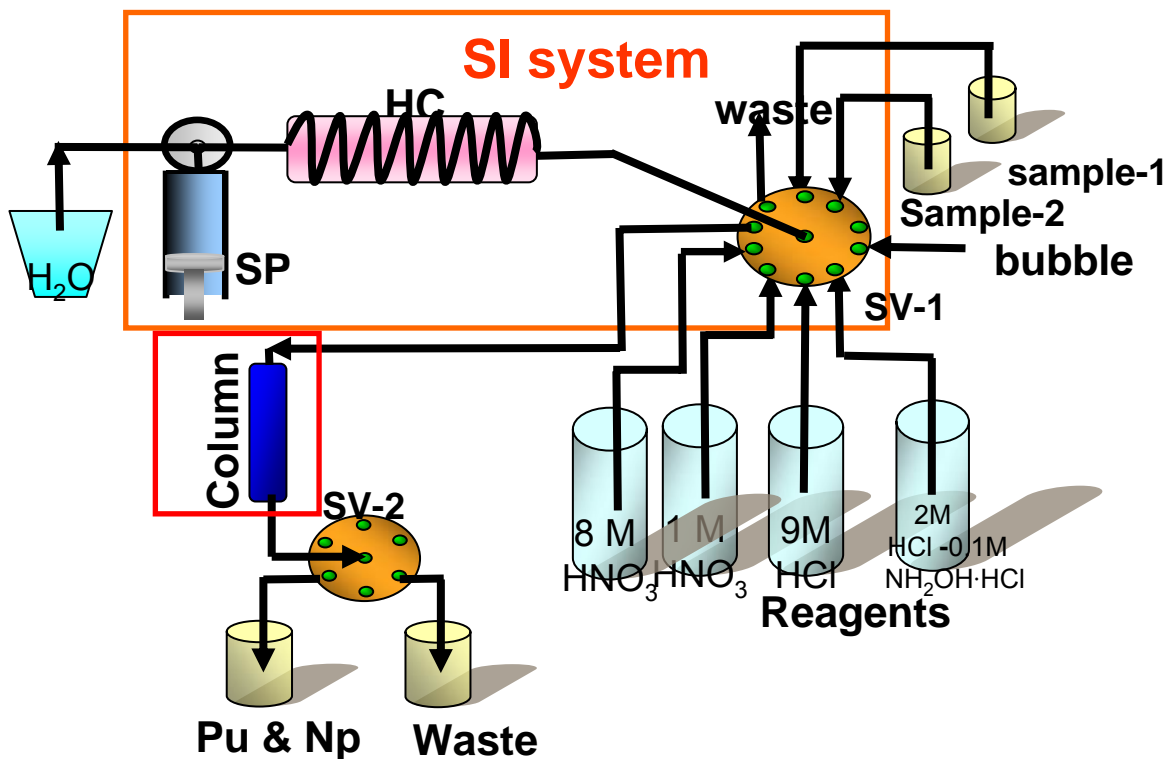


Fig. 2 Scheme of a SI system



## Samples

**Soil**: Danish soil, reference material from a laboratory round-robin intercomparison. The reference values of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are  $0.140 \pm 0.008$  and  $0.098 \pm 0.006$  Bg/kg.

**Sediment, plants, seawater...**

## Anion exchange chromatographic column

**Column size: 16mL (1.0 x 20 cm)**

**8mL (0.7 x 20 cm)**

**4mL (0.7 x 10 cm)**

**2mL (0.5 x 10 cm)**

**2mL (0.7 x 5.0 cm)**

**Resin: AG 1x2 (50-100mesh), AG 1x4 (50-100mesh),  
AG 1x4(100-200mesh), AG 1x8(50-100mesh).**

## Instrumentation

### 1) FIA lab system 3500

- ♣ Syringe/peristaltic pump
- ♣ 10-port selection valve

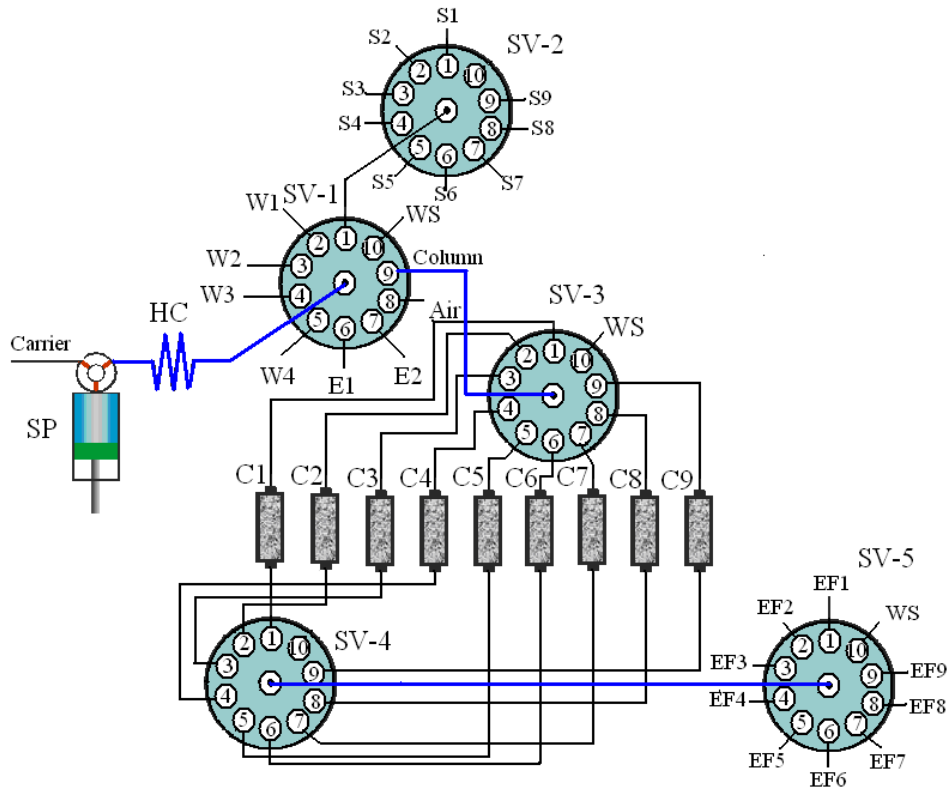
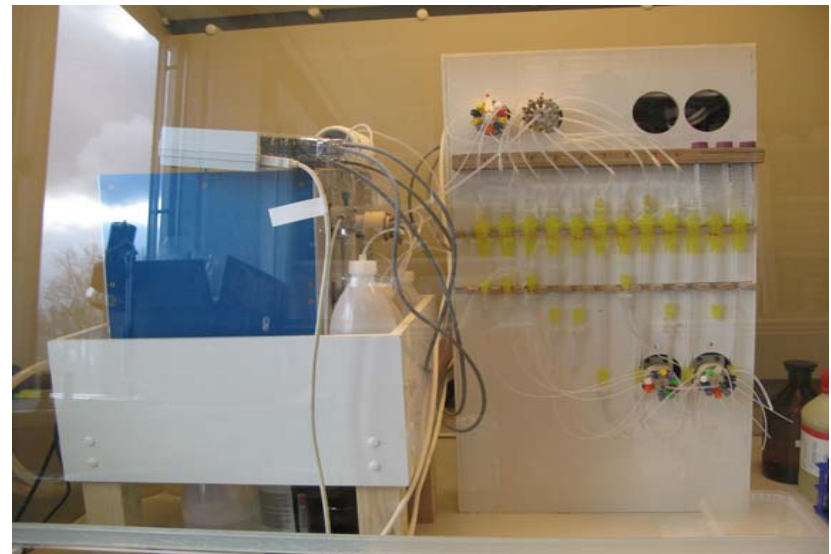
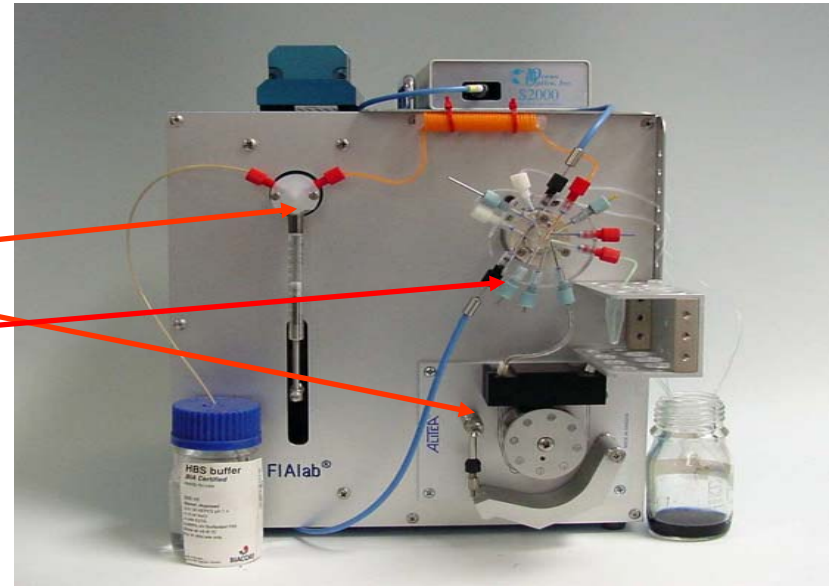


Fig.3. Scheme of the experimental setup



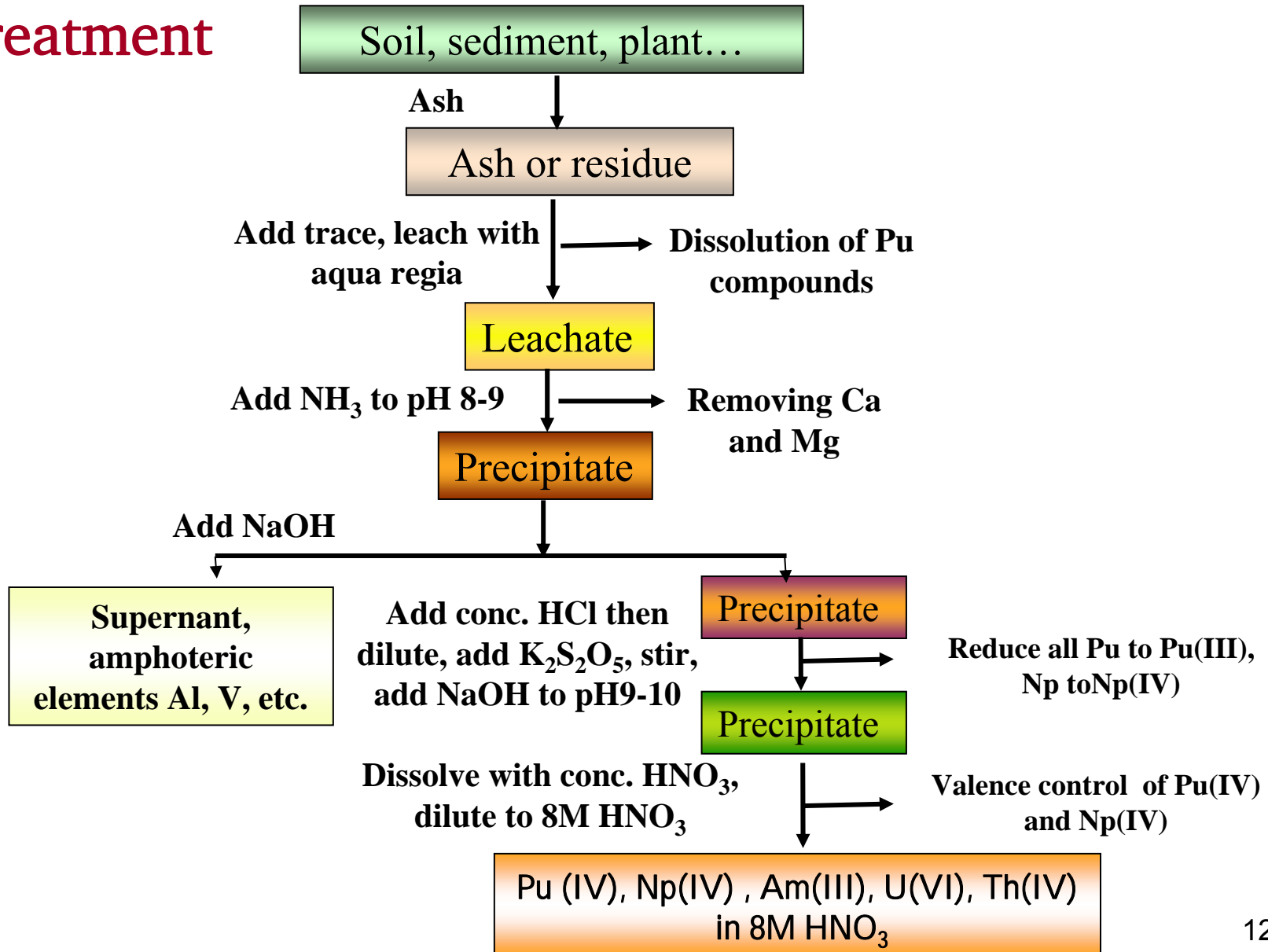
## Instrumentation

### 2) ICP-MS

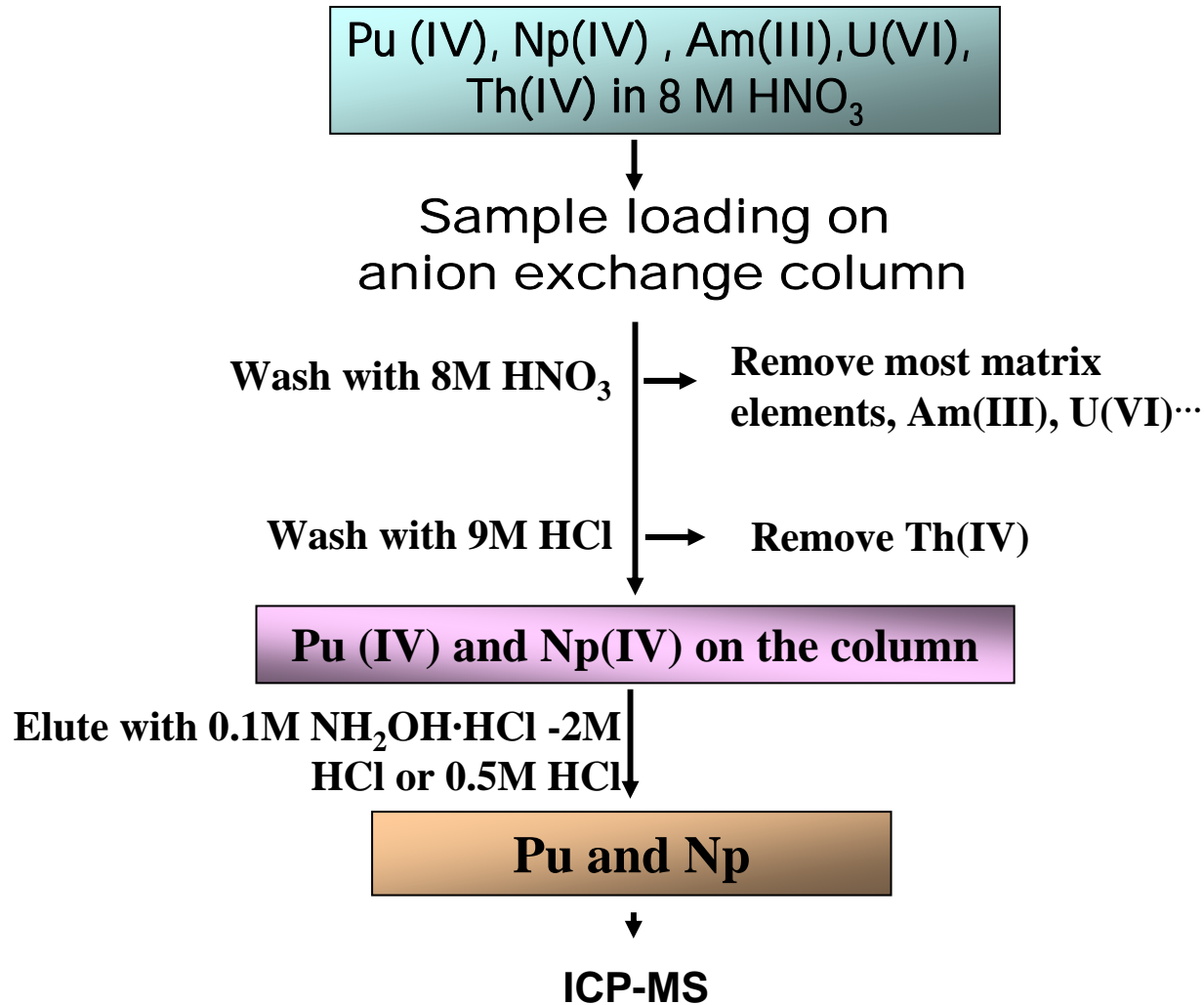
**Thermo X-series inductively coupled plasma mass spectrometry (ICP-MS)**



## Pre-treatment



## Separation & detection



## Experimental parameters for comparison

- **Column size**
- **Resin type**
- **Washing solution (1.0-8.0 mol/L HNO<sub>3</sub>)**
- **Elution solution (NH<sub>2</sub>OHHCl-HCl, 0.1-1.0mol/L HCl)**
- **Flow rate (1.0-5.0 mL/min)**

## Key factors for evaluation of experimental results

- Chemical yields of Pu and Np

😊 >85%   😞 <85%

- Ratio between the chemical yield of  $^{237}\text{Np}$  and  $^{242}\text{Pu}$

😊 0.9-1.1   😞 otherwise

- Measured values of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$

😊 agree well with the reference values

😞 otherwise

- Decontamination factors for U, Th and Pb

😊  $>10^3$    😞  $<10^3$

**Table 3. comparison of different experimental conditions for the separation of Pu and Np (1)**

Column size	Resin	Separation condition#	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
16mL (1.0 × 20cm)	AG1 × 2	W-1, 2.5 E-1, 1.0	102.3 ± 5.1	95.8 ± 4.8	0.9	0.14 ± 0.0	0.10 ± 0.01	3.3 × 10 <sup>3</sup>	1.9 × 10 <sup>4</sup>	1.4 × 10 <sup>4</sup>
	AG1 × 4		99.9 ± 5.0	94.8 ± 4.7	0.9	0.23 ± 0.02	0.08 ± 0.01	3.0 × 10 <sup>2</sup>	2.9 × 10 <sup>3</sup>	5.6 × 10 <sup>3</sup>
	AG1 × 8		96.4 ± 4.8	90.9 ± 4.5	0.9	1.39 ± 0.14	0.11 ± 0.01	4.2 × 10 <sup>1</sup>	3.8 × 10 <sup>2</sup>	1.2 × 10 <sup>4</sup>
8mL (0.7 × 20cm)	AG1 × 2	W-1, 2.5 E- 2, 1.0	71.5 ± 3.6	67.4 ± 3.4	0.9	0.17 ± 0.02	0.12 ± 0.01	1.1 × 10 <sup>3</sup>	5.2 × 10 <sup>3</sup>	3.1 × 10 <sup>4</sup>
	AG1 × 4		100.0 ± 5.4	100.0 ± 5.3	1.0	0.16 ± 0.02	0.10 ± 0.01	1.6 × 10 <sup>3</sup>	6.4 × 10 <sup>3</sup>	3.9 × 10 <sup>3</sup>
	AG1 × 8		94.2 ± 4.7	87.9 ± 4.4	0.9	0.16 ± 0.02	0.10 ± 0.01	3.2 × 10 <sup>2</sup>	7.8 × 10 <sup>2</sup>	8.1 × 10 <sup>3</sup>
	AG1 × 8	W-1, 5.0 E-2, 2.5	91.9 ± 4.6	80.1 ± 4.0	0.9	0.18 ± 0.02	0.12 ± 0.01	1.1 × 10 <sup>2</sup>	9.1 × 10 <sup>1</sup>	8.8 × 10 <sup>3</sup>
4mL (0.7 × 10cm)	AG1 × 2	W-1, 2.5 E-2, 1.0	71.2 ± 3.6	48.4 ± 2.4	0.7	0.12 ± 0.01	0.06 ± 0.01	2.1 × 10 <sup>3</sup>	4.4 × 10 <sup>3</sup>	1.1 × 10 <sup>4</sup>
	AG1 × 4		100.0 ± 5.0	98.2 ± 4.9	1.0	0.12 ± 0.01	0.10 ± 0.01	1.3 × 10 <sup>3</sup>	2.4 × 10 <sup>3</sup>	2.2 × 10 <sup>4</sup>
	AG1 × 8		98.7 ± 4.9	97.2 ± 4.9	1.0	0.16 ± 0.02	0.10 ± 0.01	1.0 × 10 <sup>3</sup>	8.9 × 10 <sup>2</sup>	8.6 × 10 <sup>3</sup>
	AG1 × 8	W-1, 5.0 E-2, 2.5	92.6 ± 4.6	86.3 ± 4.3	0.9	0.17 ± 0.02	0.12 ± 0.01	2.2 × 10 <sup>2</sup>	9.8 × 10 <sup>1</sup>	2.7 × 10 <sup>3</sup>



**Table 3. comparison of different experimental conditions for the separation of Pu and Np (2)**

Column size	Resin	Separation condition#	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
2mL (0.5 × 10cm)	AG1 × 2	W-2, 2.5 E-2, 2.5	75.0 ± 3.8	19.0 ± 1.0	0.3	0.26 ± 0.03	0.08 ± 0.01	3.2 × 10 <sup>2</sup>	2.0 × 10 <sup>2</sup>	2.6 × 10 <sup>3</sup>
		W-2, 5.0 E-2, 2.5	48.6 ± 2.4	35.7 ± 1.8	0.7	0.15 ± 0.02	0.10 ± 0.01	8.9 × 10 <sup>2</sup>	6.8 × 10 <sup>2</sup>	9.3 × 10 <sup>3</sup>
	AG1 × 4	W-2, 2.5 E-2, 2.5	103.0 ± 5.2	106.0 ± 5.3	1.0	0.14 ± 0.01	0.09 ± 0.01	3.9 × 10 <sup>3</sup>	2.4 × 10 <sup>4</sup>	2.7 × 10 <sup>4</sup>
		W-2, 5.0 E-2, 2.5	94.0 ± 4.7	89.7 ± 4.5	1.0	0.25 ± 0.03	0.09 ± 0.01	3.9 × 10 <sup>2</sup>	6.7 × 10 <sup>3</sup>	1.6 × 10 <sup>4</sup>
	AG1 × 8	W-2, 2.5 E-2, 2.5	90.5 ± 5.0	88.7 ± 4.9	1.0	0.25 ± 0.03	0.09 ± 0.01	3.4 × 10 <sup>2</sup>	2.7 × 10 <sup>2</sup>	9.0 × 10 <sup>5</sup>
		W-2, 5.0 E-2, 2.5	100.5 ± 5.0	98.7 ± 4.9	1.0	0.29 ± 0.03	0.07 ± 0.01	4.8 × 10 <sup>1</sup>	2.1 × 10 <sup>2</sup>	2.9 × 10 <sup>5</sup>
	AG1 × 4	W-2, 2.5 E-4, 2.5	72.9 ± 3.6	64.1 ± 3.2	0.9	0.18 ± 0.02	0.12 ± 0.01	1.6 × 10 <sup>3</sup>	1.7 × 10 <sup>4</sup>	6.1 × 10 <sup>3</sup>
		W-3, 2.5 E-4, 2.5	81.8 ± 4.1	69.2 ± 4.1	0.8	0.38 ± 0.04	0.10 ± 0.01	2.1 × 10 <sup>3</sup>	1.3 × 10 <sup>4</sup>	7.5 × 10 <sup>3</sup>
		W-4, 2.5 E-4, 2.5	80.2 ± 4.0	63.8 ± 4.1	0.8	0.19 ± 0.02	0.09 ± 0.01	2.4 × 10 <sup>3</sup>	1.4 × 10 <sup>4</sup>	1.5 × 10 <sup>4</sup>
		W-5, 2.5 E-4, 2.5	39.6 ± 2.0	20.6 ± 4.1	0.5	0.20 ± 0.02	0.15 ± 0.02	2.6 × 10 <sup>3</sup>	1.1 × 10 <sup>4</sup>	9.4 × 10 <sup>3</sup>
		W-6, 2.5 E-4, 2.5	31.0 ± 1.6	11.7 ± 4.1	0.4	0.19 ± 0.02	0.20 ± 0.02	3.1 × 10 <sup>3</sup>	5.0 × 10 <sup>4</sup>	1.2 × 10 <sup>5</sup>

**Table 3. comparison of different experimental conditions for the separation of Pu and Np (3)**

Column size	Resin	Separation condition #	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
2mL (0.5 × 10cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 § E-3, 2.5	61.0 ± 3.1	64.9 ± 3.2	1.1	0.18 ± 0.02	0.12 ± 0.01	3.1 × 10 <sup>3</sup>	2.8 × 10 <sup>4</sup>	1.2 × 10 <sup>4</sup>
		W-2, 2.5 E-4, 2.5	91.6 ± 4.6	91.0 ± 4.6	1.0	0.14 ± 0.01	0.10 ± 0.01	6.9 × 10 <sup>3</sup>	1.7 × 10 <sup>4</sup>	1.0 × 10 <sup>3</sup>
		W-2, 2.5 E- 5, 2.5	66.8 ± 3.3	74.6 ± 3.7	1.1	0.14 ± 0.01	0.09 ± 0.01	8.6 × 10 <sup>3</sup>	1.2 × 10 <sup>4</sup>	1.0 × 10 <sup>3</sup>
		W-2, 2.5 E-6, 2.5	78.5 ± 3.9	81.6 ± 4.1	1.0	0.14 ± 0.01	0.07 ± 0.01	6.3 × 10 <sup>3</sup>	1.9 × 10 <sup>4</sup>	1.5 × 10 <sup>3</sup>
		W-3, 2.5 E-4, 2.5	35.9 ± 1.8	61.2 ± 3.1	1.7	0.22 ± 0.02	0.19 ± 0.02	3.4 × 10 <sup>3</sup>	3.1 × 10 <sup>4</sup>	2.9 × 10 <sup>3</sup>
		W-4, 2.5 E-4, 2.5	82.3 ± 4.1	80.9 ± 4.0	1.0	0.21 ± 0.02	0.12 ± 0.01	3.8 × 10 <sup>3</sup>	1.4 × 10 <sup>4</sup>	4.3 × 10 <sup>4</sup>
		W-5, 2.5 E-4, 2.5	63.6 ± 3.2	30.1 ± 1.5	0.5	0.19 ± 0.02	0.10 ± 0.01	4.5 × 10 <sup>3</sup>	3.1 × 10 <sup>4</sup>	1.8 × 10 <sup>4</sup>
2mL (0.7 × 5cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 E-4, 2.5	40.4 ± 2.0	37.1 ± 1.9	0.9	0.18 ± 0.02	0.07 ± 0.01	1.1 × 10 <sup>4</sup>	1.6 × 10 <sup>3</sup>	7.2 × 10 <sup>3</sup>
1mL (0.5 × 5cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 E-4, 2.5	50.7 ± 2.5	44.8 ± 2.2	0.9	0.14 ± 0.02	0.10 ± 0.01	4.1 × 10 <sup>3</sup>	4.0 × 10 <sup>3</sup>	1.5 × 10 <sup>4</sup>

The reference values of <sup>239</sup>Pu and <sup>240</sup>Pu concentration in the Danish soil were reported to be 0.140 ± 0.008 Bq/kg and 0.098 ± 0.006 Bq/kg.\* Experimental results are given as the average of three replicates ± standard deviation. \*\* The relative standard deviations were in all instances better than 10%. § flow rate, mL/min.# W-1: washing sequence 200mL of 8 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-2: 100mL of 8 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-3: 100mL of 6 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-4: 100mL of 4 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-5: 100mL of 2 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-6: 100mL of 1 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; Pu eluting solution: E-1:Pu elution solution 200mL of 0.1 mol/L NH<sub>2</sub>OH-HCl-2 mol/L HCl; E-2: 100mL of 0.1 mol/L NH<sub>2</sub>OH-HCl-2 mol/L HCl; E-3: 40mL of 0.1 mol/L NH<sub>2</sub>OH-HCl-2 mol/L HCl; E-4: 40mL of 0.5 mol/L HCl; E-5: 40mL of 0.1 mol/L HCl; E-6: 40mL of 1.0 mol/L HCl.

**Table 3. Comparison of different experimental conditions for the separation of Pu and Np (1)**

Column size	Resin	Separation condition#	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
16mL (1.0 × 20cm)	AG1 × 2	W-1, 2.5 E-1, 1.0	😊	😊	😊	😊	😊	😊	😊	😊
	AG1 × 4		😊	😊	😊	😞	😞	😞	😊	😊
	AG1 × 8		😊	😊	😊	😞	😊	😞	😞	😊
8mL (0.7 × 20cm)	AG1 × 2	W-1, 2.5 E- 2, 1.0	😞	😞	😊	😊	😊	😊	😊	😊
	AG1 × 4		😊	😊	😊	😊	😊	😊	😊	😊
	AG1 × 8		😊	😊	😊	😊	😊	😞	😞	😊
	AG1 × 8	W-1, 5.0 E-2, 2.5	😊	😞	😊	😞	😞	😞	😞	😊
4mL (0.7 × 10cm)	AG1 × 2	W-1, 2.5 E-2, 1.0	😞	😞	😞	😊	😞	😊	😊	😊
	AG1 × 4		😊	😊	😊	😊	😊	😊	😊	😊
	AG1 × 8		😊	😊	😊	😞	😊	😊	😞	😊
	AG1 × 8	W-1, 5.0 E-2, 2.5	😊	😊	😊	😞	😞	😞	😞	😊

**Table 3. Comparison of different experimental conditions for the separation of Pu and Np (2)**

Column size	Resin	Separation condition#	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
2mL (0.5 × 10cm)	AG1 × 2	W-2, 2.5 E-3, 2.5	☹	☹	☹	☹	☹	☹	☹	☺
		W-2, 5.0 E-3, 2.5	☹	☹	☹	☺	☺	☹	☹	☺
	AG1 × 4	W-2, 2.5 E-3, 2.5	☺	☺	☺	☺	☺	☺	☺	☺
		W-2, 5.0 E-3, 2.5	☺	☺	☺	☹	☺	☹	☺	☺
	AG1 × 8	W-2, 2.5 E-3, 2.5	☺	☺	☺	☹	☺	☹	☹	☺
		W-2, 5.0 E-3, 2.5	☺	☺	☺	☹	☹	☹	☹	☺
	AG1 × 4	W-2, 2.5 E-4, 2.5	☹	☹	☺	☹	☹	☺	☺	☺
		W-3, 2.5 E-4, 2.5	☹	☹	☹	☹	☺	☺	☺	☺
		W-4, 2.5 E-4, 2.5	☹	☹	☹	☹	☺	☺	☺	☺
		W-5, 2.5 E-4, 2.5	☹	☹	☹	☹	☹	☺	☺	☺
W-6, 2.5 E-4, 2.5		☹	☹	☹	☹	☹	☺	☺	☺	

**Table 3. Comparison of different experimental conditions for the separation of Pu and Np (3)**

Column size	Resin	Separation condition #	Chemical yield of <sup>242</sup> Pu, Y <sub>Pu</sub> (%)	Chemical yield of <sup>237</sup> Np, Y <sub>Np</sub> (%)	Ration of Y <sub>Np</sub> /Y <sub>Pu</sub>	<sup>239</sup> Pu measured (Bq/kg) *	<sup>240</sup> Pu measured (Bq/kg) *	Decontamination factor **		
								<sup>238</sup> U	<sup>232</sup> Th	Pb
2mL (0.5 × 10cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 § E-3, 2.5	☹	☹	☺	☹	☹	☺	☺	☺
		W-2, 2.5 E-4, 2.5	☺	☺	☺	☺	☺	☺	☺	☺
		W-2, 2.5 E- 5, 2.5	☹	☹	☺	☺	☺	☺	☺	☺
		W-2, 2.5 E-6, 2.5	☹	☹	☺	☺	☹	☺	☺	☺
		W-3, 2.5 E-4, 2.5	☹	☹	☹	☹	☹	☺	☺	☺
		W-4, 2.5 E-4, 2.5	☹	☹	☺	☹	☹	☺	☺	☺
		W-5, 2.5 E-4, 2.5	☹	☹	☹	☹	☺	☺	☺	☺
		W-2, 2.5 E-4, 2.5	☹	☹	☺	☹	☹	☺	☺	☺
2mL (0.7 × 5cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 E-4, 2.5	☹	☹	☺	☹	☹	☺	☺	☺
1mL (0.5 × 5cm)	AG1 × 4, 100-200 mesh	W-2, 2.5 E-4, 2.5	☹	☹	☺	☺	☺	☺	☺	☺

The reference values of <sup>239</sup>Pu and <sup>240</sup>Pu concentration in the Danish soil were reported to be  $0.140 \pm 0.008$  Bq/kg and  $0.098 \pm 0.006$  Bq/kg.\* Experimental results are given as the average of three replicates  $\pm$  standard deviation. \*\* The relative standard deviations were in all instances better than 10%. § flow rate, mL/min.# W-1: washing sequence 200mL of 8 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-2: 100mL of 8 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-3: 100mL of 6 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-4: 100mL of 4 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-5: 100mL of 2 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; W-6: 100mL of 1 mol/L HNO<sub>3</sub> + 100mL of 9 mol/L HCl; Pu eluting solution: E-1:Pu elution solution 200mL of 0.1 mol/L NH<sub>2</sub>OH·HCl-2 mol/L HCl; E-2: 100mL of 0.1 mol/L NH<sub>2</sub>OH·HCl-2 mol/L HCl; E-3: 40mL of 0.1 mol/L NH<sub>2</sub>OH·HCl-2 mol/L HCl; E-4: 40mL of 0.5 mol/L HCl; E-5: 40mL of 0.1 mol/L HCl; E-6: 40mL of 1.0 mol/L HCl.

## Main Results

- 1)  $^{242}\text{Pu}$  performs well as a tracer for both Pu isotope and  $^{237}\text{Np}$ .
- 2) Cross-link of the resins has significant influence on the separation efficiency. **Finally, AG 1x4 resin was chosen as the optimum.**
- 3) **Small-sized column packed with 2mL resin suffices up to 10g of soil.**

**Table 4. Selected results from the experiment (10g of soil)**

Column size	Resin	Chemical yield of $^{242}\text{Pu}$ , $Y_{\text{Pu}}$ (%)	Chemical yield of $^{237}\text{Np}$ , $Y_{\text{Np}}$ (%)	Ration of $Y_{\text{Np}}/Y_{\text{Pu}}$	$^{239}\text{Pu}$ measured (Bq/kg) *	$^{240}\text{Pu}$ measured (Bg/kg)**	Decontamination factor ***		
							$^{238}\text{U}$	$^{232}\text{Th}$	$^{208}\text{Pb}$
2mL (0.5 × 10cm)	AG1 × 4, 50-100 mesh	103.0 ± 5.2	106.0 ± 5.3	1.0	0.14 ± 0.01	0.09 ± 0.01	3.9 × 10 <sup>3</sup>	2.4 × 10 <sup>4</sup>	2.7 × 10 <sup>4</sup>
	AG1 × 4, 100-200 mesh	91.6 ± 4.6	91.0 ± 4.6	1.0	0.14 ± 0.01	0.10 ± 0.01	6.9 × 10 <sup>3</sup>	1.7 × 10 <sup>4</sup>	1.0 × 10 <sup>3</sup>

\*The reference value is  $0.140 \pm 0.008$  Bg/kg. \*\*The reference value is  $0.098 \pm 0.006$  Bg/kg.

\*\* The relative standard deviations were in all instances better than 10%.

## Main Results

- 4) The total time of on-line separation for a single sample is ~ **2.5h**.  
**For comparison: 2-3days is need for off-line separation.**
- 5) Chemical yields of Pu and Np equally range from 90% to 100%.
- 6) Decontamination factor for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{208}\text{Pb}$  are in the range of  $10^3$  to  $10^4$ .

Table 4. Selected results from the experiment (10g of soil)

Column size	Resin	Chemical yield of $^{242}\text{Pu}$ , $Y_{\text{Pu}}$ (%)	Chemical yield of $^{237}\text{Np}$ , $Y_{\text{Np}}$ (%)	Ration of $Y_{\text{Np}}/Y_{\text{Pu}}$	$^{239}\text{Pu}$ measured (Bq/kg) *	$^{240}\text{Pu}$ measured (Bg/kg)**	Decontamination factor ***		
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	AG1 × 4, 100-200 mesh	91.6 ± 4.6	91.0 ± 4.6	1.0	0.14 ± 0.01	0.10 ± 0.01	6.9 × 10 <sup>3</sup>	1.7 × 10 <sup>4</sup>	1.0 × 10 <sup>3</sup>

\*The reference value is  $0.140 \pm 0.008$  Bg/kg. \*\*The reference value is  $0.098 \pm 0.006$  Bg/kg.

\*\*\* The relative standard deviations were in all instances better than 10%.

- 1) Innovation:** Automatic  
Rapid  
Simultaneous  
Low consumption of resins  
Low generation of wastes
- 2) Next step:** Stability of Np(IV) and Pu(IV)  
Capacity of the SI system  
Reusability of the resin



- **Xiaolin Hou**
- **Per Roos**
- **Manuel Miró**
- **Radioecology and Tracers Programme (headed by Sven P. Nielsen), Radiation Research Division, Risø-DTU, Denmark.**

**THANK YOU !**

Personal e-mail: [qiaojixin2004@gmail.com](mailto:qiaojixin2004@gmail.com)