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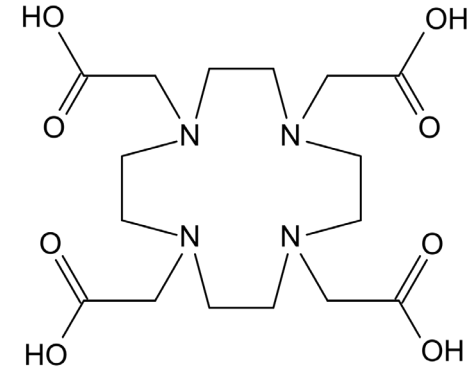
Separations from Buffers

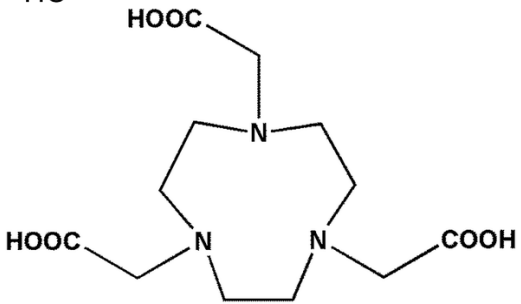
Daniel McAlister and Madeleine Eddy

66th RRMC, West Palm Beach, FL, Oct 29 – Nov 3, 2023

RRMC
Radiobioassay & Radiochemical Measurements Conference

Diagnostic and Therapeutic Nuclear Medicine





$^{64/67}Cu$

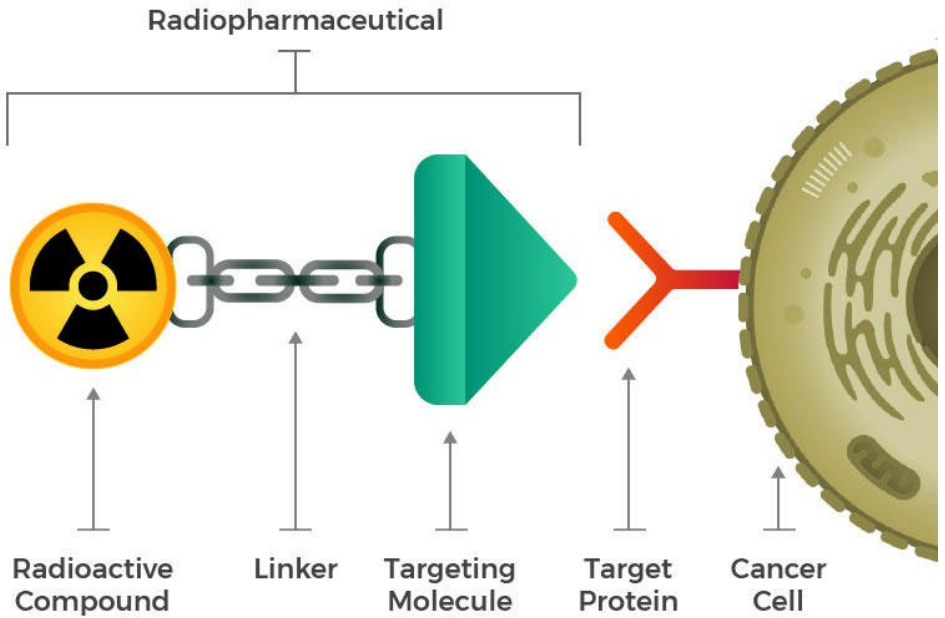
^{68}Ga

^{90}Y

^{177}Lu

$^{203/212}Pb$

^{225}Ac



Chelate radiometals from pH 4-7 buffer.

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https://www.cancer.gov/news-events/cancer-currents-blog/2020/radiopharmaceuticals-cancer-radiation-therapy

Acidic solutions vs pH 3-6

- Adsorption, hydrolysis, oxidation state control and the differences between high specific activity (low mass) and low specific activity/stable metal (μgs) can be very different in highly acidic vs pH 3-6 solutions.

- Maintaining higher ionic strength can help (NaCl, buffer concentrations).

- Initial work can be done with stable metals, but results should be verified with high specific activity radionuclides.

Goals

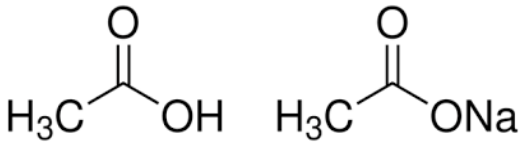
1) Study elution of nuclear medicine radionuclides from acidic solution into buffers

2) Choose systems where you can achieve

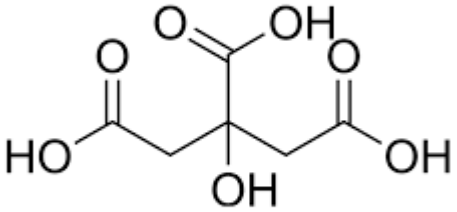
    a) High yield
    b) Elution in a small volume
    c) Concentration factor and purification from impurities

3) Purification of chelated radionuclides from buffer solutions

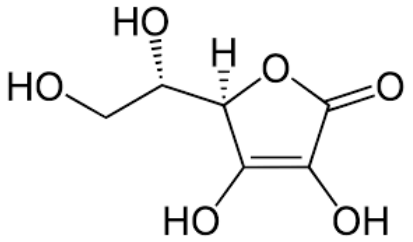
Common buffer components



Sodium Acetate
Buffer pH = 3.6-5.6



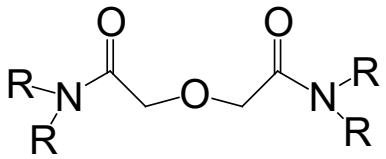
Citric acid
Buffer pH = 3.0 - 6.2



Ascorbic acid
Buffer pH = 3.5 – 5.5
Antioxidant/reducing agent
Helps reduce impact of radiation damage

NaCl
Sodium chloride
Adjust ionic strength

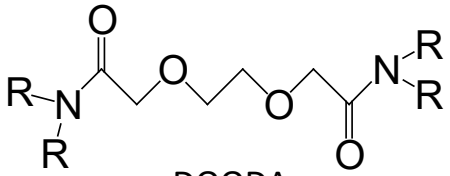
Neutral



DGA

Normal = n-octyl

Branched = 1-ethyl-2-hexyl

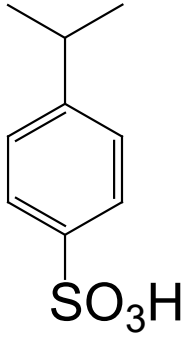


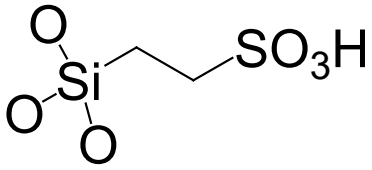
DOODA

R = n-octyl

Retain metals from >1 HNO3/HCl.
Recover metals in dilute HCl or buffer solution.

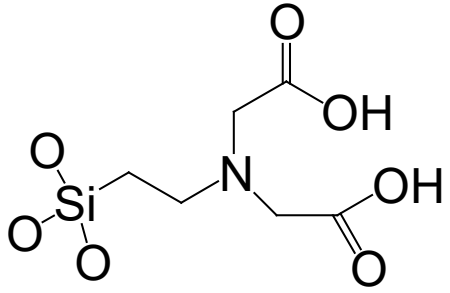
SCX





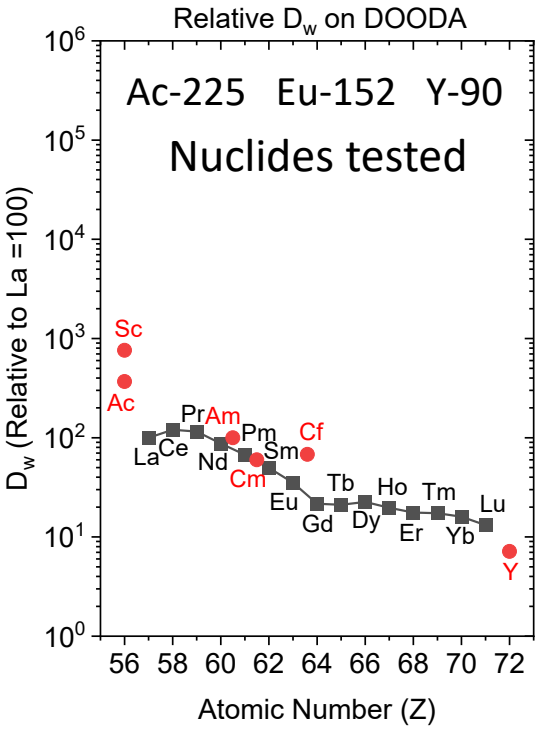
Retain metals from dilute HNO3/HCl.
Recover metals in >1M HCl or buffer solution.

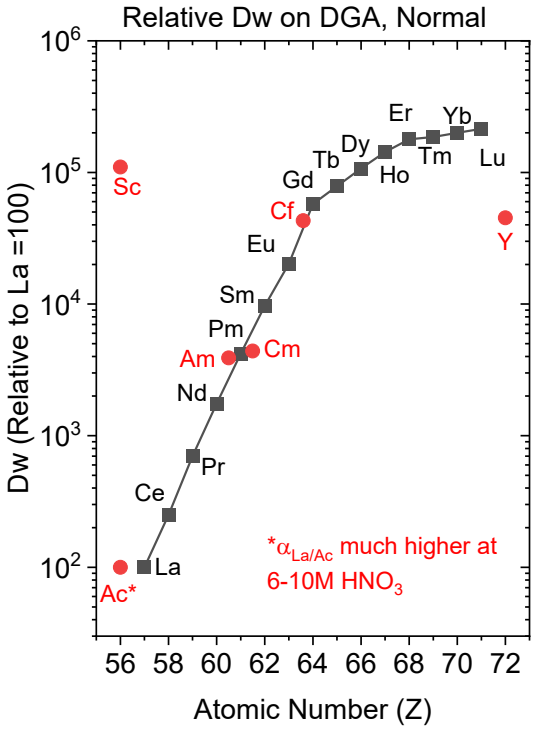
WCX (CM)

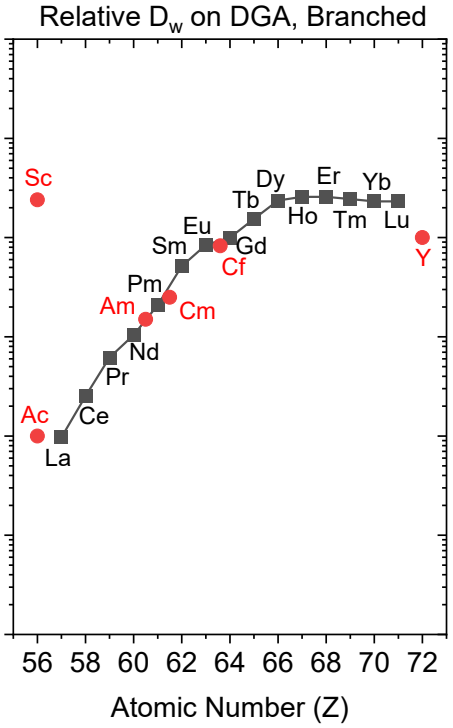


Retain metals from >1 buffer
Recover metals in dilute HCl.

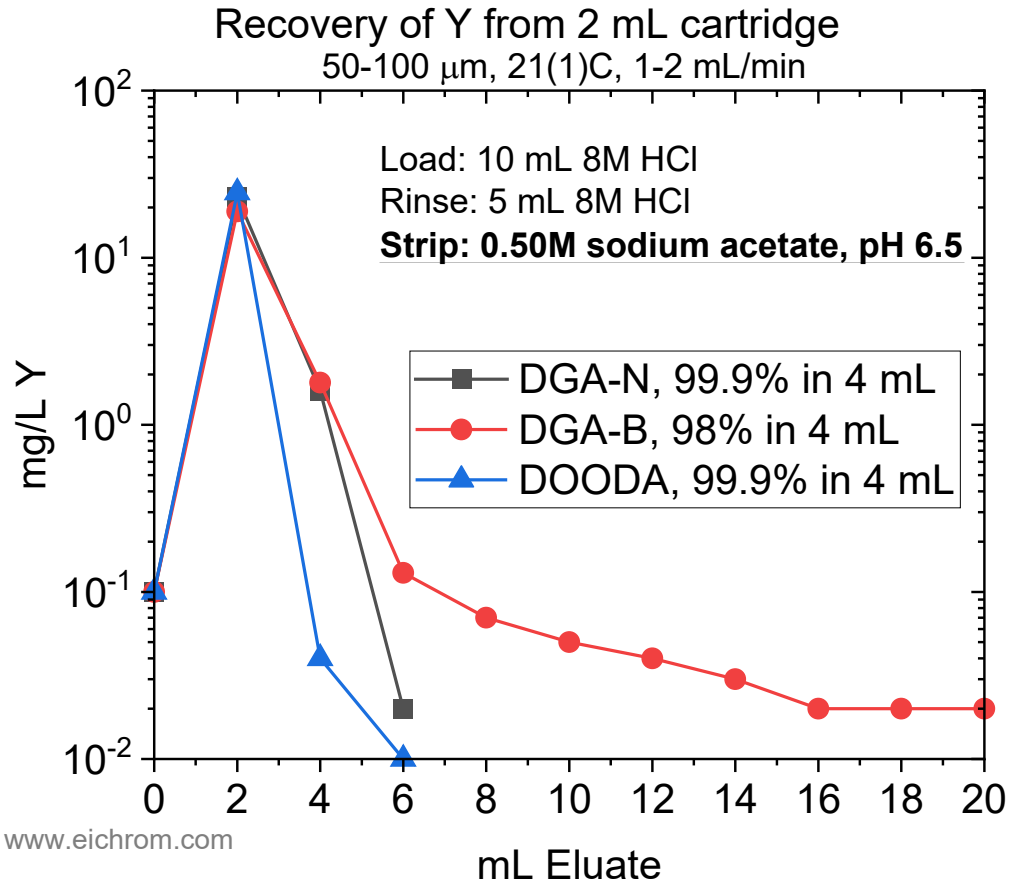
DGA/DOODA


Relative D_w on DOODA
Ac-225 Eu-152 Y-90
Nuclides tested
D_w (Relative to La =100)
Atomic Number (Z)
Sc
Ac
La
Ce
Pr
Nd
Pm
Am
Sm
Cm
Eu
Gd
Cf
Tb
Dy
Ho
Er
Tm
Yb
Lu
Y


Relative Dw on DGA, Normal
Dw (Relative to La =100)
Atomic Number (Z)
Sc
Ac*
La
Ce
Pr
Nd
Pm
Am
Sm
Cm
Eu
Gd
Cf
Tb
Dy
Ho
Er
Tm
Yb
Lu
Y
*$\alpha_{La/Ac}$ much higher at
6-10M HNO_3


Relative D_w on DGA, Branched
Atomic Number (Z)
Sc
Ac
La
Ce
Pr
Nd
Pm
Am
Sm
Cm
Eu
Gd
Cf
Tb
Dy
Ho
Er
Tm
Yb
Lu
Y

DGA/DOODA



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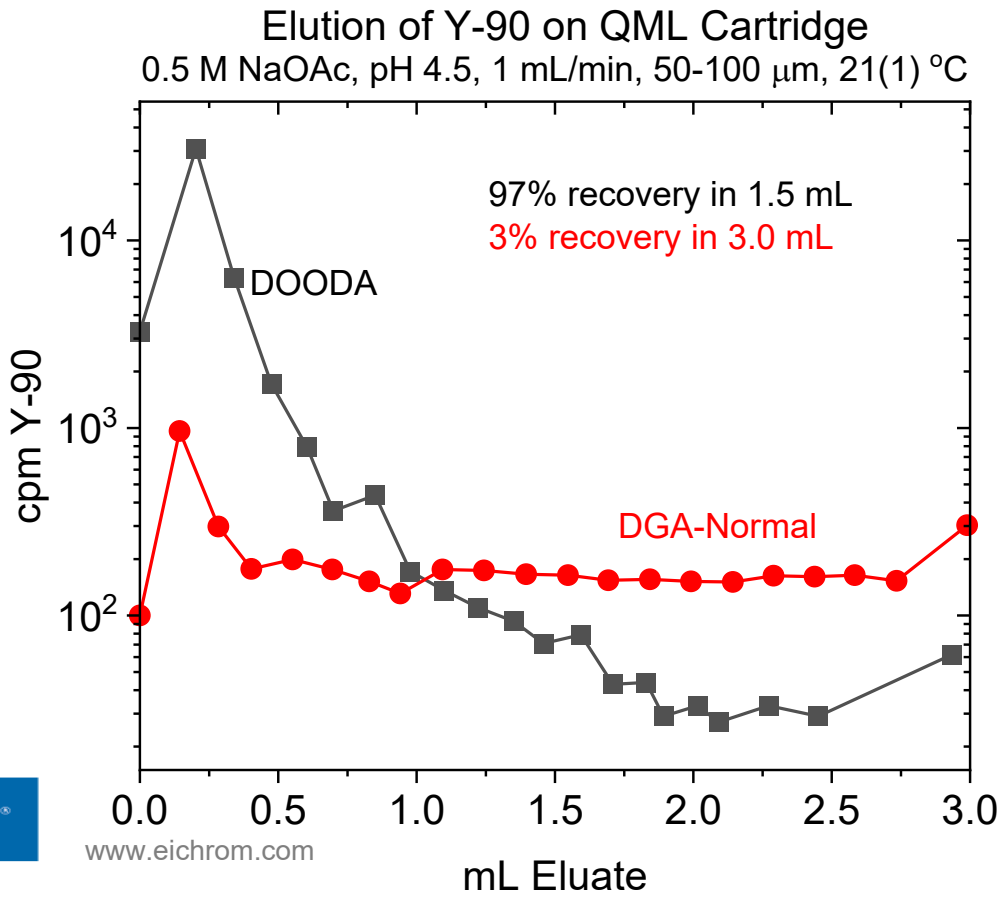
DGA/DOODA

Ac-225
>99% in 3 mL for
DGA-N, DGA-B
and DOODA



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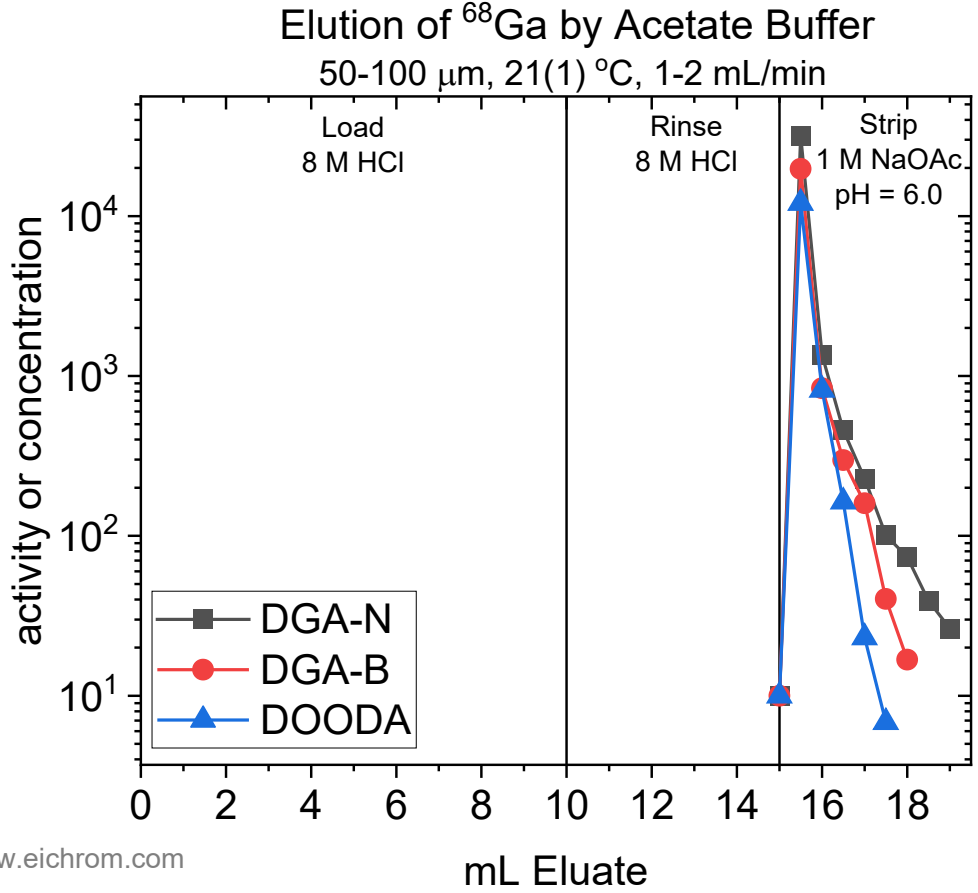
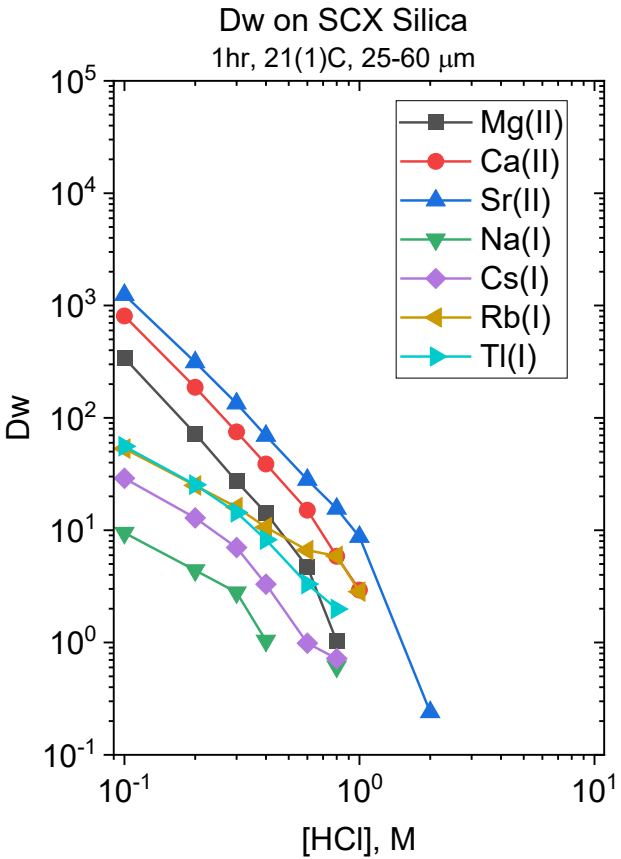
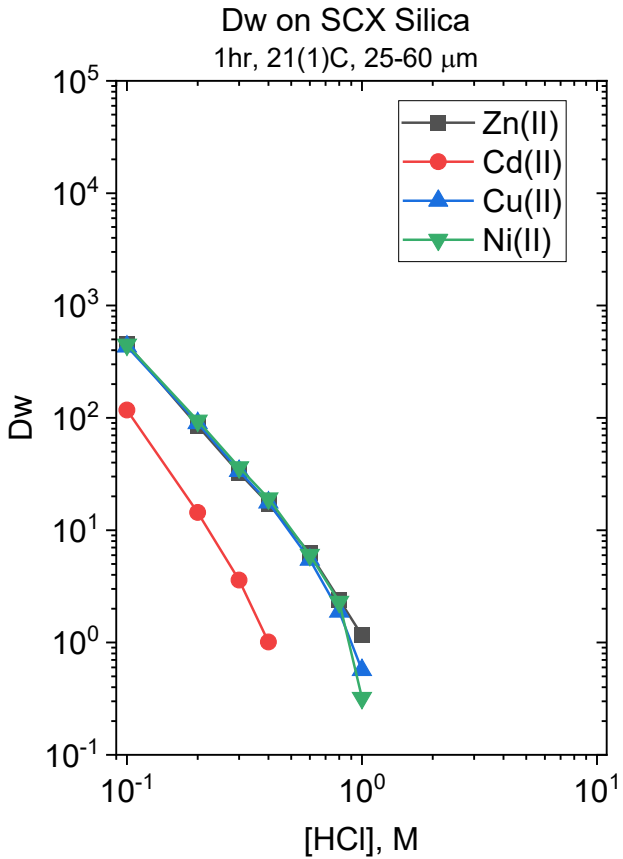
DGA/DOODA

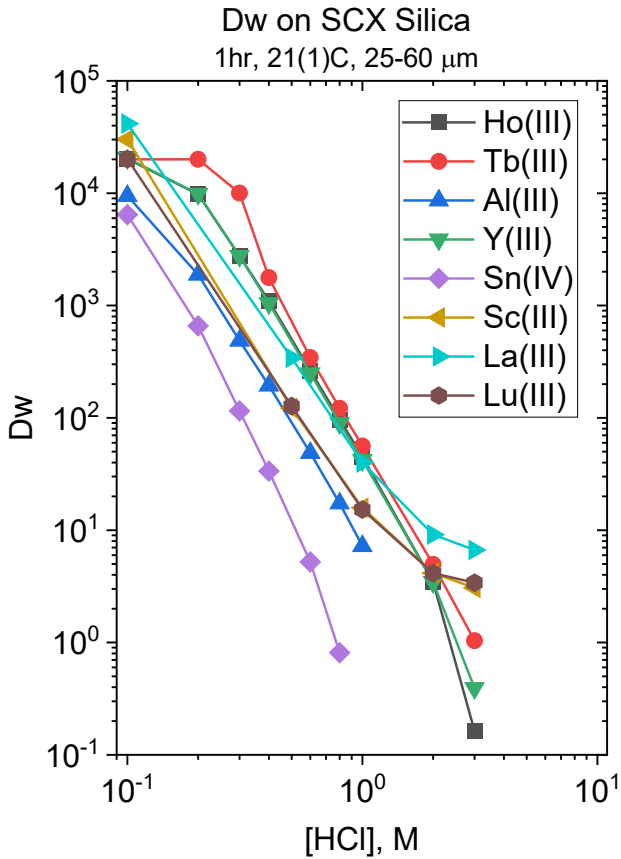
Neutral extractants can achieve transfer to buffer and concentration, but require loading 1-8M HNO3 or HCl.



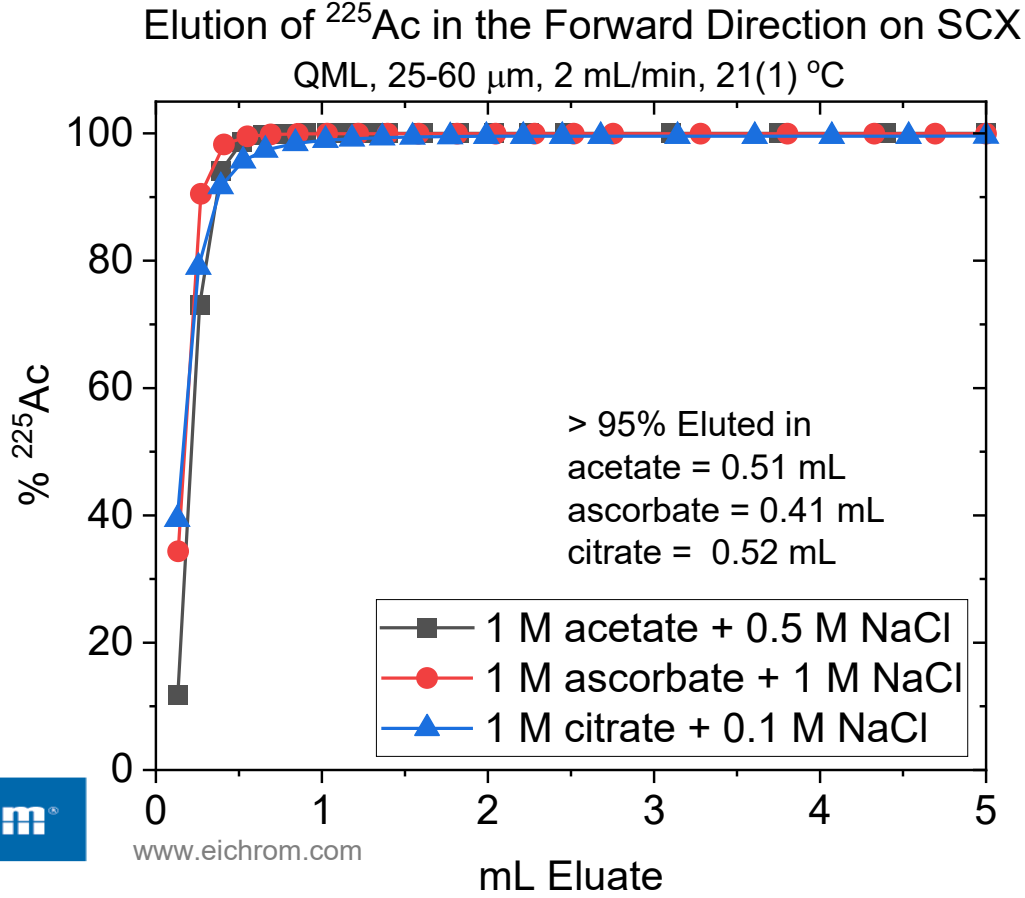
SCX


Dw on SCX Silica
1hr, 21(1)C, 25-60 μm
Dw
[HCl], M
Mg(II)
Ca(II)
Sr(II)
Na(I)
Cs(I)
Rb(I)
Tl(I)


Dw on SCX Silica
1hr, 21(1)C, 25-60 μm
Dw
[HCl], M
Zn(II)
Cd(II)
Cu(II)
Ni(II)


Dw on SCX Silica
1hr, 21(1)C, 25-60 μm
Dw
[HCl], M
Ho(III)
Tb(III)
Al(III)
Y(III)
Sn(IV)
Sc(III)
La(III)
Lu(III)

SCX



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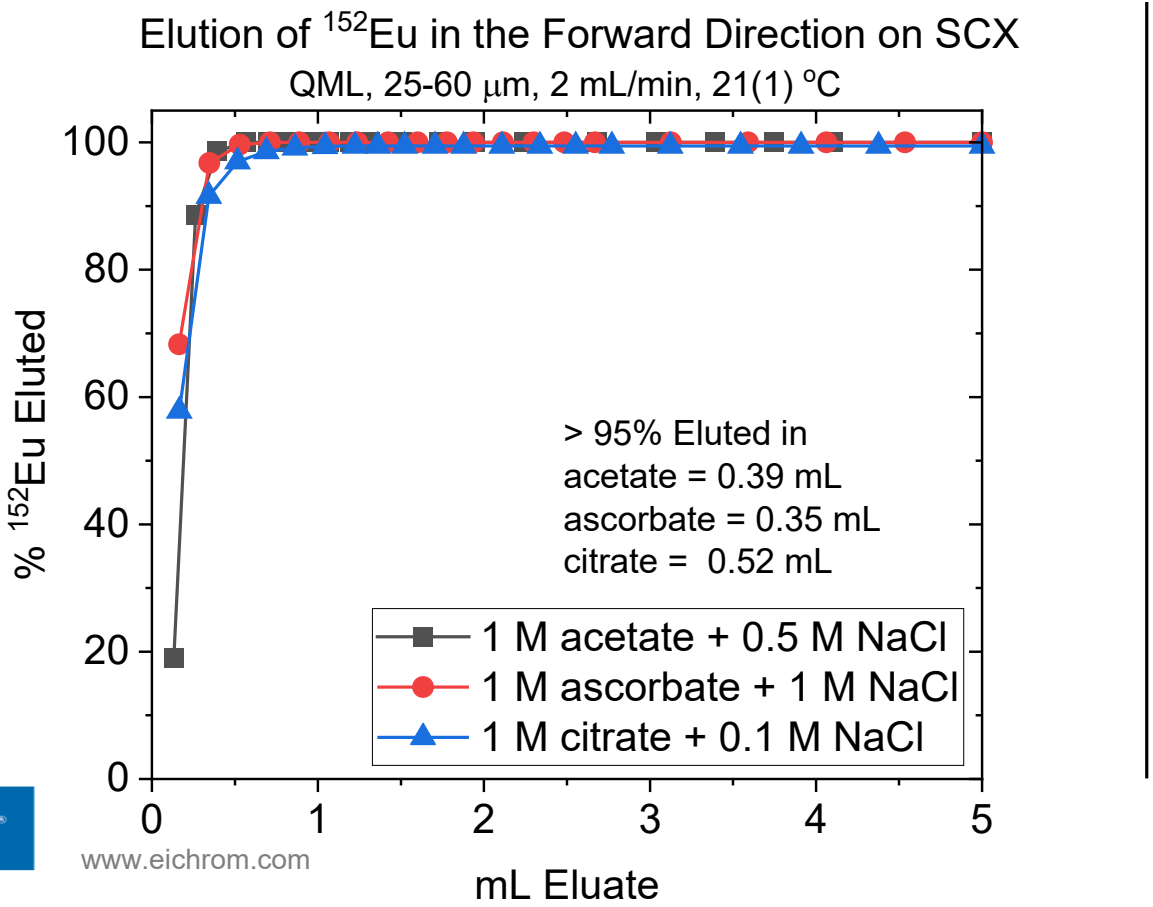
SCX



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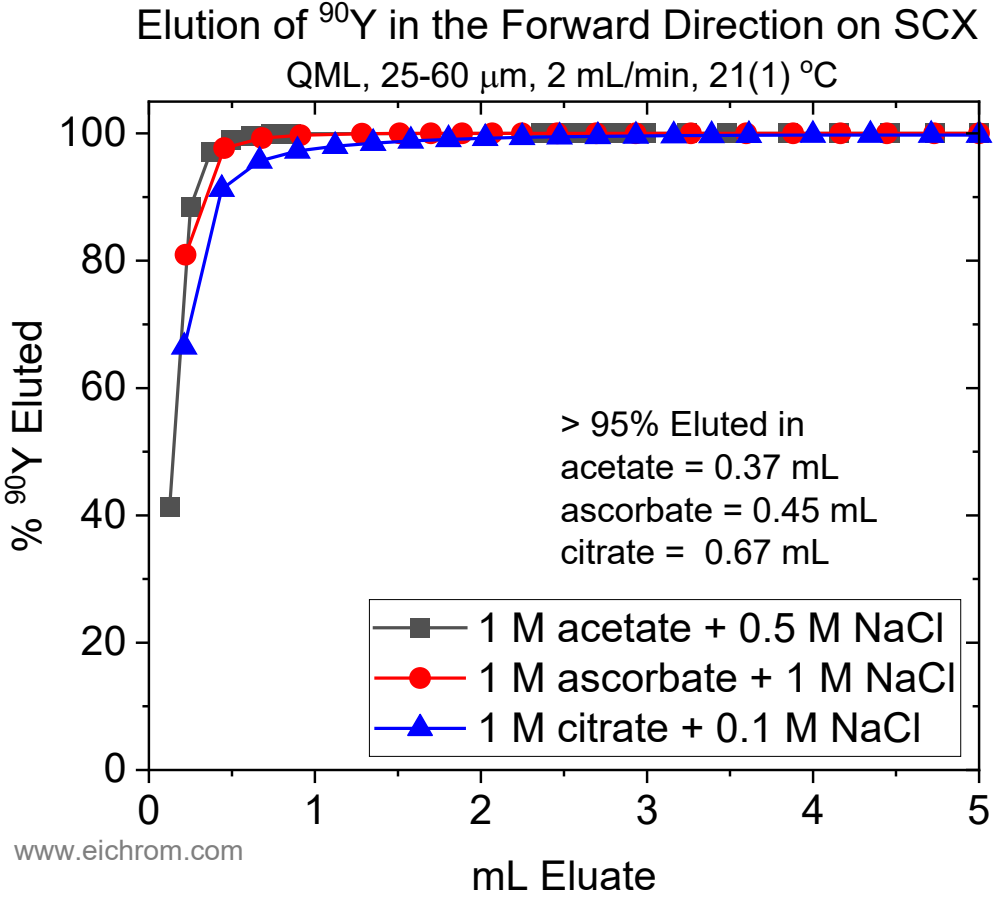
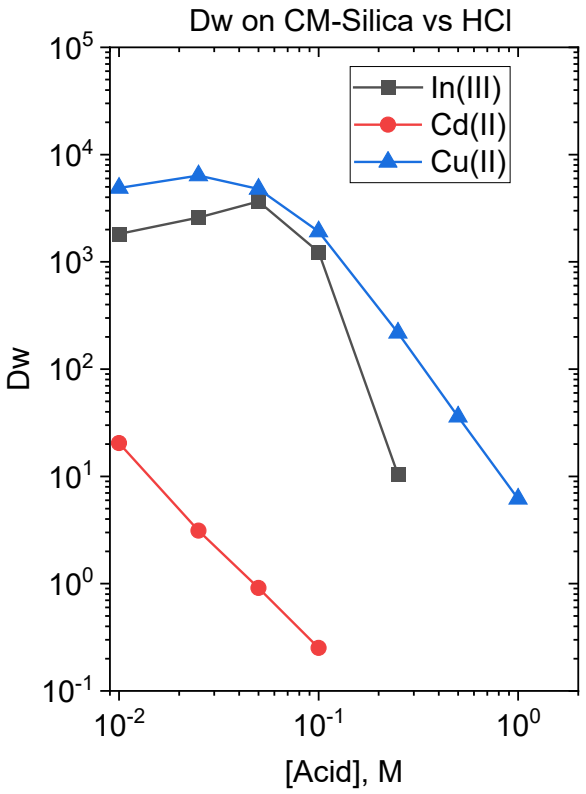
SCX

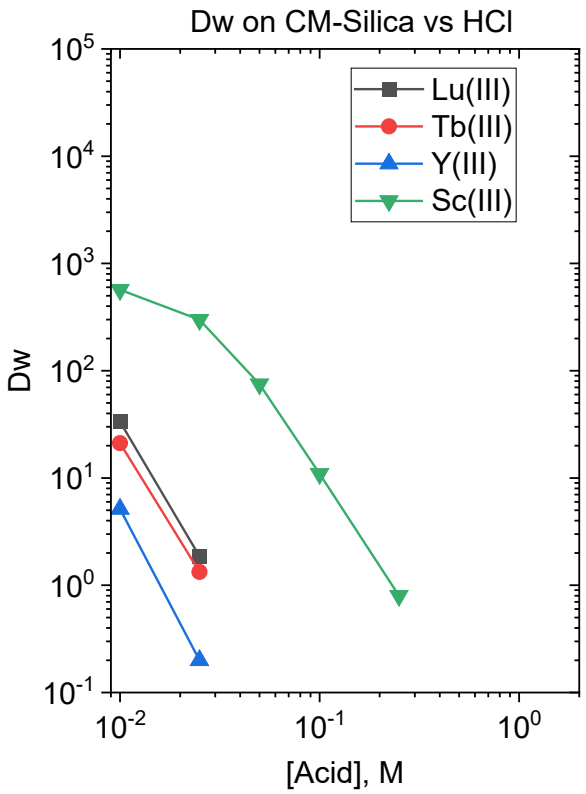
Try with ^{68}Ga and buffers.

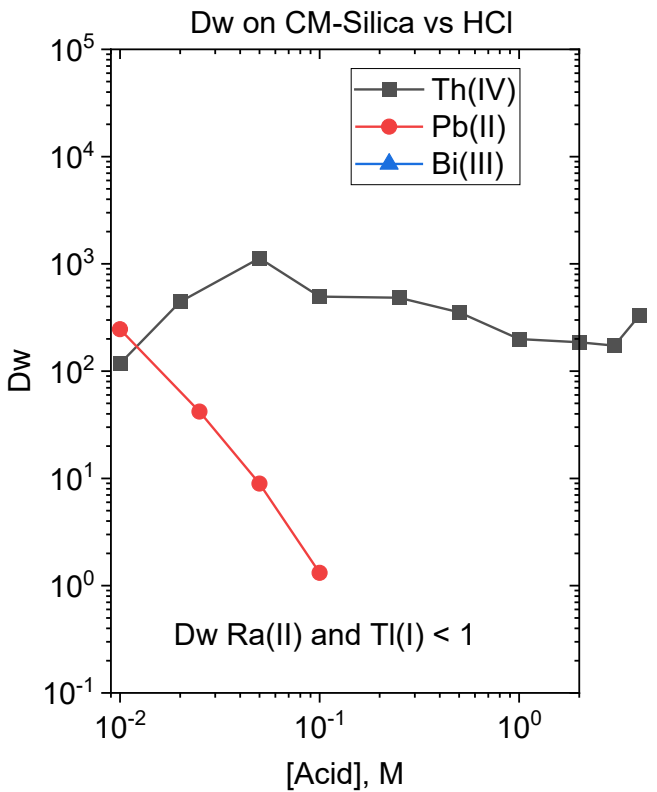
Works for ^{68}Ga loading with 0.1M HCl and recovering in 5M NaCl-0.1M HCl.



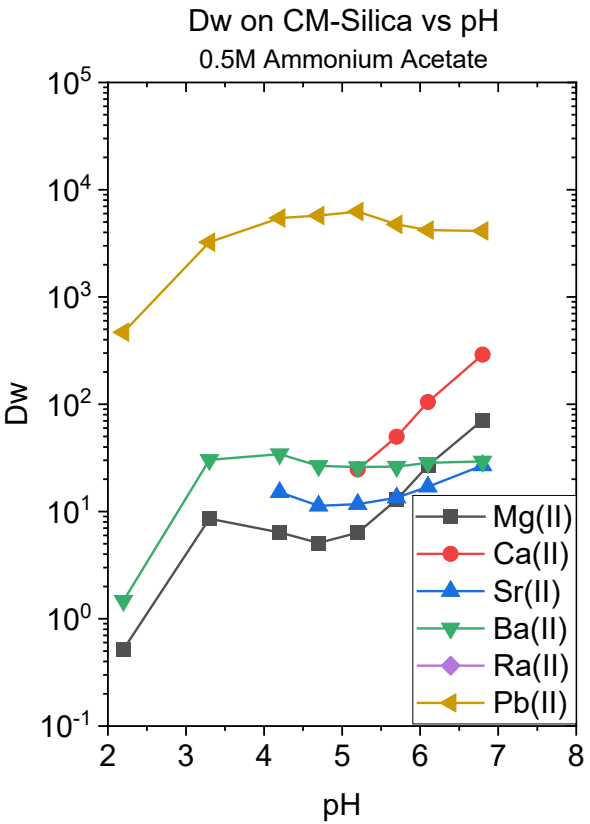
WCX (CM)

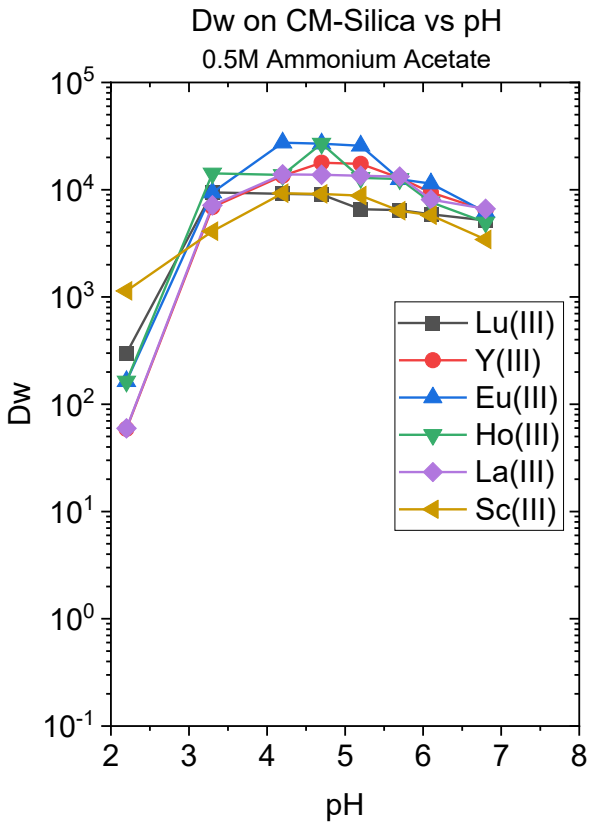

Dw on CM-Silica vs HCl
In(III)
Cd(II)
Cu(II)
Dw
[Acid], M

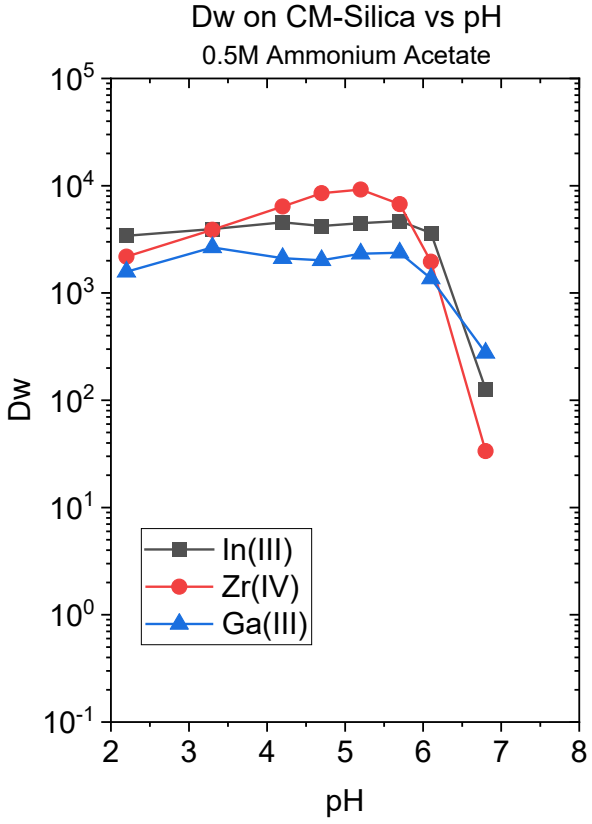

Dw on CM-Silica vs HCl
Lu(III)
Tb(III)
Y(III)
Sc(III)
Dw
[Acid], M


Dw on CM-Silica vs HCl
Th(IV)
Pb(II)
Bi(III)
Dw Ra(II) and Tl(I) < 1
Dw
[Acid], M

WCX (CM)


Dw on CM-Silica vs pH
0.5M Ammonium Acetate
Dw
pH
Mg(II)
Ca(II)
Sr(II)
Ba(II)
Ra(II)
Pb(II)


Dw on CM-Silica vs pH
0.5M Ammonium Acetate
Dw
pH
Lu(III)
Y(III)
Eu(III)
Ho(III)
La(III)
Sc(III)


Dw on CM-Silica vs pH
0.5M Ammonium Acetate
Dw
pH
In(III)
Zr(IV)
Ga(III)

Sodium acetate vs Ammonium Acetate

- Use ammonium acetate as surrogate for sodium when measuring metal ions by AES

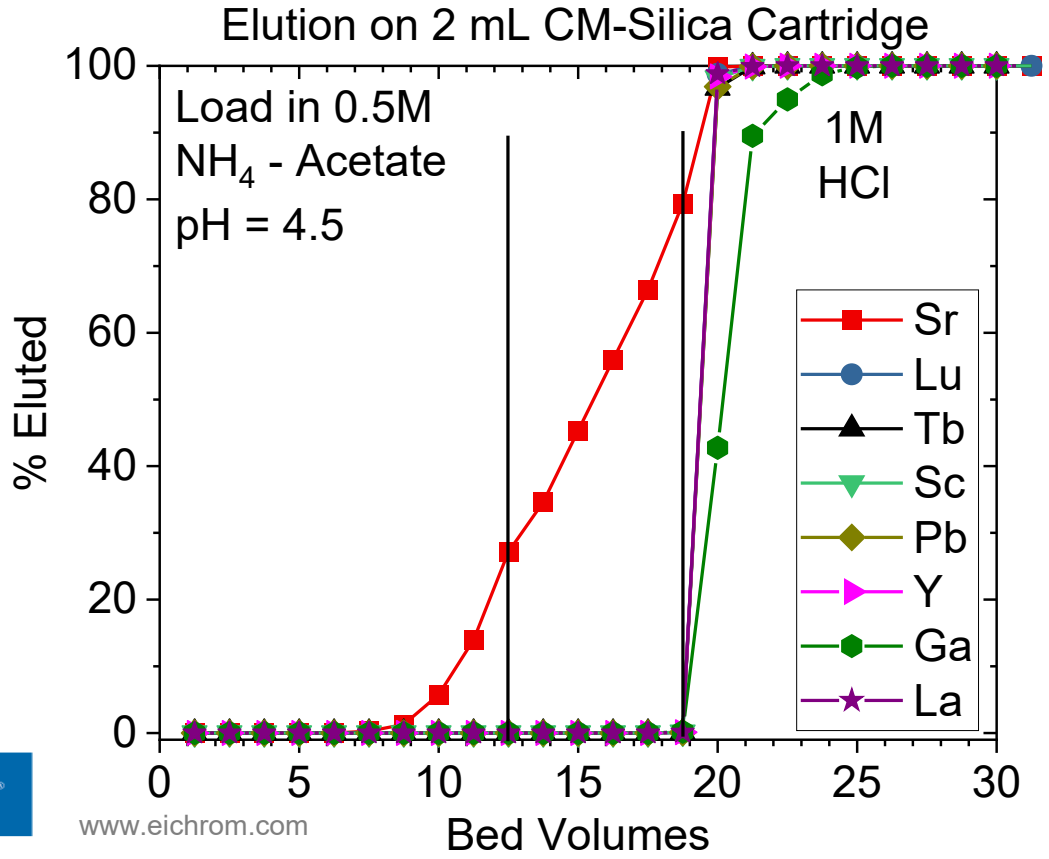
- Na^+ causes interference with many metal ions
    - ionization interference (axial, ionic lines, ionization potential)
    - intense yellow emission

- NH_4^+ is less prone to interference

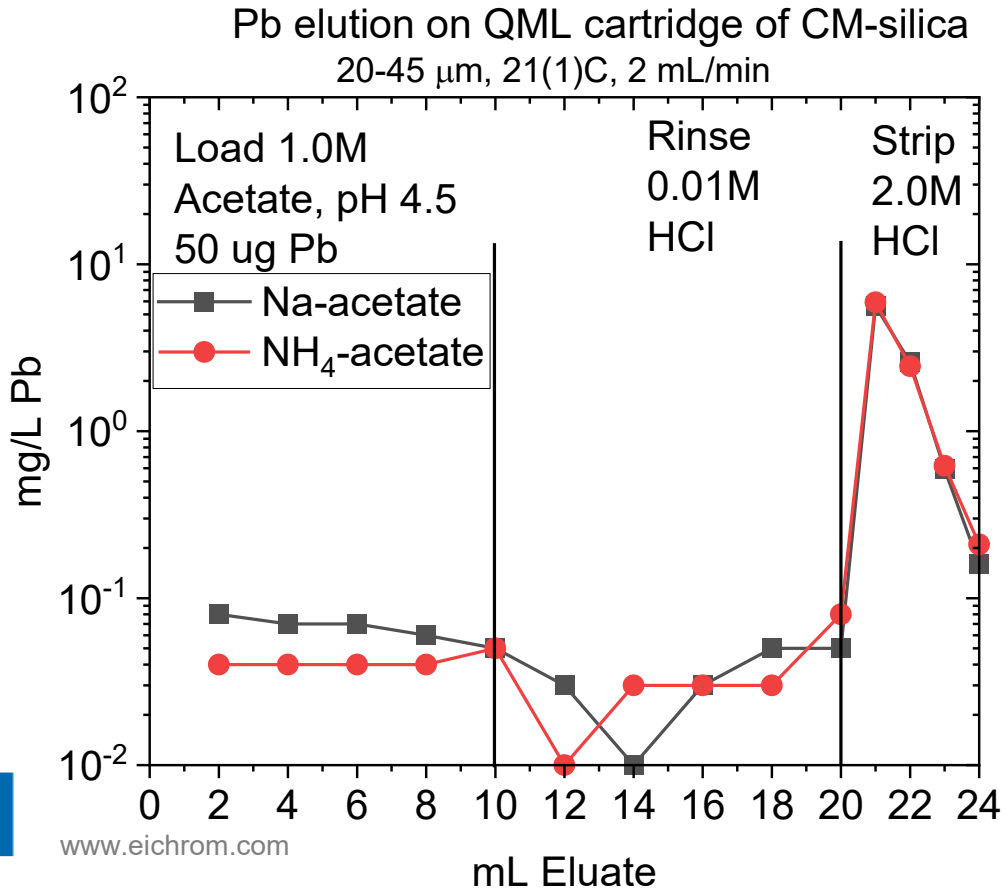
- Ammonium acetate = Spirit of Mindererus
    - weak acid + weak base
    - additional buffer capacity at high pH
    - good substitute for sodium acetate, but not perfect

WCX (CM)

50 µg each metal



WCX (CM)



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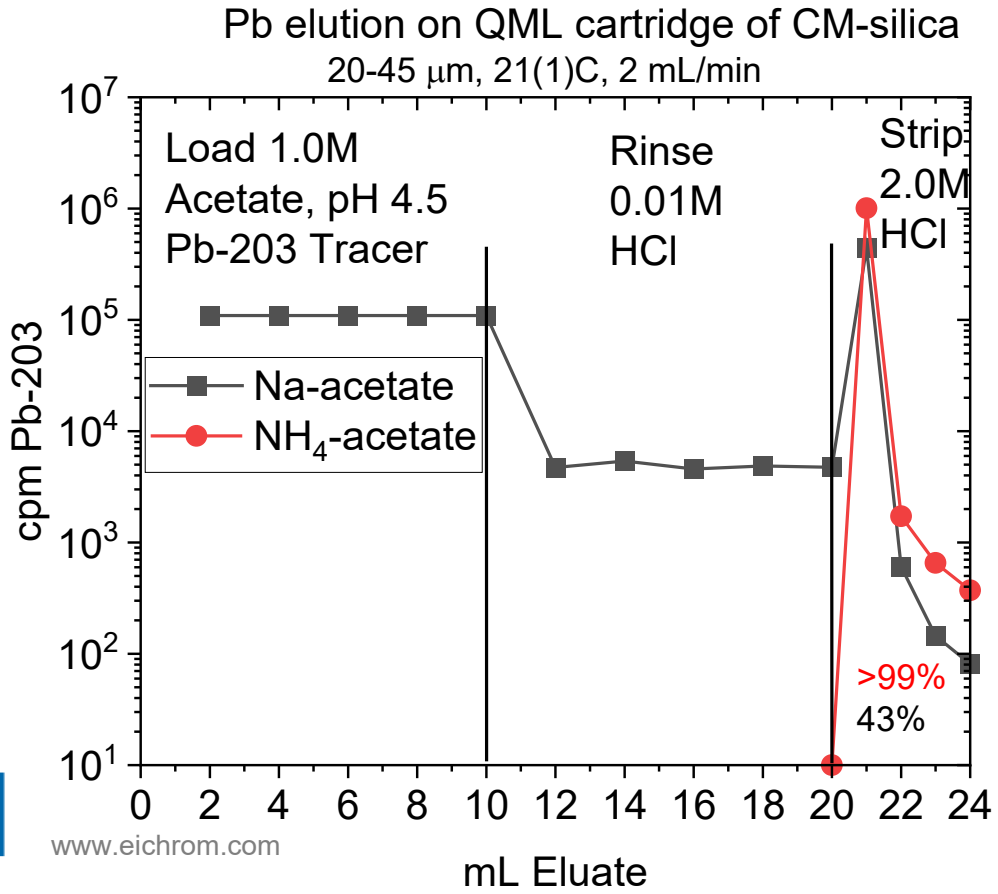
WCX (CM)



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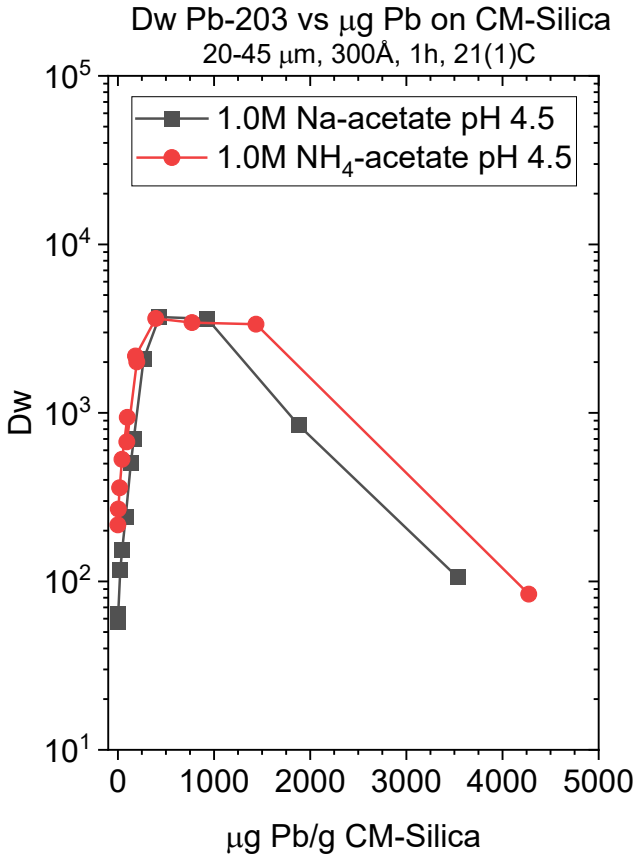
WCX (CM)

- higher Pb mass improves uptake until capacity exceeded.

- large difference between Na-acetate and NH4-acetate for tracer Pb-203.

- not seen with other systems studied, but need to evaluate more thoroughy.



Future work

- Optimize AES procedure for use in high Na matrix

- Study addition systems (tracer vs ug chemistry)

- Further work to understand interesting cases (Pb-acetate, others)

- Test CM-Silica in other high salt matrices (sea water)