The Rapid Determination of Strontium-89 and Strontium-90 in Environmental Samples

Terence O'Brien*, Lawrence Jassin,
E. Philip Horwitz and Daniel McAlister
Eichrom Technologies and PGRF

Presented at the MARC IX: Conference

Kailua-Kona, HI
March 29, 2012

eichrom
expertise. collaboration. results.

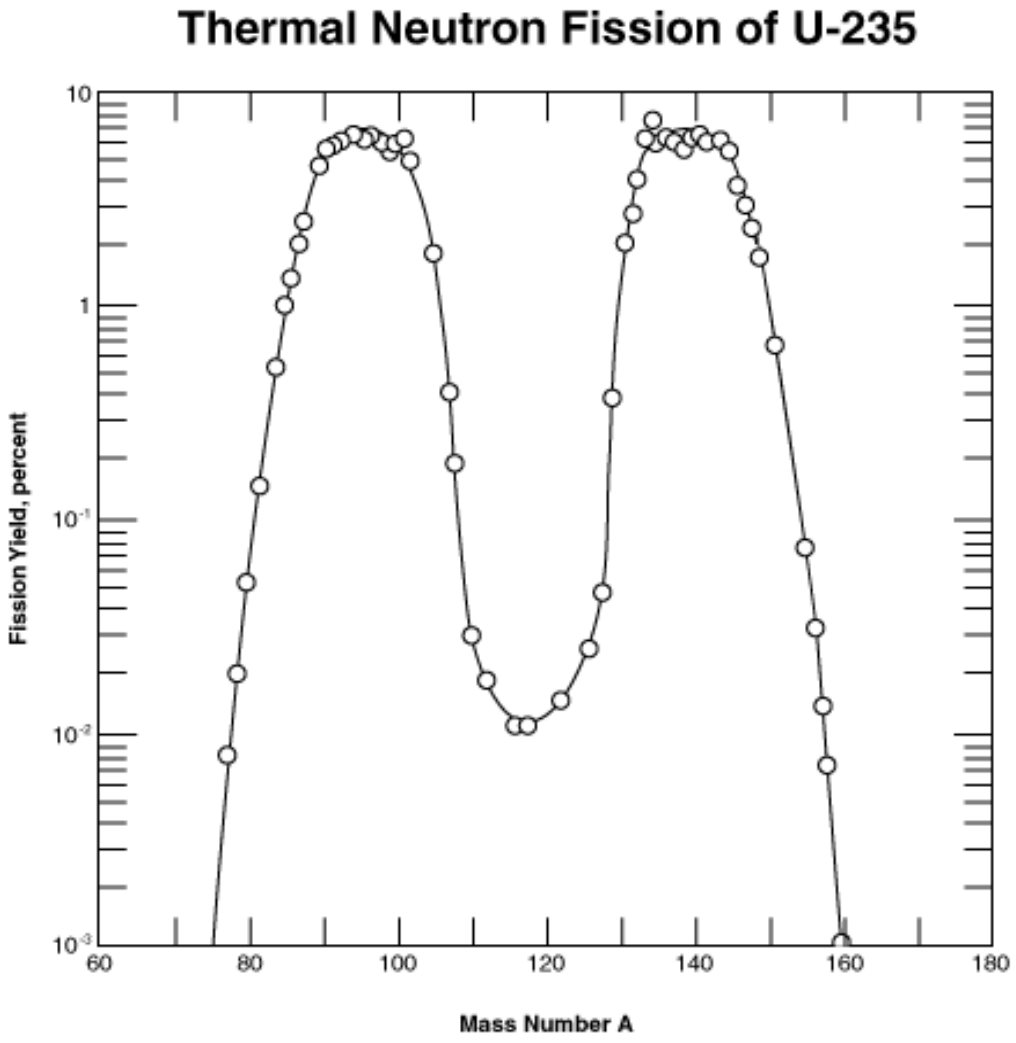
www.eichrom.com

Monday, December 6th, 2011: Japan Today

Some 45 tons of highly radioactive water leaked Sunday from desalination equipment used to decontaminate the radioactive water in Tokyo Electric Power Co.'s Fukushima No. 1 nuclear plant and it is unclear if any made it to the sea, a Tepco official said Monday…

The utility is testing seawater samples taken off the coast near the plant to see if it is contaminated with strontium. It will know the results in about two to three weeks, Tepco said.

Strontium-89 & Strontium-90



eichrom
expertise. collaboration. results.

www.eichrom.com

Strontium-89 & Strontium-90

Strontium-89

$T_{1/2}$ = **50.53 days**

Mean Beta energy = 584.6 keV

Mean beta-dose = 0.5846 MeV/Bq-s

Daughter Yttrium-89 Stable

Strontium-90

$T_{1/2}$ = **28.90 years**

Mean Beta Energy = 195.8 keV

Mean beta-dose = 0.196 MeV/Bq-s

Daughter

Yttrium-90

$T_{1/2}$ = **64.0 hours**

Mean Beta Energy = 933.6 keV

Mean beta-dose = 0.9336 MeV/Bq-s

Daughter Zirconium-90 Stable

The Issues

Some methods for the determination of radioactive strontium take one of the following approaches.

1. Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Water for Environmental Restoration Following Homeland Security Events:

2.2.1 This test assumes that it is reasonable to assume the absence of ^{89}Sr in the sample.

2. Eichrom SRW01 – Strontium 89, 90 in Water

7.9.3 “After yttrium ingrowth of approximately 1 week”

The Issues

3. Rapid separation of actinides and radiostrontium in vegetation samples

If $^{89/90}$Sr differentiation is needed, there are Čerenkov counting techniques for more rapid determination of ^{89}Sr and ^{90}Sr. ^{89}Sr can be measured directly by Čerenkov counting, employing methodology that takes advantage of the high Čerenkov counting efficiency of ^{89}Sr relative to ^{90}Sr [11].

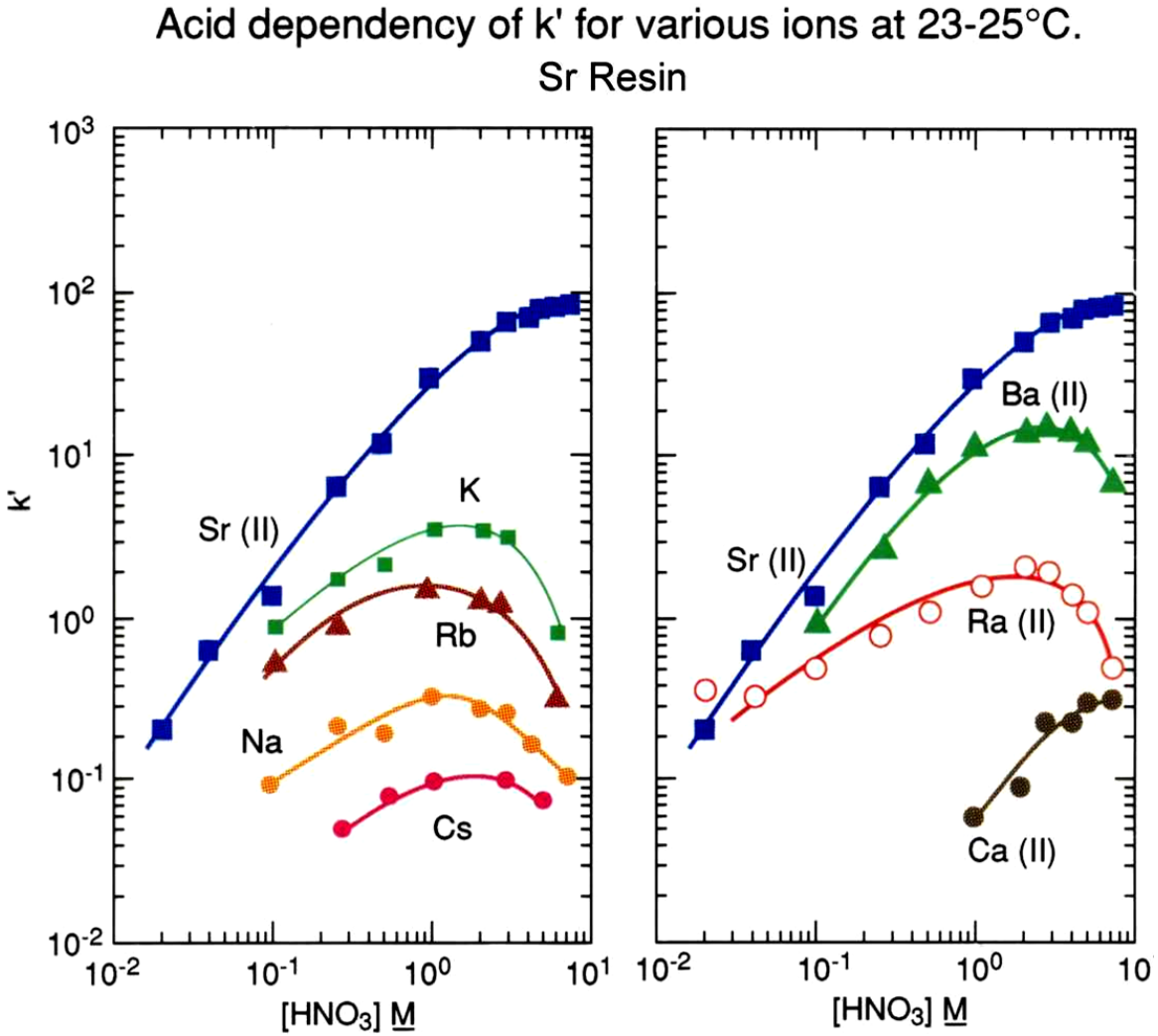
Assumptions

1. Strontium-89 and Strontium-90 could be present in a sample.
2. Strontium-90 and Yttrium-90 will likely be in equilibrium.
3. Rapid sample turnaround required. – 8 Hrs
4. Many samples to be analyzed. – Multi-Detector

1st Tool: Preconcentration

1. For Sr Resin – Sr k' around 80-90. Therefor sample volumes above 50 mL should be avoided to minimize Sr break through.
2. Water sample evaporation is an option but the clock is ticking.
3. Cation Exchange
4. A fast calcium phosphate precipitation can concentrate both Sr and Y.

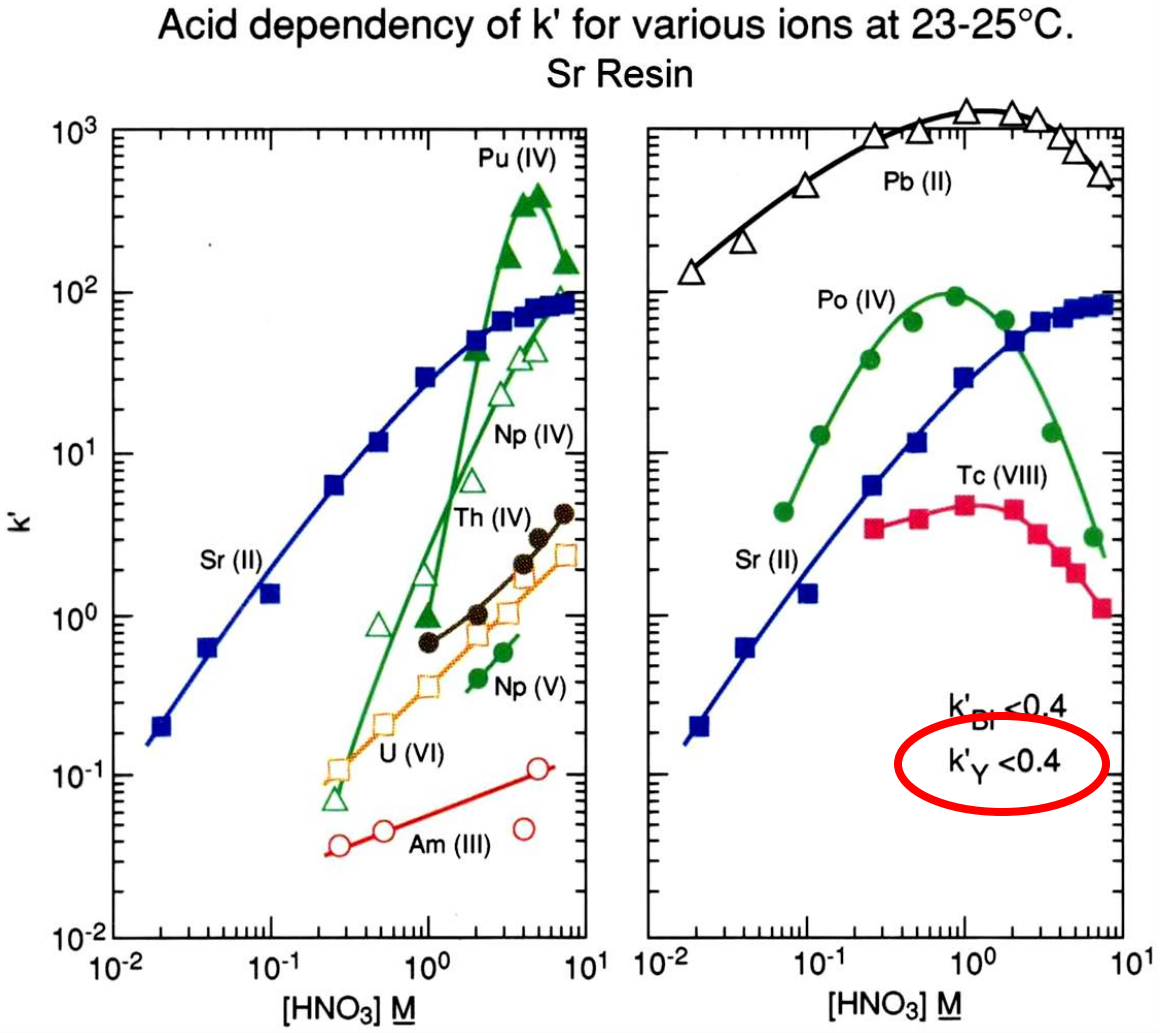
2nd Tool: Sr Resin



eichrom
expertise. collaboration. results.

www.eichrom.com

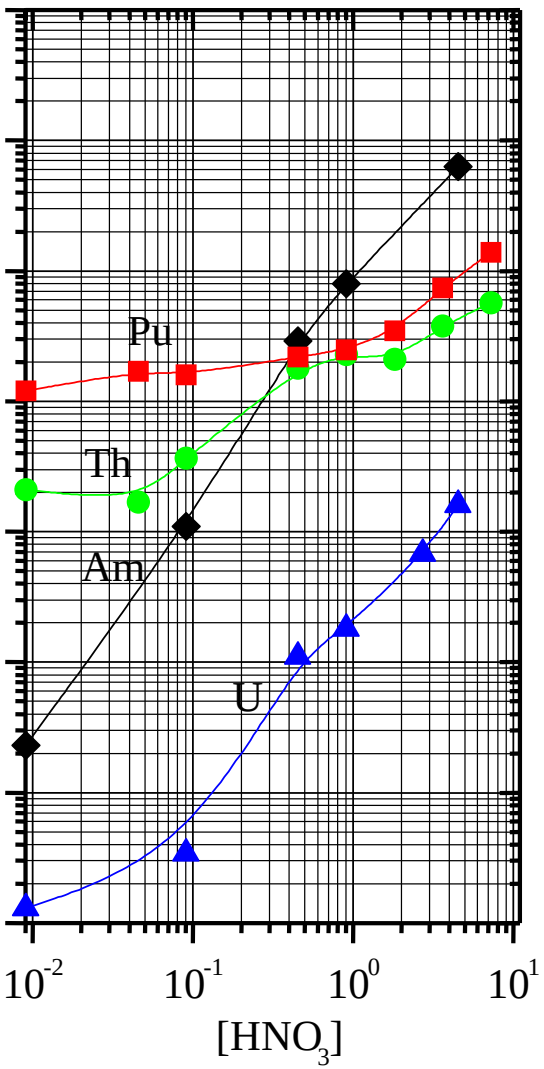
Sr Resin

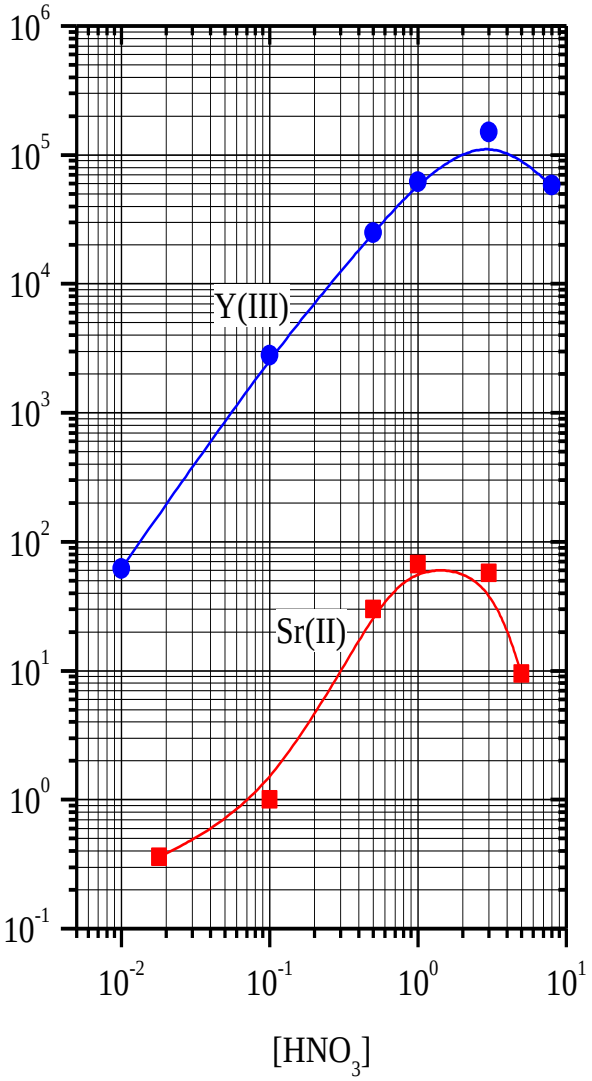


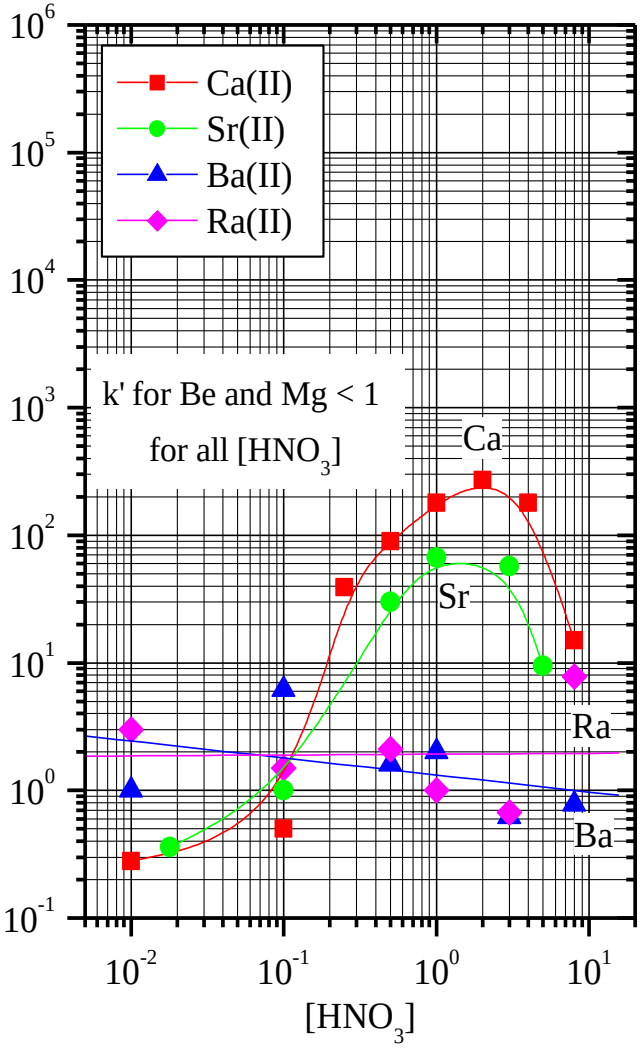
eichrom
expertise. collaboration. results.

www.eichrom.com

3rd Tool: DGA Resin







4th Tool Vacuum Box

Sample flow rates

1. Recommended: Load/Elution 1 mL / min
   Rinse rates of 3 mL / min
2. 2007 RRMC - Julie Gostic reports work with flow rates of 6-7 mL/min with good recoveries for Am-241 on TRU and DGA
3. 2009 Maxwell publishes the use of 2-4 mL/min for rapid / emergency sample analysis.

Flow Sheet: Rapid Sr-89 & Sr-90 Preconcentration

Aliquot & Acidify <pH 2

↓

Add Sr & Y Carrier/Tracer and Matrix Spikes

↓

Add Calcium Nitrate, Barium Chloride, Ammonium Phosphate, Phenolphthalein

↓

Add Concentrated Ammonium Hydroxide To dark pink phenolphthalein end point

↓

Centrifuge 2000-3000 rpm 5-10 minutes

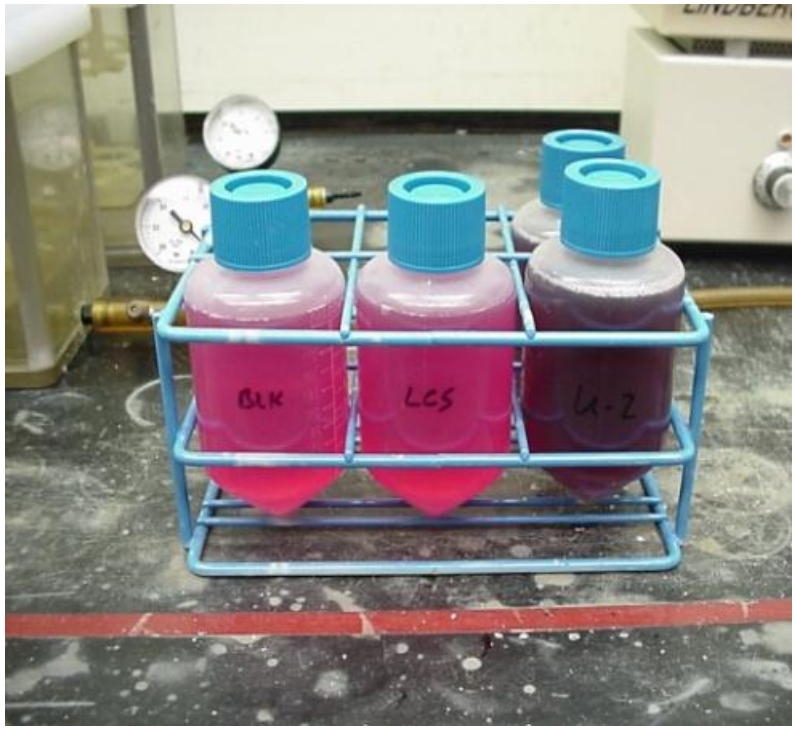
↓

Pour off supernate

→ Dissolve Precipitate with 8M Nitric Acid and 0.5M Aluminum Nitrate

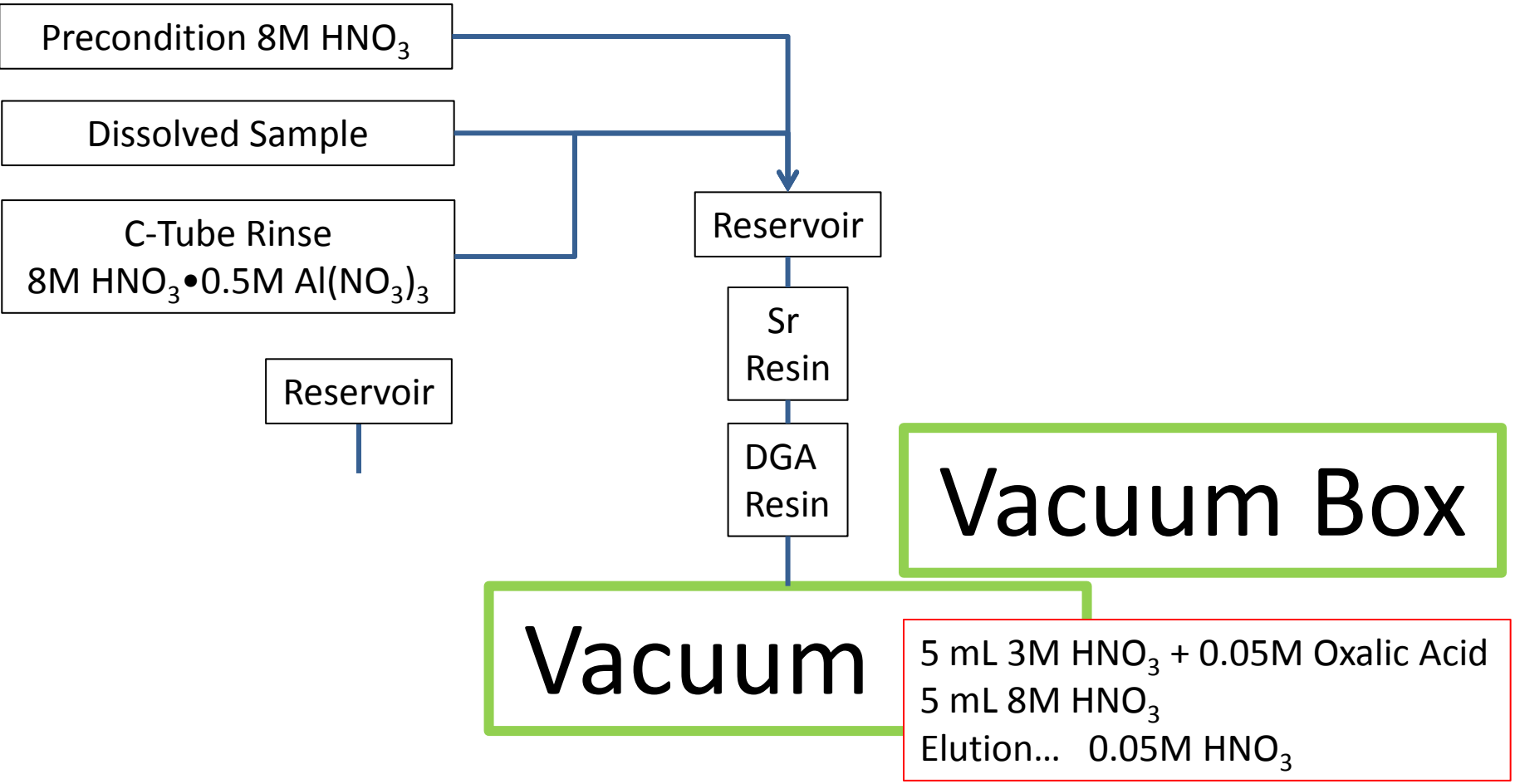
↓

Dispose of Liquid

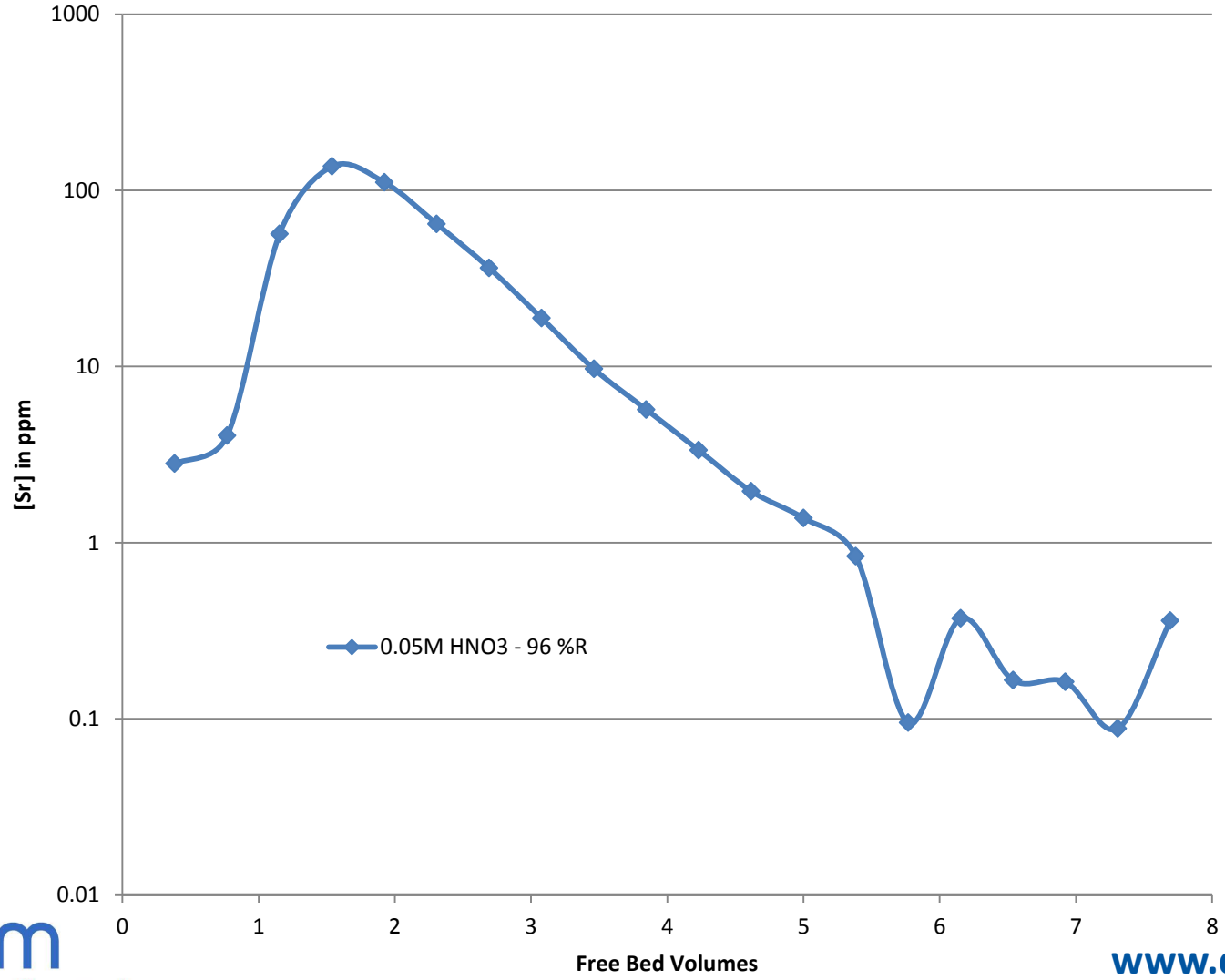


eichrom
expertise. collaboration. results.

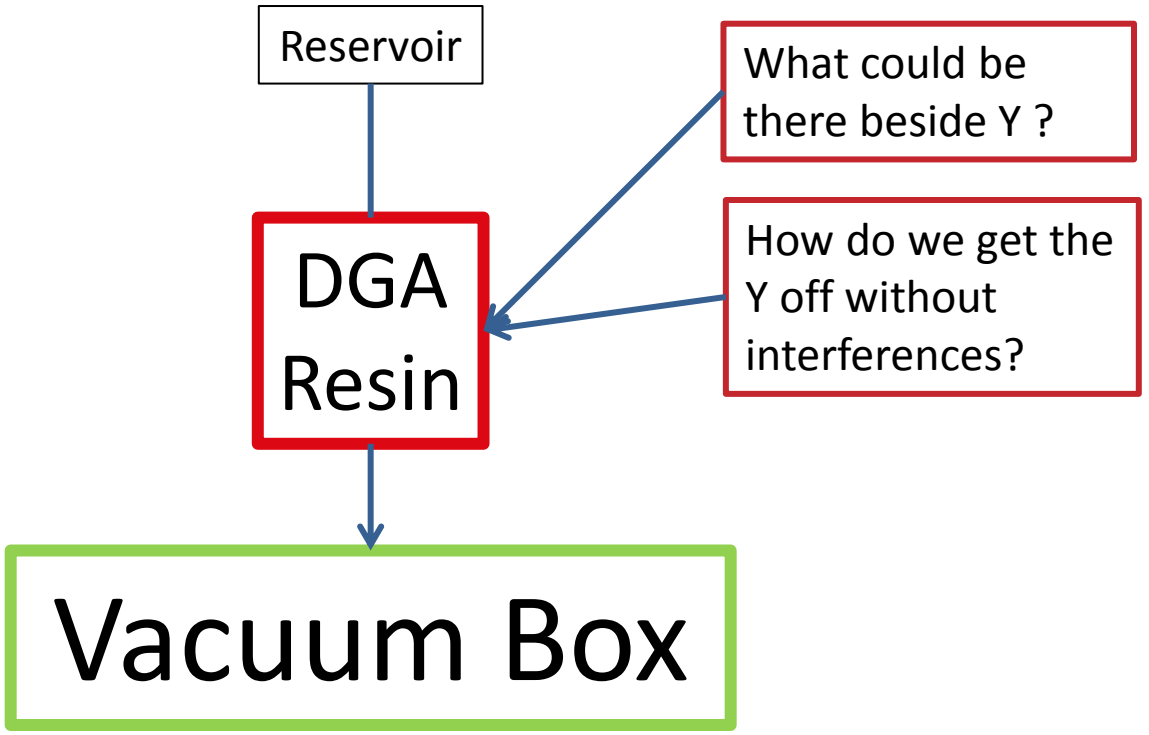
Strontium / Yttrium Separation and Retention

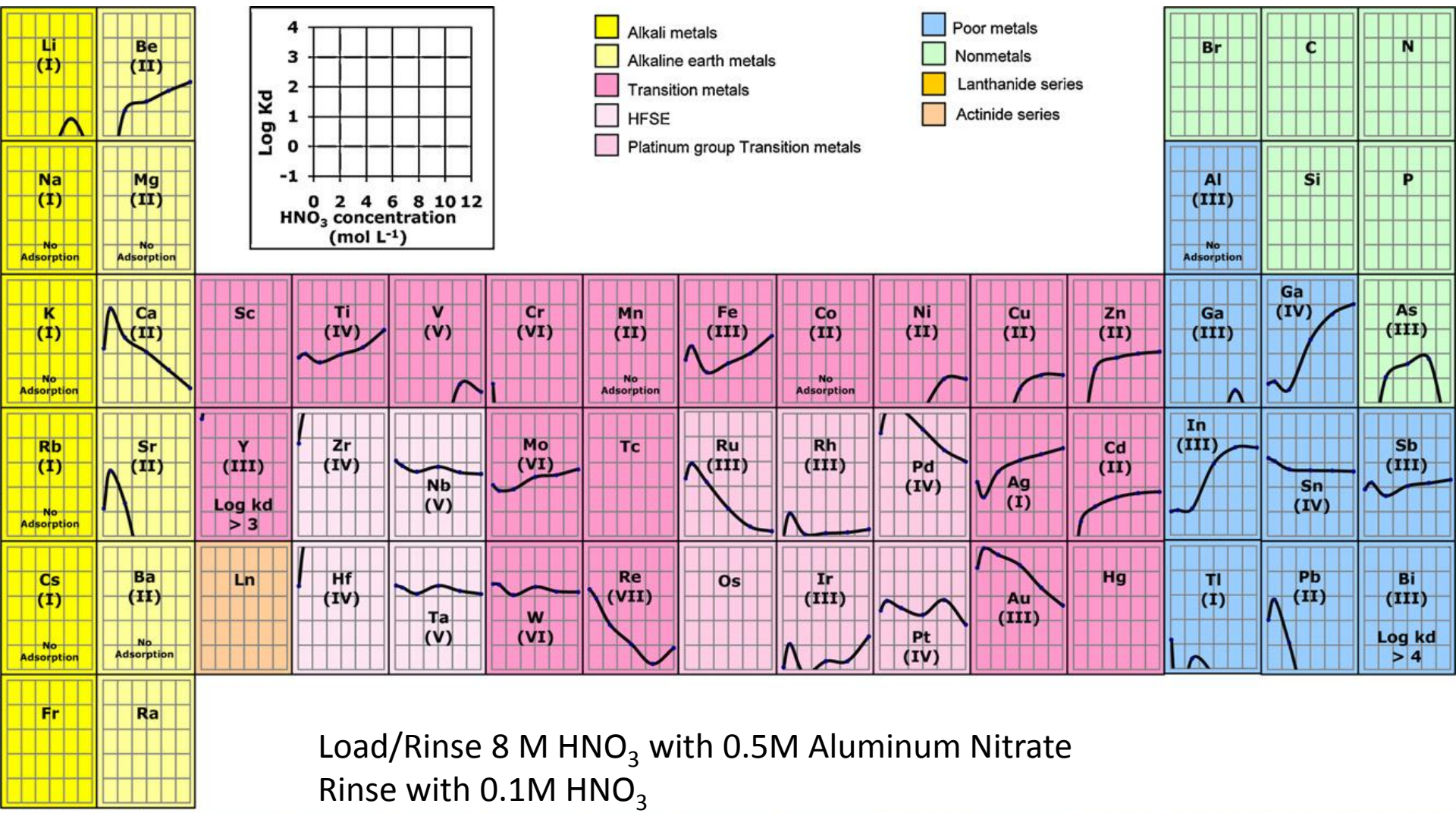


Strontium Elution



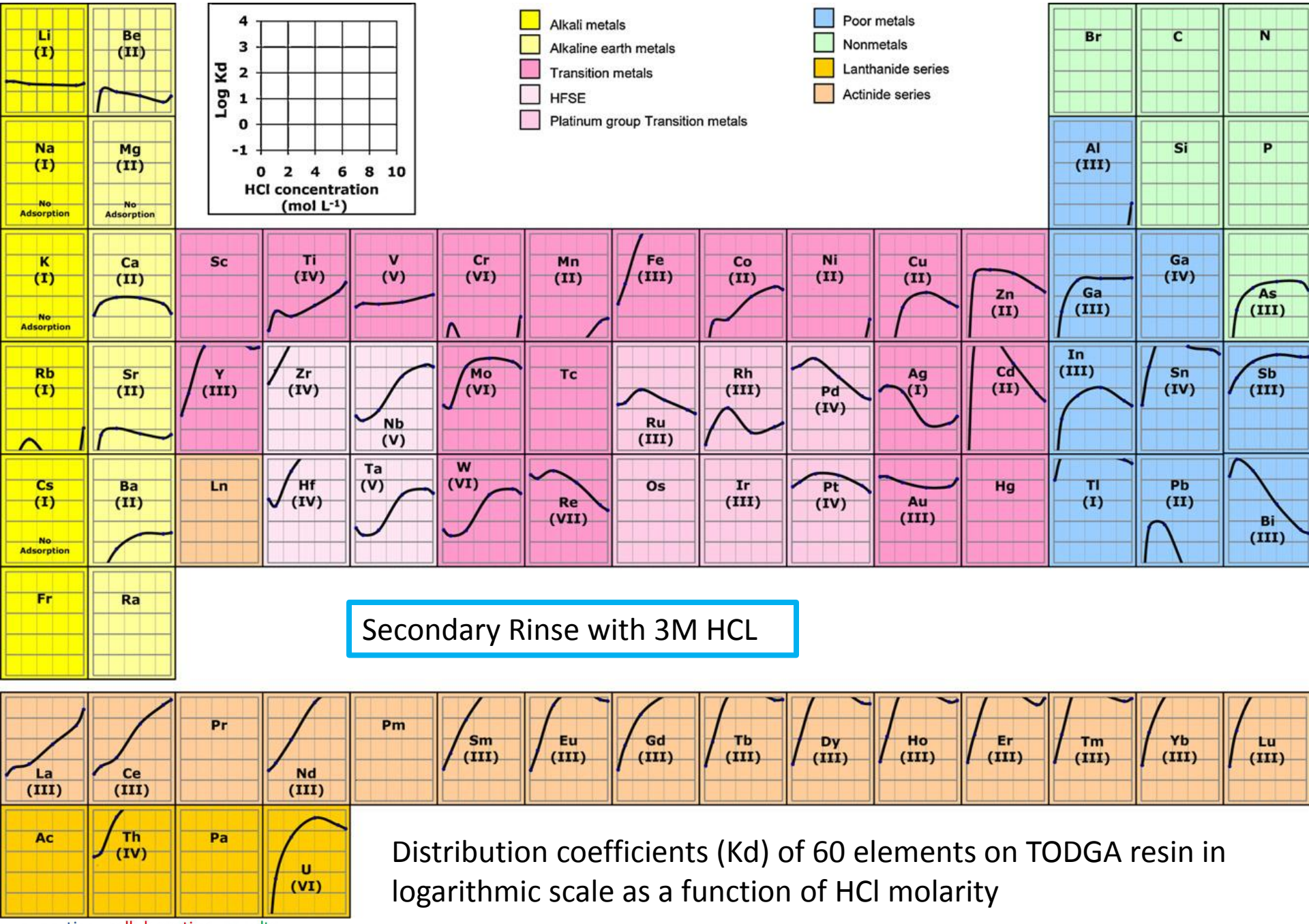
Yttrium on DGA…





Load/Rinse 8 M HNO_3 with 0.5M Aluminum Nitrate
Rinse with 0.1M HNO_3

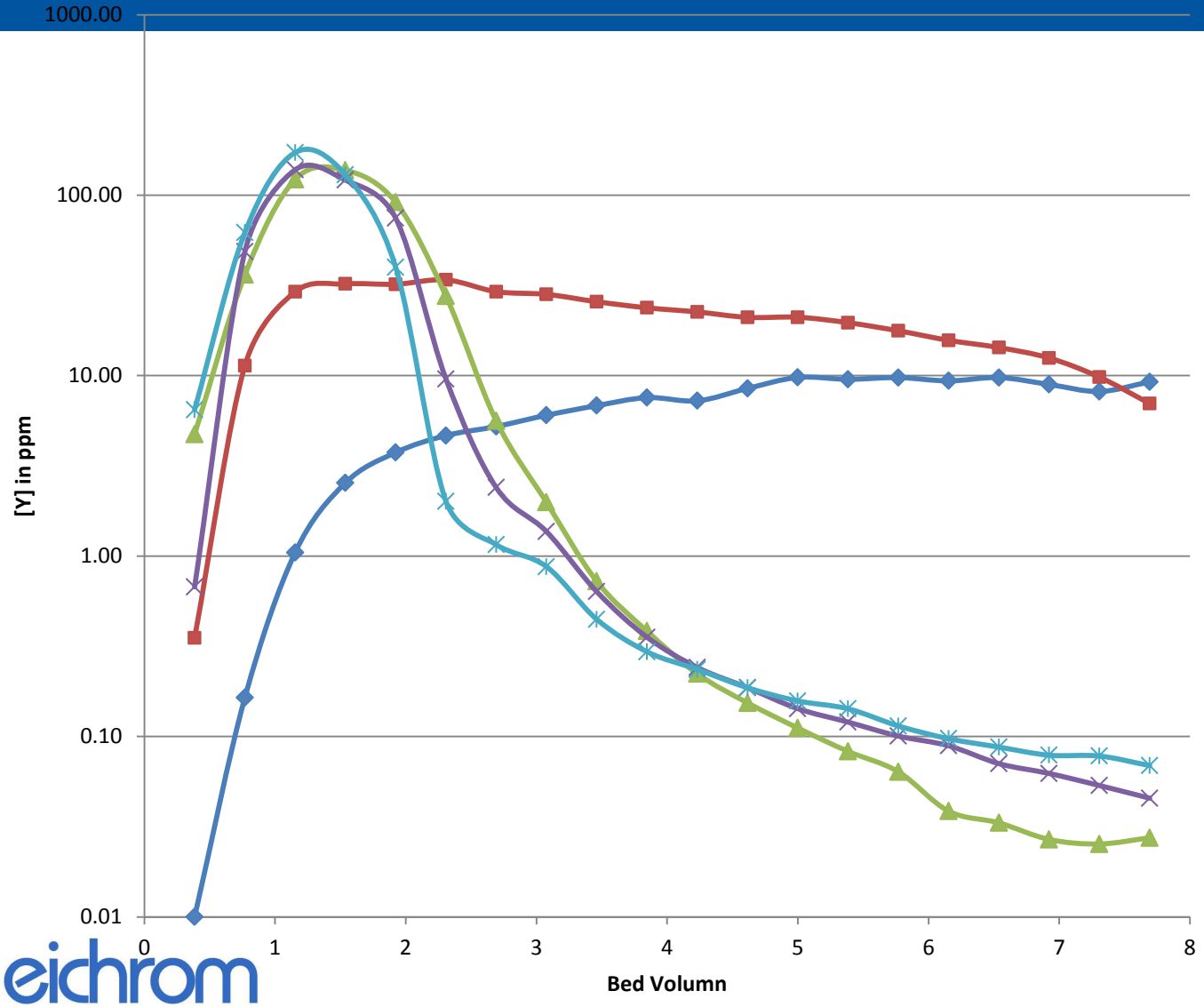
Distribution coefficients (Kd) of 60 elements on TODGA resin in logarithmic scale as a function of HNO_3 molarity



Secondary Rinse with 3M HCL

Distribution coefficients (Kd) of 60 elements on TODGA resin in logarithmic scale as a function of HCl molarity

Yttrium elution from DGA



1M HCl - 27.1 %R
0.5M HCl - 86.2 %R
0.1M HCl - 90.8 %R
1M Acetic - 84.4 %R
0.5M Acetic - 88.1 %R

eichrom
expertise. collaboration. results.

www.eichrom.com

Beta Detection

LSC/Cherenkov

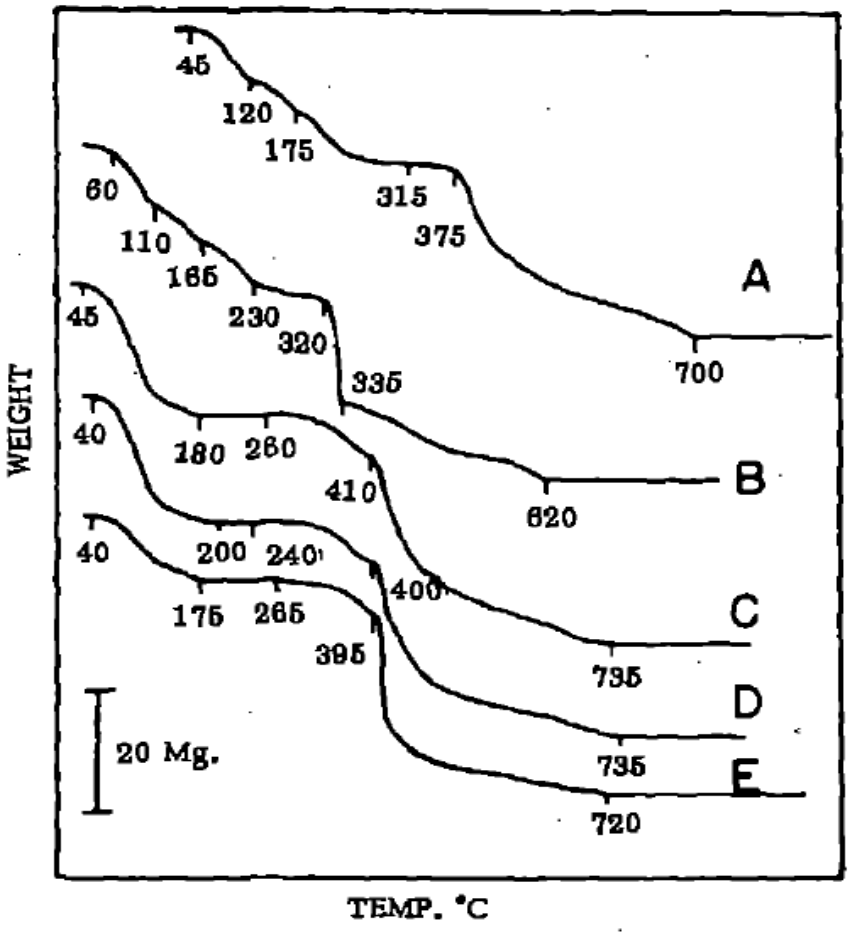
GPC Low Background Counters

Carrier Recoveries

Matrix/Carrier	Avg.Recovery	RSD%
De-ionized Water		
Strontium	84.9%	4%
Yttrium	--	--
Tap Water:		
Strontium	75.0%	7%
Yttrium	--	

Waters; n=4

Yttrium and REEs Waters of Hydration
Thermal Gravimetric Analysis



The issue:
C: Yttrium Oxalate has many waters of Hydration

The solution:
Conversion to Yttrium Oxide at 800

	Sr-89			Sr-90		
Sample	Certified Value	Measured Value	Difference ±%	Certified Value	Measured Value	Difference ±%
Blank		-0.35±0.37			0.30±023	
De-ionized Water-A	2.39 ±0.05	2.8±1.2	+18	1.20 ±0.02	0.72±0.27	-40
De-ionized Water-B		2.4±1.0	+2		0.97±0.33	-19
De-ionized Water-C		2.0±.81	-16		1.21±0.38	+1
Tap Water-A	2.39 ±0.05	2.7±1.1	+13	1.20 ±0.02	1.12±0.38	-7
Tap Water-B		2.5±1.0	+5		1.24±0.39	3
Tap Water-C		3.1±1.3	+30		1.18±0.38	-2

Sample count time was 20 minutes and results are in Bq/L

Estimated Time Line (10 samples)

Step	Minutes	Elapsed Time (hrs)
Sample Aliquot & Spiking	30	0:30
Sample Pre-Concentration	45+	1:15
Sample Separation Sr & DGA	30	1:45
Sr Purification	30	2:15
Y Purification (Concurrent with Sr Purification)	30	--
Sample Mounting (Sr & Y)	45	3:00
Counting Time Sr-89 – 3.5 Bq/L MDA Sr-90 – 3.0 Bq/L MDA	20+	3:20
Data Reduction	20	4:00

Future Work

1. Yttrium Yield Measurement Technique
    a) Oxalates and Hydroxides
    b) Gravimetric vs AA/ICP

2. Interference Rejection

3. Additional Matrix Preparation

Acknowledgments

Phil Horwitz, Dan McAlister, Larry Jassin at
PRGF and Eichrom
Tim Winters at GEL Labs
Bob Shannon at Quality Radioanalytical Support
Environmental Resource Associates

Thank you for you attention and your questions.