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## **Title page**

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3 Title: Improving the Recovery of Ba from Sr Resin Columns Using Chelating Agents

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# 9 **Improving the Recovery of Ba from Sr Resin Columns** 10 **Using Chelating Agents**

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## 15 **Abstract**

16 Age dating of Cs-137 sources requires the accurate and precise quantification of the isotope  
17 Ba-137. In previous efforts to determine the age of Cs-137 source material, variation in  
18 Ba recovery was the largest source of uncertainty. This work investigates the use of several  
19 common chelating agents (EDTA, CDTA, DTPA, and NTA) to strip barium from Sr Resin  
20 columns quantitatively in an effort to improve the reproducibility of the separation. Batch  
21 studies showed conditions in which CDTA, EDTA, and NTA may suppress the retention  
22 of Ba on the column. Follow-up column studies showed excellent removal of barium from  
23 the column with quantitative recovery in the appropriate elution fraction. This work builds  
24 upon previous studies and establishes additional chelating agents that could be used in an  
25 age dating separation procedure.

## 26 **Keywords**

27 Age Dating, Cesium, Barium, Chelating Agents, Sr Resin, Extraction chromatography

## 28 **Introduction**

29 The fission product Cs-137 is used in a variety of commercial and medical applications in  
30 the form of sealed radioactive sources. These sources are commonly used in self-contained  
31 irradiators, radiotherapy units, density gauges, well-logging devices, and as calibration  
32 sources.[1] It finds utility in many of these applications due to its favorable half-life (30.07  
33 yrs) and gamma emission energy (661.7 keV). Unfortunately, these same characteristics  
34 also make it attractive for use in a radiological dispersal device (RDD), or “dirty bomb.”

35 Current events have driven a concerted effort to develop methods that can be used to  
36 identify the source of Cs-137 if the material is found outside of regulatory control. Because  
37 different source manufacturers have produced Cs-137 at different times through the years,  
38 the age of any interdicted material can help narrow down its point of origin. The “age” of  
39 a Cs-137 source, corresponding to its last purification date, can be determined using the  
40 ratio of Cs-137 to its daughter, Ba-137, using the following equation:

$$41 \quad t = \ln\left(1 + \frac{N_D(t)}{N_P(t)}\right) \times \frac{1}{\lambda}$$

42 where  $t$  is the elapsed time since purification,  $N_D(t)$  is the atom amount of the daughter  
43 nuclide at present time,  $N_P(t)$  is the atom amount of parent nuclide at present time, and  $\lambda$   
44 is the decay constant of the parent nuclide. Although gamma spectrometry can quantify  
45 Cs-137, Ba-137 is stable and therefore mass spectrometry is required to quantify Ba-137.  
46 Unfortunately, Cs-137 causes isobaric interference on Ba-137 mass spectrometric  
47 measurements and the two must therefore be separated prior to barium analysis.

48 Previous work by *Sommers et al.* and *Steeb et al.* used extraction chromatography with Sr  
49 Resin to separate the parent and daughter with success.[2,3] A subsequent attempt by *Steeb*  
50 *et al.* to apply the same procedure to a cesium ceramic matrix determined that the largest  
51 source of uncertainty in the procedure was the recovery of Ba from the column.[4] The  
52 variability in Ba recovery accounted for 85% of the variability in the determined age, which  
53 was  $\leq 3\%$  ( $k=2$ ) with respect to Cs (pure Ba recovery numbers were not reported). This  
54 corresponds to  $\pm 2$  years for a 60 year old source. In order to improve Ba recovery, *Steeb*  
55 *et al.* added the chelating agent ethylenediaminetetraacetic acid (EDTA) to the elution  
56 matrix to effectively back-extract the Ba into the aqueous phase, similarly to previous anion

57 exchange procedures.[5,6,4] Doing this improved recovery and repeatability, thereby  
58 decreasing the uncertainty in the associated age calculations to  $\leq 1.7\%$  ( $k=2$ ).[4]  
59 Subsequently, *Surrao* investigated the use of EDTA and diethylenetriaminepentaacetic  
60 acid (DTPA) and found that adjustment to pH 9 further increased the Ba recovery and  
61 reproducibility.[7] In that work, Ba recoveries of  $100.9 \pm 2.2\%$  were achieved. This work  
62 expands the suite of chelating agents examined and takes a closer look at the effects of pH  
63 on Ba recovery.

## 64 **Experimental**

### 65 ***Materials***

66 Samples and standards for the separation improvement experiments were prepared using  
67 ultra-high purity solvents and reagents to avoid unintentional contamination. A variety of  
68 solutions were prepared by dissolving solid  $\text{KNO}_3$ ,  $\text{NaNO}_3$ , ethylenediaminetetraacetic  
69 acid (EDTA), cyclohexyldiaminetetraacetic acid (CDTA), diethylenetriaminepentaacetic  
70 acid (DTPA), or nitrilotriacetic acid (NTA) in de-ionized water as elution reagents.  
71 Potassium and sodium nitrate salts were Puratronic grade, obtained from Alfa Aesar  
72 (Haverhill, MA), while EDTA, NTA, and CDTA were 99%, obtained from Acros Organics  
73 (New Jersey, NJ), and DTPA was  $\geq 99.0\%$ , obtained from Fluka (Morris Plains, NJ). After  
74 dissolution of the solids, the pH was adjusted by adding solid NaOH (TraceSELECT,  
75 Fluka) until the desired level was reached, after which the solutions were diluted to 50 ml.  
76 Standards and spike solutions were prepared using certified natural elemental standards of  
77 Ba ( $998 \pm 5 \mu\text{g/ml}$ ; density =  $1.000 \text{ g/ml @ } 20^\circ\text{C}$ ) and In ( $997 \pm 3 \mu\text{g/ml}$ ; density =  $1.011$   
78  $\text{g/ml @ } 20^\circ\text{C}$ ) from Inorganic Ventures (Christiansburg, VA). By using the certified  
79 density of each elemental standard, it was possible to determine the concentrations in terms  
80 of  $\mu\text{g/g}$ . Doing so allowed for the use of mass-based measurements and calculations to  
81 minimize error. All solutions were prepared with deionized water (DIW) from a Thermo  
82 Scientific™ Barnstead™ MicroPure™ water purification system.

83 The Sr Resin used in both batch and column studies contains 1.0 M 4,4'(5')-di-t-  
84 butylcyclohexano 18-crown-6 in 1-octanol as the extractant, and was manufactured by

85 Eichrom Technologies (Lisle, IL). All batch and column studies were carried out using  
86 their 50 – 100  $\mu\text{m}$  particle size resin. All resin products were used as received from the  
87 manufacturer.

## 88 ***Procedure***

89 Both batch studies and column studies were employed to optimize the chelating agent back-  
90 extraction conditions.

91 *Batch Studies:* Batch studies were carried out as described by *McLain, et al.*[8-10] Briefly,  
92 Sr Resin was weighed into 2 ml centrifuge tubes and preconditioned with chelating agent  
93 solution for 1 hour, then spiked with Ba solution and mixed for an additional hour to ensure  
94 equilibration. At this point, the aqueous phase was separated from the resin using a 0.45  
95  $\mu\text{m}$  PTFE syringe filter and an aliquot of the aqueous phase was prepared for ICP-MS  
96 analysis. Results are displayed as  $k'$  values, which indicate the number of free column  
97 volumes (FCVs) required to reach the peak maximum of the elution profile when eluting  
98 from a chromatography column. Mathematical operations to transform batch  
99 concentration data to  $k'$  can be found in *Chiarizia et al.*[11]

100 Initially, chelating agent solutions were prepared by adjusting DIW to the desired pH, then  
101 dissolving the chelating agent. It was discovered, however, that the chelating agents were  
102 buffering the solutions to much different pH levels, especially when present in high  
103 concentrations. The preparation procedure was therefore switched to adding the chelating  
104 agents to 30-40 ml DIW, then adding solid NaOH until the pH stabilized at the desired  
105 level, and finally diluting to 50 ml. The most concentrated solutions were prepared first,  
106 and all subsequent batch study solutions were prepared via serial dilution from these. Batch  
107 studies were carried out at pH 7, 9, and 11 with varying concentrations of all four chelating  
108 agents. Batch studies were also performed with varying concentrations of  $\text{NaNO}_3$  and  
109  $\text{KNO}_3$  without pH adjustment to test the hypothesis laid out by *Surrao et al.* that  $\text{Na}^+$  and  
110  $\text{K}^+$  could effectively compete with the  $\text{Ba}^{2+}$  enough to improve Ba recovery.[7]  
111 Concentrations of the salts analyzed were 0.01, 0.05, 0.1, 0.5, and 1 M. Finally, batch  
112 studies were carried out at pH 7.6, 8.7, and 10.8 (adjusted with NaOH) without any

113 chelating agent added. A summary of the batch studies carried out in this work is found in  
 114 Table 1.

115 **Table 1** Summary of all batch studies carried out as part of this work. N/A denotes the field  
 116 is not applicable. N/C denotes the parameter was not controlled.

Chelating/Competing Agent	pH	Concentrations analyzed (M)				
EDTA	7	0.01	0.025	0.05	0.1	0.25
	9					
	11					
CDTA	7	0.01	0.025	0.05	0.1	0.25
	9					
	11					
DTPA	7	0.01	0.025	0.05	0.1	0.25
	9					
	11					
NTA	7	0.01	0.025	0.05	0.1	0.25
	9					
	11					
None	7.6	N/A	N/A	N/A	N/A	N/A
	8.7					
	10.8					
KNO <sub>3</sub>	N/C	0.01	0.05	0.1	0.05	1
NaNO <sub>3</sub>	N/C	0.01	0.05	0.1	0.05	1

117 *Column Studies:* Column studies were performed using 1-ml polypropylene Bio-Scale™  
 118 Mini Cartridges (Bio-Rad Laboratories) made of polypropylene with a height of 38 mm  
 119 and an inner diameter of 6 mm, and were slurry packed with Sr Resin in DIW. Each column  
 120 was slurry packed by mixing an amount of resin with DIW, adding a portion to the column,  
 121 pulling the solution through the column with a peristaltic pump until the liquid level in the  
 122 column was just higher than the resin, and then repeating the process until the entire 1-ml  
 123 column was filled with resin. Once packed, the column was closed and all separations  
 124 were run with solution pumped in the reverse direction. All separations were run at room  
 125 temperature at a flow rate of 0.5 ml min<sup>-1</sup>. In order to compare the recoveries achieved for  
 126 the different chelating agents, three column studies were performed using 0.05M solutions  
 127 of NTA, DTPA, and CDTA at pH 9. These conditions were selected based on the results  
 128 of the batch studies. An additional separation was performed using 0.05 M HNO<sub>3</sub> to

129 establish a baseline using conditions recommended by the manufacturer. The EDTA  
130 system was omitted, as it was thoroughly investigated previously.[4,7]

131 Column studies were carried out as described by *Steeb, et al.*[4] Briefly, each column was  
132 conditioned with 5 ml of 3 M HNO<sub>3</sub> at a flow rate of 1 ml min<sup>-1</sup>. The column was loaded  
133 with 0.5 ml of 500 ppb Ba solution in 3 M HNO<sub>3</sub>, and then was rinsed with 3 ml of the  
134 same solution used to condition the column. In a real separation, the rinse fractions would  
135 contain any Cs that was initially in solution with the Ba. Previously published literature  
136 indicates all Cs would be eluted from the column in the first 1-4 free column volumes (0.6-  
137 2.4 ml).[4,12,2,13] Following this, a 0.5 ml plug of DIW was added to the column to  
138 prevent any chelating agent precipitation, and finally 5.5 ml of chelating agent solution was  
139 used to remove Ba from the column. Once the recovery was determined, high-fidelity  
140 measurements of the top two performers were carried out. For these trials, three columns  
141 were packed for each chelating agent, and only two fractions (rinse and Ba elution) were  
142 collected for each.

143 For all column studies, fractions were collected in labeled 10 ml PTFE beakers containing  
144 250 ng of In as an internal standard. Following collection, 1 ml of 3 M HNO<sub>3</sub> was added  
145 to each fraction and refluxed for 1 hr at 135 °C to destroy as much of the organic content  
146 as possible, then evaporated to dryness. This was repeated, and then the residues were  
147 reconstituted in 2% HNO<sub>3</sub>, transferred to a sterile 50 ml centrifuge tube, diluted to 25 g  
148 using 2% HNO<sub>3</sub>, and sent for ICP-MS analysis. In order to monitor recovery, “spike”  
149 samples were prepared using the same solution that was loaded on the column. In order to  
150 account for Ba found in the reagents, blanks were prepared by processing samples of the  
151 reagents in the same manner as the rest of the samples.

## 152 ***Measurements***

153 Elemental concentrations of In and Ba in each collected fraction were measured with a  
154 NexION 2000 Inductively Coupled Plasma Mass Spectrometer (ICP-MS) (PerkinElmer,  
155 Shelton, CT). This system was equipped with a PerkinElmer S-10 autosampler and a  
156 discrete dynode electron multiplier detector. A PFA Barrel spray chamber and a Type C  
157 PFA ST3 nebulizer form the sample introduction-system. All ICP-MS samples and

158 calibration standards were prepared by mass and ratio-based data reduction and calibration  
159 formulas were employed to reduce systematic error, as recommended by *Salit et al.*[14-16]

160 The samples were analyzed in the pulse-counting mode with a dead-time correction of 62  
161 ns. The following parameters were utilized for each analysis run: a 100-ms dwell time, 15  
162 sweeps per replicate, and 5 replicates per sample. All data were acquired in the quantitative  
163 analysis mode and were divided by the measured In intensity to account for instrument  
164 noise and drift. Based on the standard deviation of the analyzed samples, minimum and  
165 maximum  $k'$  values able to be determined were 0.13 and 12,500, respectively.

## 166 **Results and discussion**

### 167 *Batch Studies*

168 Results of the different batch studies were somewhat predictable in that under all conditions  
169 an increase in chelating agent concentration resulted in a lower  $k'$  value. This is  
170 demonstrated at pH 7, pH 9, and pH 11 in Fig. 1, Fig. 2, and Fig. 3, respectively.  
171 Interestingly, increasing pH did not uniformly improve the performance of the chelating  
172 agents. NTA and EDTA perform much better at pH 9 and pH 11 than at pH 7. CDTA has  
173 a similar performance in each matrix, with a mildly better performance at pH 7. In contrast,  
174 it appears that DTPA performs effectively the same at pH 7 and pH 11, with a marginal  
175 increase in  $k'$  at pH 9. This variety of performance characteristics is likely due to the  
176 different levels of deprotonation for the various chelating agents. For comparison, the  
177 various pKa values have been compiled in Table 2, along with the stability constants of the  
178 chelating agents with Ba.[17-21] Both EDTA and NTA have a significant concentration  
179 of completely deprotonated molecules by pH 8, while DTPA only achieves similar levels  
180 at pH 9 and CDTA at pH 10. The variety of deprotonation constants also made it difficult  
181 to adjust the pH of solutions exactly. As a result, all pH levels are approximate ( $\pm 0.5$  of  
182 what is cited). It is possible that this also had some effect on the observed retention of Ba.

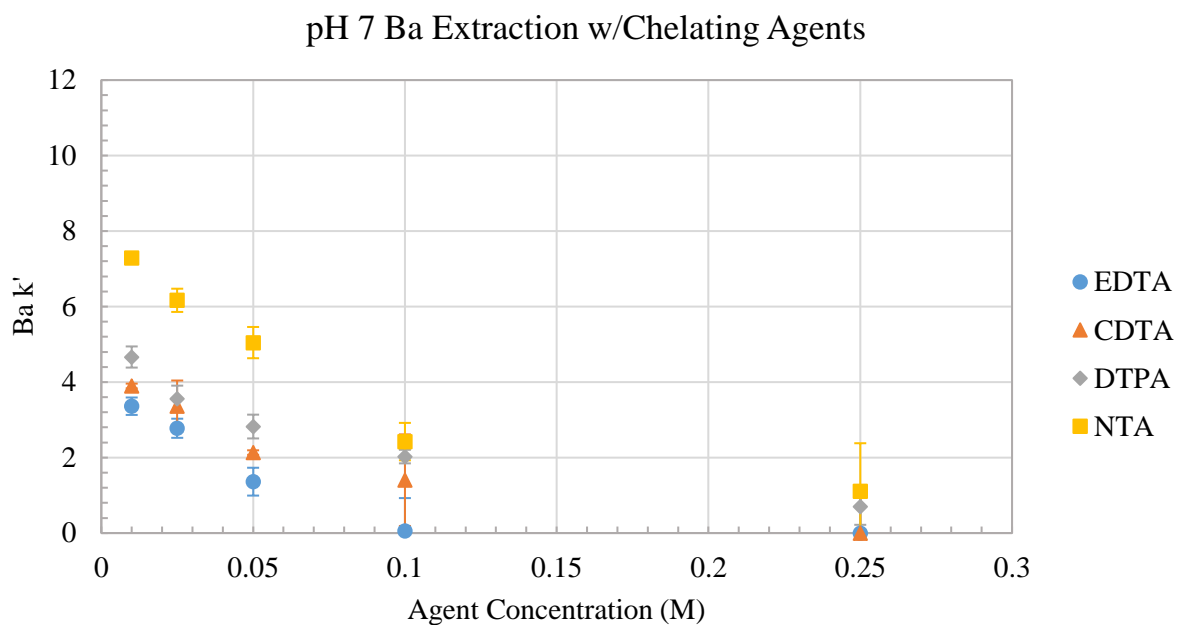
183 At the highest concentrations of chelating agent, it was observed that material would  
184 precipitate out of solution over time after solutions were separated from the resin and



185 prepared for ICP-MS analysis. This is likely due to the chelating agents being exposed to  
 186 acid solutions, in which they are only sparingly soluble. This precipitation was only  
 187 observed when the solutions were being disposed of, however, so it is not believed that it  
 188 had any impact on the analysis.

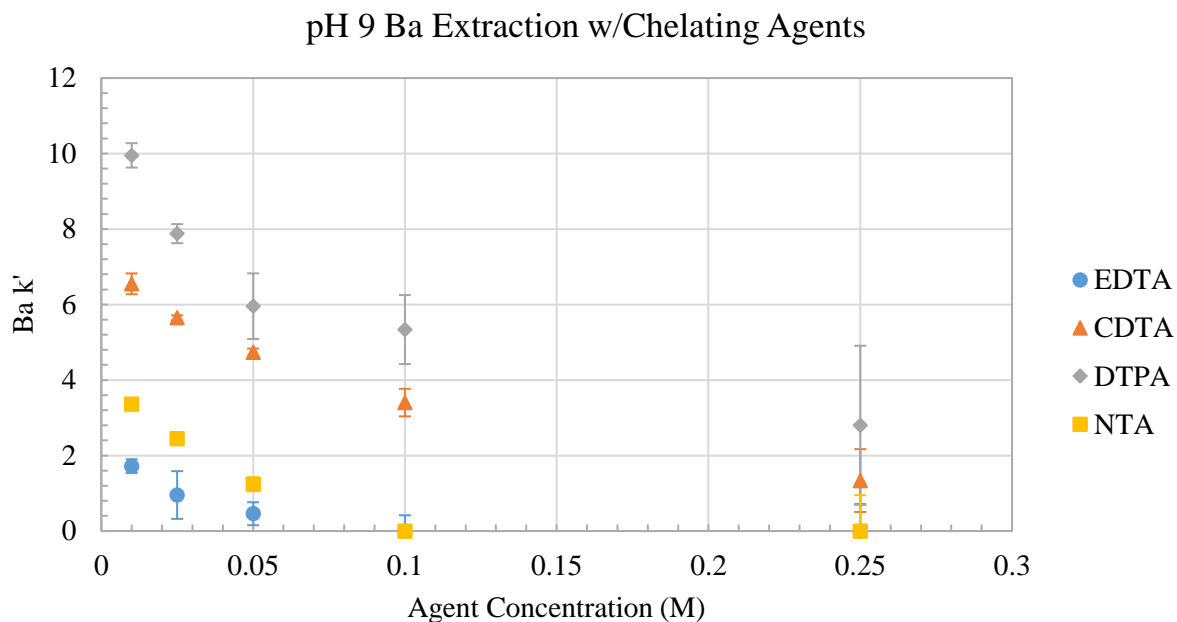
189 **Table 2** The various pKa values for the selected chelating agents, along with the stability  
 190 constant of each with Ba. [17-21]

Compound	pKa1	pKa2	pKa3	pKa4	pKa5	pKf - Ba
NTA	1.89	2.49	9.73			4.8
EDTA	1.99	2.76	6.16	10.26		7.9
CDTA	2.43	3.52	6.12	11.7		8.0
DTPA	2.14	2.38	4.26	8.6	10.53	8.6



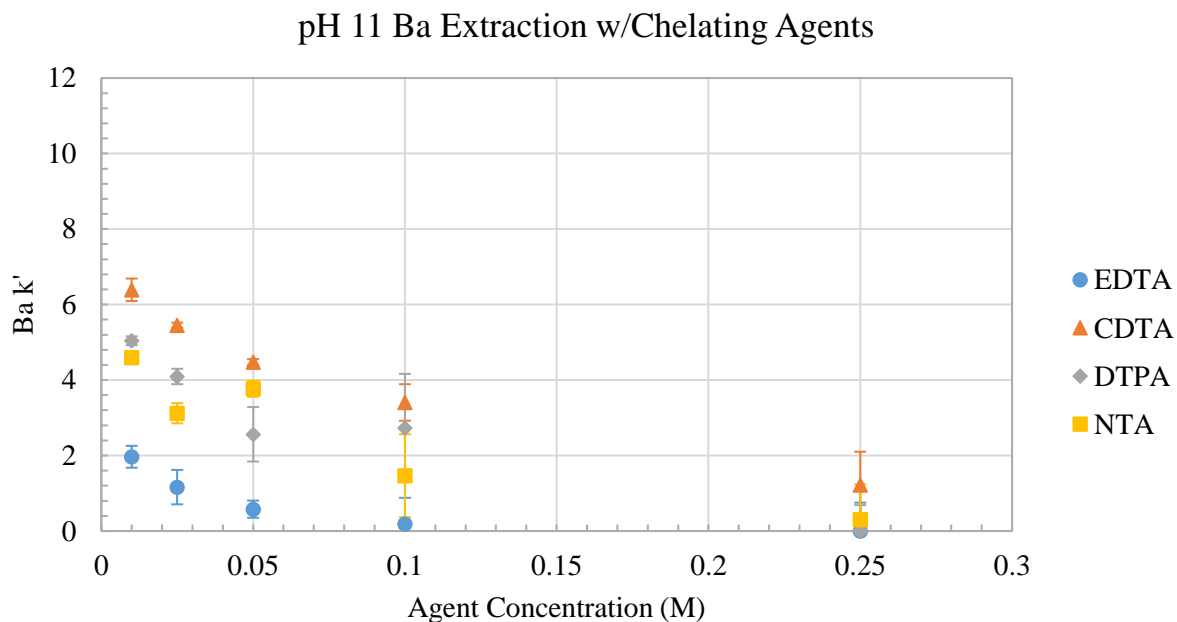
191

192 **Fig. 1** Batch study results for the various chelating agent concentrations when adjusted to  
 193 pH 7 with NaOH. Error bars are  $1\sigma$



194

195 **Fig. 2** Batch study results for the various chelating agent concentrations when adjusted to  
196 pH 9 with NaOH. Error bars are  $1\sigma$



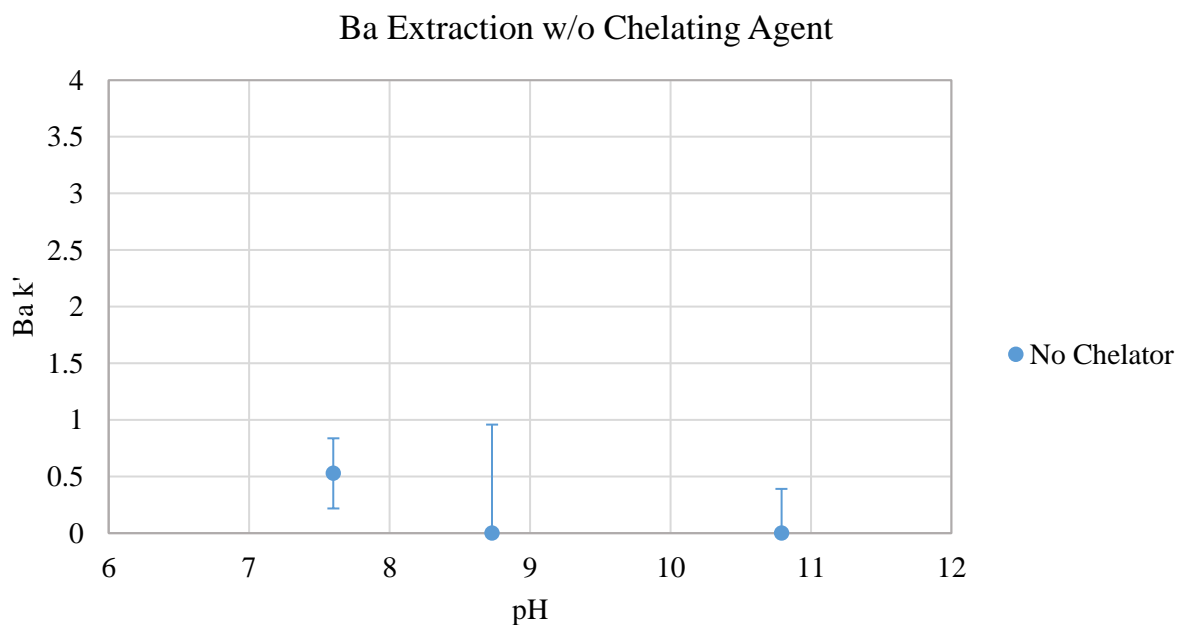
197

198 **Fig. 3** Batch study results for the various chelating agents concentrations when adjusted  
199 to pH 11 with NaOH. Error bars are  $1\sigma$

200 In any event, EDTA and NTA exhibit their best performance (lowest Ba k') at pH 9 while  
201 DTPA and CDTA exhibit their best performance at pH 7. The overall best performance for  
202 all chelators and pH values tested was for EDTA at pH 9.

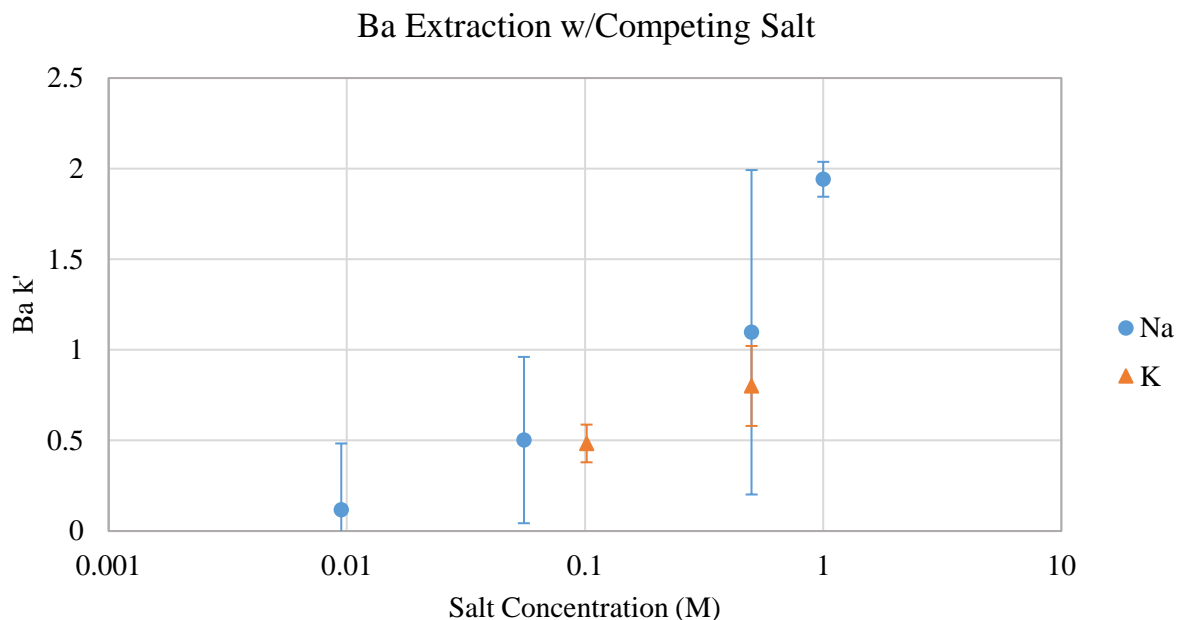
203 A batch study was also performed where the solutions were adjusted to pH 7.6, 8.7, and  
204 10.8 but no chelating agent was added. Interestingly, the retention was extremely low for  
205 all of these systems, indicating that it may be possible to elute the Ba under slightly basic  
206 ( $7 \leq \text{pH} \leq 11$ ) conditions (see Fig. 4). Though  $\text{Ba}(\text{HO})_2$  is generally considered "slightly  
207 soluble," with a  $K_{\text{sp}}$  of  $2.55 \times 10^{-4}$  even 10  $\mu\text{g}$  of Ba will be completely miscible at all three  
208 pH levels investigated.[22]

209 Finally, batch studies were conducted with varying amounts of  $\text{NaNO}_3$  and  $\text{KNO}_3$  added  
210 to the systems to test the hypothesis laid out by *Surrao*. [7] Namely, that the  $\text{Na}^+$  and  $\text{K}^+$   
211 cations would compete with  $\text{Ba}^{2+}$  on the resin enough to increase Ba recovery. The results  
212 of these batch studies are found in Fig. 5.



213

214 **Fig. 4** Batch study results when solution is adjusted to a given pH with NaOH, but no  
215 chelating agent is added. Error bars are  $1\sigma$



216

217 **Fig. 5** Batch study results when competing salt solutions of  $\text{NaNO}_3$  and  $\text{KNO}_3$  are added.  
 218 Error bars are  $1\sigma$

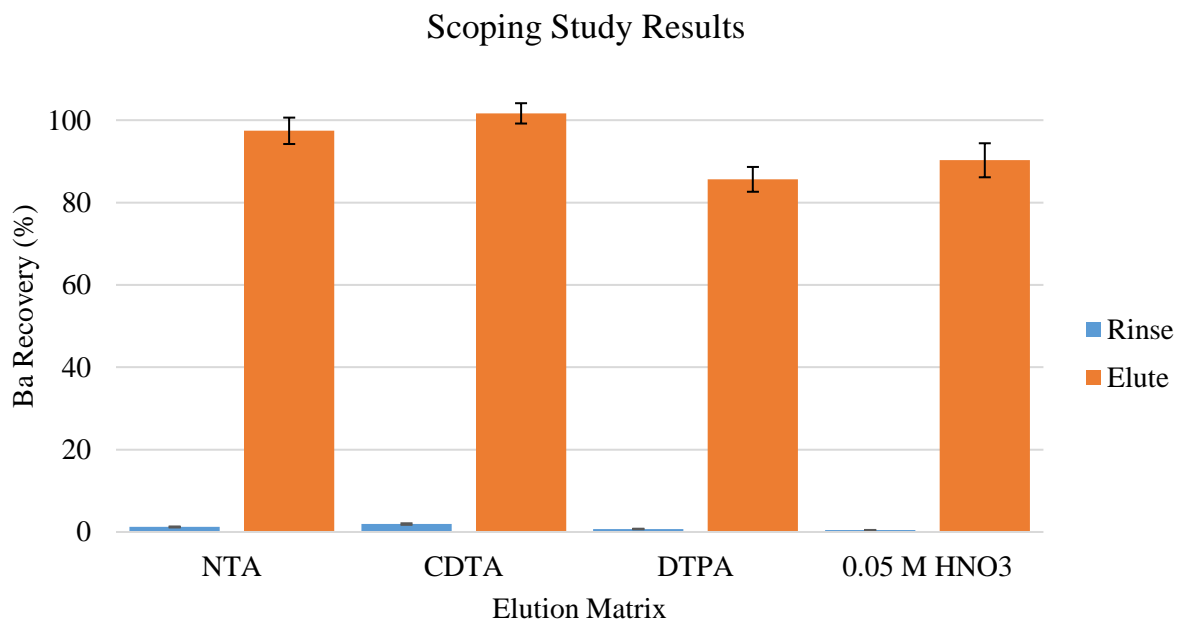
219 While it appears that the retention of Ba increases as Na and K are added to the system, it  
 220 is possible this is from the additional nitrate that was also added. As free nitrate is added,  
 221 the probability of Ba combining to make the neutral  $\text{Ba}(\text{NO}_3)_2$  species increases, which  
 222 correspondingly increases the probability of Ba being extracted since the resin has been  
 223 shown to extract the neutral alkali earth metals.[23] It should also be noted that both  
 224 sodium and potassium nitrate salts were tested at 0.01, 0.05, 0.1, 0.5, and 1 M  
 225 concentrations. Points that are missing from Fig. 5 were below the limit of detection ( $k' =$   
 226 0.13). It is suspected that the reason for this is not that the Ba is being extracted so little,  
 227 but that there is a significant amount of Ba present in the  $\text{KNO}_3$  used in the study, even  
 228 though it was a high-purity reagent (99.997% metals basis). Because the increase in  $k'$  over  
 229 the course of the study was somewhat smaller for the potassium salt than the sodium salt,  
 230 indicating some possible competition, this system was also included in the set of high-  
 231 fidelity column studies.

232 Based on the batch studies performed, additional experiments with CDTA, DTPA, and  
 233 NTA at 0.05 M. This concentration was identified as optimal in that it minimized the  
 234 retention of Ba on the column without eventually resulting in precipitation of the chelating

235 agents in the samples that were acidified for analysis. Though DTPA worked better at pH  
236 11, the solubility limit of Ba drops quickly starting at pH 11 (dropping 2 orders of  
237 magnitude for every pH integer). For this reason, all chelating agents were studied at pH 9  
238 moving forward.

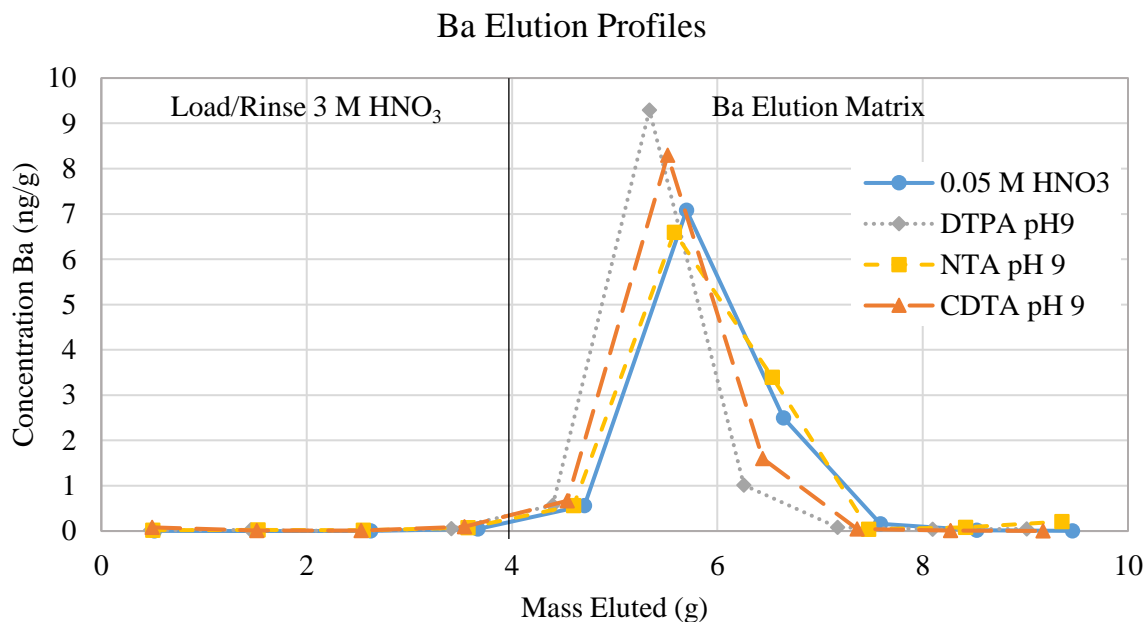
### 239 *Column Studies*

240 *Scoping Study:* Each of the systems identified as "optimal" were tested with the chelating  
241 agent at 0.05 M to help combat the possibility of chelator precipitation on the column  
242 during operation. Unfortunately, the pH 7 systems of DTPA and CDTA still had issues  
243 with precipitation. Because of this, these systems were also tested at pH 9. Small amounts  
244 of Ba were detected in the load/rinse fractions, and recoveries were above 80% for all  
245 columns (See Fig. 6). The columns eluted with NTA and CDTA had the best recoveries,  
246 though CDTA had nearly 2% of the Ba come out in the rinse fraction. Elution profiles  
247 from the four scoping study columns can be seen in Fig. 7. All of the elution matrices  
248 resulted in a peak maximum between 5.3-5.7 g.



249

250 **Fig. 6** Barium recovery using various elution matrices. NTA, CDTA, and DTPA solutions  
251 were adjusted to pH 9, all using NaOH



252

253 **Fig. 7** Elution profiles of barium from scoping study

254 *High Fidelity Measurements:* Following the results of the initial scoping study, high  
 255 fidelity measurements were made using NTA, CDTA, and KNO<sub>3</sub> as elution matrices. NTA  
 256 and CDTA were chosen because of the excellent recovery seen in the scoping study, and  
 257 KNO<sub>3</sub> was chosen because of the favorable competition shown in the batch studies. The  
 258 consistency seen in the elution peak location made it likely that the KNO<sub>3</sub> elution point  
 259 would also be in the 5.3-5.7 g elution range, and as a result, the volumes of eluent were not  
 260 changed at all.

261 The results of the high-fidelity measurements are found in Table 3, below. Notably, the  
 262 recovery of Ba using the K<sup>+</sup> competing cation showed much lower recovery than the  
 263 chelating agents. It is possible that this was partially due to not adjusting the pH of the  
 264 KNO<sub>3</sub> solution prior to its use as an elution matrix. However, even if it were adjusted and  
 265 the recovery improved, it is unlikely that the uncertainty of the recovery would be reduced  
 266 any further. This is because, even though high purity KNO<sub>3</sub> was used, a significant amount  
 267 of Ba still had to be subtracted from each elution fraction.

268 **Table 3** Results from the high-fidelity tests. Uncertainty is  $2\sigma$ 

Column	Fraction	NTA		CDTA		KNO <sub>3</sub>	
		Ba Recovery (%)		Ba Recovery (%)		Ba Recovery (%)	
1	Rinse	1.020	$\pm 0.008$	0.73	$\pm 0.03$	0.110	$\pm 0.003$
1	Elute	103	$\pm 3$	99	$\pm 1$	86	$\pm 6$
2	Rinse	0.64	$\pm 0.02$	0.34	$\pm 0.01$	0.95	$\pm 0.03$
2	Elute	102	$\pm 2$	102	$\pm 2$	89	$\pm 4$
3	Rinse	0.559	$\pm 0.007$	0.43	$\pm 0.01$	2.5	$\pm 0.2$
3	Elute	102	$\pm 1$	102	$\pm 1$	89	$\pm 2$
<b>Avg Rinse Recovery</b>		0.74	$\pm 0.03$	0.50	$\pm 0.03$	1.19	$\pm 0.08$
<b>Avg Elute Recovery</b>		102	$\pm 3$	101	$\pm 3$	88	$\pm 7$

269 In contrast to the KNO<sub>3</sub> trials, the NTA and CDTA columns had negligible amounts of Ba  
 270 contamination that needed to be compensated for. The recoveries associated with both of  
 271 the chelating agents is within  $2\sigma$  of 100%, indicating quantitative recovery. The  
 272 uncertainties associated with the data presented here are slightly larger than those achieved  
 273 by *Surrao et al.* with EDTA ( $100.9 \pm 2.2\%$ ).[7] This indicates that the two chelating agents  
 274 should be viable alternatives to EDTA for removing Ba from Sr Resin columns. This is  
 275 evidenced further by the two-fold improvement in uncertainty when compared to the KNO<sub>3</sub>  
 276 data, and in the uncertainties being less than those observed for elution with 0.05 M HNO<sub>3</sub>  
 277 in the earlier scoping study ( $90 \pm 4\%$ ).

## 278 **Conclusions**

279 The batch studies carried out in this work show that Ba retention on Sr Resin decreases  
 280 with increasing concentrations of all the chelating agents tested. The results of the batch  
 281 studies show that EDTA and NTA exhibit their best Ba retention reduction at pH 9, while  
 282 DTPA and CDTA exhibit their best at pH 7. Increasing pH alone also decreases Ba  
 283 retention, though barium's relatively low solubility in high-pH systems decreases the  
 284 attractiveness of such an elution matrix. It was also shown that Ba retention increased from  
 285  $k' = \min(0.13)$  to  $k' = 1-2$  with the addition of sodium and potassium nitrate salts, though  
 286 this is likely due to the addition of nitrate to the system. When compared with the retention  
 287 of Ba in nitric acid at the corresponding concentrations, the retention is an order of  
 288 magnitude lower, indicating at least some competition.[24]

289 The column chromatography experiments carried out indicate that the addition of a  
290 competing cation like  $K^+$  is not an effective means of improving Ba recovery from Sr Resin.  
291 They do show, however, that the chelating agents CDTA and NTA are viable alternative  
292 options to EDTA that improve both recovery and repeatability of Ba separations. The  
293 addition of these tools to the nuclear forensic examiner's selection improve the likelihood  
294 of an accurate age determination. Subsequently, it will be possible to make a better  
295 estimation of where illicit radiological materials may have come from.

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