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**Intermittent Elevated Radium Concentrations in Coastal Plain  
Groundwater of South Carolina, U.S.A.**

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**Abstract**

To learn the cause of intermittent radium concentrations in groundwater of Coastal Plain aquifers, 31 groundwater wells in South Carolina, U.S.A. were sampled for radium and other geochemical parameters. Sediments cored from near the well screens were also sampled to examine any relationship between sediment properties and radium concentration in the groundwater.

Elevated radium concentrations only occurred in groundwater with low electrical conductivity and pH values below 6.3. The adsorption edge for radium on hematite – a major surface active mineral in these aquifers -- is at a pH value of about 6. Near this value, small changes in pH can result in significant adsorption or desorption of radium. In groundwater with initially low alkalinity, small intermittent decreases in partial pressure of carbon dioxide in groundwater cause decreases in pH and desorption of radium. The result is intermittent elevated radium concentrations.

**Key Words**

Radium, groundwater, Coastal Plain, South Carolina

## 1. Introduction

Elevated natural radium concentrations in groundwater of the Upper Coastal Plain of South Carolina, as well as other coastal plain aquifers, are common (Edgar, 1963; Michel and Moore, 1980; Michel and Cothorn, 1986; Michel and Price, 1989). These concentrations often exceed the combined ( $^{228}\text{Ra} + ^{226}\text{Ra}$ ) radium drinking water limit of 0.18 Bq/L (5 pCi/L) and are of concern to a large portion of the population that obtains their drinking water from subsurface aquifers Zapecza and Szabo (1988). Wells that consistently exceed radium drinking water standards can generally be explained by local geological or mineralogical factors. For example, consistently elevated radium concentrations in Southeast Coastal Plain aquifers tend to occur near the Fall Line that separates Piedmont from Coastal Plain aquifers (Michel and Moore, 1980; Zapecza and Szabo, 1988). In the South Carolina Coastal Plain, mineralogical factors include the presence of placer deposits containing monazite (Mertie, 1953) and the occurrence of uranium- and thorium-rich deposits of gorceixite (Ferguson et al., 1979). More difficult to explain are intermittent concentrations of elevated radium common to many wells of this area. Groundwater from these wells occasionally has elevated radium concentrations. The persistence of this in particular wells strongly suggests that the phenomenon is not a result of analytical or sampling error. Intermittent elevated radium concentrations are of concern to water supply well owners, as well as those responsible for monitoring waste sites for radium emissions. In addition, occasional elevated concentrations of other metals of environmental concern may result from the same mechanisms that cause intermittent radium concentrations.

Intermittent elevated concentrations of radium in groundwater are not adequately explained by mechanisms invoked to explain consistent radium concentrations. Radium derived from direct dissolution of radium-bearing minerals should occur at fairly consistent concentrations. Furthermore, at the pH values typical of coastal plain aquifers (pH>5) most radium-bearing minerals are sparingly soluble. Radium flux into groundwater caused by alpha recoil should also result in consistent radium concentrations. Likewise, these mechanisms would suggest that elevated radium concentrations should occur wherever there is an abundance of uranium- and thorium-bearing minerals. However, this does not seem to be the case. Hence, the mechanism of intermittent radium occurrence must be more complicated and involve factors that vary with time.

In this study we analyzed groundwater samples from 31 background wells on or near the Savannah River Site (Figure 1) for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and other geochemical parameters. The Savannah River Site is a large nuclear facility operated by the United States Department of Energy. Because of the history of nuclear processing at this site, it is important to understand all radionuclide occurrences in groundwater, including those that are natural. The wells sampled for this study were installed specifically to monitor groundwater composition in uncontaminated areas for comparison to groundwater in contaminant plumes. Each of the wells is screened to monitor the top or bottom of a major aquifer. The 31 wells sampled cover all of the major aquifers in the area. Several municipal water supplies in the vicinity have had problems with intermittent radium concentrations, and groundwater sampled for this study is representative of that tapped by these systems.

## 2.0 Methods

The wells sampled have dedicated single or variable speed submersible pumps. Before sampling, each well was purged in excess of two well volumes until field parameters stabilized. A YSI Series 6 Water Quality Logging system with a flow through cell was used to measure water quality parameters, which included pH, conductivity, temperature, oxidation-reduction potential (ORP), and dissolved oxygen. A DRT-15 turbidimeter provided measurements of turbidity. Water samples collected for cation, anion, and radium analyses were filtered using in-line 0.45 micron filters.

3M Co. Empore™ RAD solid phase extraction (SPE) disks were used to concentrate radium from well water samples prior to quantification of the  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  by high resolution gamma-ray spectrometry. This served to concentrate the radium from a large volume sample (~1.8 liters) onto a 47mm SPE disk for a more efficient counting geometry. The SPE disks are composed of 90% by weight elemental specific resin beads, with the balance being Teflon microfibrals. To prepare a sample the disk was placed on a vacuum flask and the aqueous sample pulled through the disk at a flow rate of up to 300 mL per minute Beals et al. (2001). Barium-133 was added as a yield monitor although radium is generally quantitatively retained by the disk under most conditions. The disks were then counted in the Underground Counting Facility using a high purity germanium (HPGe) well-type gamma-ray spectrometer.

Samples for cation analyses were preserved with nitric acid and analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES), Varian Vista AX

Simultaneous ICP-AES. Unacidified samples were analyzed for anions using a Dionex DX500 ion chromatograph.

Sediment samples for heavy mineral separation, bulk geochemical, and grain size analysis came from archived geologic cores from locations of sampled wells. Sediment was collected from cores either above or at the same depths as the screen zones for nine of the monitoring wells. Mineralogical analysis was conducted using a Siemen D500 automated diffractometer in addition to optical microscopy. Density separations were used to concentrate heavy minerals for analysis. Analyses were done using optical microscopy, scanning electron microscopy with energy dispersive spectrometry, and x-ray diffraction.

Geochemical calculations were done using the United States Geological Survey program PHREEQC (Parkhurst, 1995). The radium adsorption curves were generated assuming an aquifer with 1 wt. % hematite as the dominant surface reactive mineral. It was further assumed that only 60% of the hematite surface was available for reaction. Surface complexation parameters were extrapolated from those for barium and strontium presented in Dzombak and Morel (1990) and the specific surface area used was 29 m<sup>2</sup>/g Fokkink, et al. (1986).

### **3.0 Results**

Results presented in Table 1 show that of the 31 wells sampled, <sup>228</sup>Ra was detected in 15 and <sup>226</sup>Ra was detected in 8. Only 1 well exceeded the combined radium standard of 0.18 Bq/L (5 pCi/L), though 3 additional wells exceeded 0.11 Bq/L (3 pCi/L) combined radium. The

low calcium and magnesium concentrations, as well as the low carbonate alkalinity indicate that the aquifers from which the groundwater is derived are predominantly silicate aquifers with little influence from carbonate minerals. This is consistent with the known geology of the area.

### *3.1 Relation of Radium Concentrations to Sediment Properties*

Relationships between chemical components in groundwater and physical properties of aquifers are common. Michel and Cothern (1986) found correlations of radium concentrations with aquifer rock type. The aquifers we sampled were all sandy coastal plain aquifers, and thus such analysis was not possible. However, we did analyze grain-size distribution and abundance of heavy minerals in aquifer material from cores taken when the wells were installed. The grain-size distribution is important because fine-grained sediments adsorb more radium than coarse sediments. Thus, fine-grained sediments can be a sink for radium and remove it from groundwater. Conversely, if conditions change to mobilize radium, fine sediments may release more to groundwater than coarse-grained sediments. Heavy mineral content of the sediments is also important because the heavy mineral fraction typically contains more uranium and thorium than the non-heavy fraction and can, thus, be a source of radium.

The solid samples analyzed were from the same elevations as the screen zones of the wells, often with multiple samples from each screen zone interval. These analyses were only done for wells with detectable radium concentrations in groundwater and results are shown in Table 2. The aquifers are dominated by coarse to medium sand, with the silt plus clay fraction ranging up to 16 wt.%. The mean wt.% of heavy minerals is 1.74 and the range is



from 0.15 to 8.45. The highest value may be the result of inclusion of several rock fragments in the heavy mineral fraction. The mineralogy of the heavy minerals was dominated by tourmaline, staurolite, and rutile, with minor amounts of zircon and garnet. The presence of monazite was confirmed in 1 sample. Despite the potential importance of these factors, no definitive relationships between them and radium concentrations were observed. These relationships may truly not exist or their absence may be an artifact of the small number of samples analyzed for heavy minerals.

### *3.2 Relation of Radium Concentrations to Groundwater Composition*

None of the geochemical relationships observed in systems with consistent elevated radium concentrations are present in this system. Several investigators have observed a positive correlation between total dissolved solids and radium concentrations (Herczeg, et al., 1988; Sidhu and Breithart, 1998; Sturchio et al., 2001). This is not true for the groundwater we analyzed, but this groundwater had much lower total dissolved solids than other studies. Others have also observed that radium behaves similarly to other alkali earths such as barium, strontium, and calcium (Sturchio et al., 2001). Yet, in the groundwaters we analyzed there is no correlation between concentrations of these metals and radium.

Nevertheless, there are relationships between radium concentrations and other parameters. Figure 2 illustrates that elevated radium concentrations only occur at pH values below about 6.3. Likewise, elevated radium concentrations only occur at low electrical conductivities (Figure 3). The relatively high detection limits for the radium analysis method tend to

obscure these relationships somewhat, but when detectable radium is differentiated from non-detectable the relationships are much clearer.

#### **4.0 Discussion**

The relationship between radium concentrations and conductivities is a reflection of a relationship between carbonate alkalinity and radium concentration. The anion suite in most Savannah River Site groundwater is dominated by bicarbonate ion (Strom and Kaback ,1992) which is a major contributor of electrical conductivity. Figure 4 is a plot of SRS groundwater analyses from Strom and Kaback (1992) illustrating that carbonate alkalinity is strongly correlated to electrical conductivity. Analyses from our study are also plotted in Figure 4 showing a similar trend.

These results are most consistent with adsorption controlling the behavior of radium in the aquifers sampled. Figure 5 is a calculated curve showing adsorption of radium on hematite versus pH. The adsorption edge is at a pH value of about 5.8, meaning that small deviations in pH near this value can result in large changes in amount of radium adsorbed. This is consistent with experimental results of Beneš et al. (1984). The adsorption edge depends, in part, on the amount of the surface reactive mineral in the aquifer. The dotted lines in Figure 5 show the variation from 70.5 to 23.5 grams of hematite. Most of the pH values of groundwater sampled are between 5.5 and 7.0, very close to the adsorption edge. Thus, intermittent decreases in pH could cause intermittent elevated radium concentrations from desorption of radium on surfaces of aquifer minerals.

In non-carbonate aquifers at relatively oxidized conditions, the strongest control on groundwater pH is the partial pressure of CO<sub>2</sub> (PCO<sub>2</sub>). Figure 6 shows the effect of PCO<sub>2</sub> on hydrogen ion concentration in water with different initial bicarbonate alkalinities. At low alkalinity there is little buffering capacity in the water and small increases in PCO<sub>2</sub> result in relatively large increases in hydrogen ion activity, or decreases in pH. In the low alkalinity groundwater sampled, small increases in PCO<sub>2</sub> could account for intermittent elevated radium concentrations. A model calculation of radium concentration in groundwater versus PCO<sub>2</sub> is presented in Figure 7. At amounts of reactive hematite less than 47 g/Liter of pore space and PCO<sub>2</sub> values less than 0.02 atm., small changes in PCO<sub>2</sub> result in large changes in dissolved radium concentration. Thus, intermittent small increases in PCO<sub>2</sub> could readily cause the intermittent elevated radium concentrations observed in coastal plain groundwater. Interestingly, the lack of pH buffering capacity in samples with low concentrations of fine-grained material (e.g. hematite) results in larger pH changes with varying PCO<sub>2</sub>, and in turn, larger variations in radium concentration. This may explain the lack of correlation between grain size distribution and aqueous radium concentrations, despite the tendency for fine-grained material to contain more radium.

## 5.0 Conclusions

These data suggest a conceptual model for intermittent elevated radium concentrations in groundwater that involves three intersecting factors. Foremost, there must be a source of radium. We have established that the sediments of the aquifers sampled are relatively enriched in heavy minerals, some of which contain elevated concentrations of uranium and thorium. The source minerals provide a continuous flux of radium that adsorbs to minerals in

the vicinity. Secondly, there must be fluctuations in pH of the groundwater, most likely caused by variations in  $PCO_2$ . When pH drops below the adsorption edge, radium is released to the groundwater. Finally, to allow variations in  $PCO_2$  to cause sufficient pH changes, the alkalinity of the groundwater must be relatively low. When these three factors intersect near a monitoring or water supply well, intermittent elevated radium concentrations may occur.

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**Figure Captions**

Figure 1: Location map of southeastern United States; inset shows Savannah River Site vicinity. Black circles on inset map show well locations.

Figure 2: Radium concentrations in groundwater versus pH. Closed symbols had detectable radium, open symbols were below the detection limit for that sample.

Figure 3: Radium concentrations in groundwater versus electrical conductivity. Closed symbols had detectable radium, open symbols were below the detection limit for that sample.

Figure 4: Electrical conductivity versus alkalinity for groundwater from the Savannah River Site. Closed circles are data from Strom and Kaback (1992), open squares are from this study.

Figure 5: Calculated adsorption curve for radium onto hematite. Solid line shows adsorption for system with 47 g hematite/L, dotted line shows adsorption for system with half this amount and twice this amount of hematite.



Figure 6: Calculated curves of hydrogen ion concentration versus partial pressure of carbon dioxide ( $\text{PCO}_2$ ) for water with different initial concentrations of bicarbonate ion.

Figure 7: Calculated curves of dissolved radium versus partial pressure of carbon dioxide for a system that includes adsorption of radium to hematite.

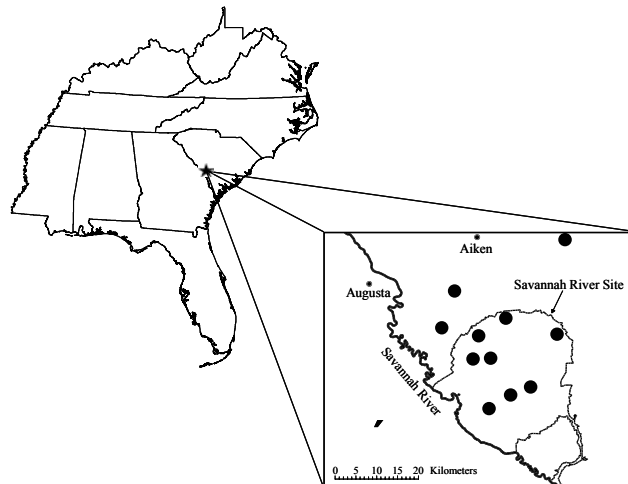
Table 1: Groundwater composition data from 31 wells. Gray highlights wells with radium concentrations above detection limit.

Well	Sample Date	pH	ORP	Dissolved	Specific	Turbidity	<sup>226</sup> Ra	<sup>228</sup> Ra	Ca	Mg	Na	K	Fe	Si	Cl	SO4
				Oxygen	Conductivity		pCi/L	pCi/L								
			mV	mg/L	uS/cm	NTU	pCi/L	pCi/L								
C1 AIK 2378	5/19/2003	5.57	NA	9.7	20	0	<0.62	0.58	1.990	<0.004	1.24	0.330	NA	5.34	2.00	2.29
C1 AIK 2380	5/19/2003	5.99	NA	8.02	19	6	<0.8	0.45	0.240	<0.004	1.04	0.285	NA	4.44	1.82	1.14
C2 AIK 818	5/21/2003	5.69	NA	9.06	14	1	1.2	<0.58	0.324	<0.004	0.816	0.254	NA	4.25	1.68	1.00
C2 AIK 825	5/21/2003	6	NA	8.06	18	0	<1.4	0.66	1.42	0.113	1.60	0.287	NA	3.21	1.90	1.00
C3 AIK 826	5/27/2003	5.58	NA	8.63	22	6	1.9	2.3	1.19	0.073	1.48	0.654	NA	5.36	1.70	5.14
C3 AIK 845	5/27/2003	5.44	NA	8.66	28	0	6.2	6.2	2.21	<0.004	0.856	0.520	NA	4.98	1.61	7.85
FC-2C	4/15/2003	7.11	139	4.28	171	0.69	<2.5	<1.1	NA	NA	NA	NA	NA	NA	NA	NA
FC-2D	4/15/2003	6.69	293	5.04	87	0.18	<2.2	<1.1	NA	NA	NA	NA	NA	NA	NA	NA
P-15C	4/29/2003	5.85	149	8.62	58	4.5	<1.7	<0.8	NA	NA	NA	NA	NA	NA	NA	NA
P-15TB	4/29/2003	6.98	-120	0.2	60	6	<2.0	<1.0	NA	NA	NA	NA	NA	NA	NA	NA
P-16A	4/15/2003	4.9	443	9.59	17	0.34	<1.1	1.3	0.452	0.346	0.675	0.311	<0.004	2.99	1.82	1.00
P-16B	4/16/2003	4.9	402	9.3	15	0.16	<2.1	1.5	0.327	0.348	0.529	0.303	<0.004	2.67	1.97	1.00
P-16TA	4/15/2003	5.58	268.3	5.48	23	14.3	<0.9	0.72	1.01	0.102	0.704	0.347	1.07	4.17	1.55	1.00
P-16TC	4/15/2003	5.63	293	6.68	18	12.8	<2.5	<1.2	1.75	0.134	0.705	0.462	1.34	4.46	1.87	1.78
P-17TA	4/16/2003	5.37	-28	0.11	38	0.54	2.5	2.3	8.38	0.274	0.477	0.591	0.431	6.08	1.53	9.42
P-17TC	4/21/2003	6.03	-2	0.12	49	0.76	<2.5	<1.1	2.04	0.314	0.717	0.704	3.41	4.85	1.69	11.1
P-23B	4/22/2003	7	-104	0.09	157	0.34	<0.62	<0.30	4.95	1.46	2.25	4.11	2.26	5.67	1.41	12.7
P-23TA	4/23/2003	5.53	NA	NA	61.1	1.14	<1.8	<0.84	1.68	0.354	3.37	3.24	3.40	5.33	1.37	12.5
P-23TC	4/22/2003	6.29	-58	0.13	70	14.7	1	0.64	38.2	0.727	1.09	1.05	0.221	7.41	2.40	10.8
P-24A	4/22/2003	9.14	216	5.22	84	1.3	<2.5	<1.1	8.38	0.204	1.54	1.31	1.54	4.46	1.54	11.7
P-24TA	4/21/2003	7.22	-225	0.05	65	6.24	<2.7	<1.1	9.06	0.431	0.631	0.950	1.27	5.66	1.92	8.67
P-24TC	4/22/2003	6.12	-25	0.11	59	0.98	<2.3	<1.1	13.8	0.216	1.24	1.16	<0.004	5.44	2.27	1.05
P-26A	4/23/2003	5.01	NA	NA	35.6	0.24	<1.5	<0.72	3.23	0.476	0.637	1.09	0.798	14.1	1.82	7.4
P-26B	4/23/2003	5.92	NA	NA	72.1	0.84	<2.3	<1.0	5.09	0.945	1.28	2.81	4.57	5.36	1.59	10.9
P-26TC	4/23/2003	5.84	NA	NA	65.4	0.16	<1.8	<0.88	10.5	0.575	1.28	0.922	<0.004	4.22	2.39	3.39
P-28TE	4/10/2003	6.15	32	0.15	60	0.4	<2.5	<1.3	9.25	0.217	0.619	0.675	0.062	6.21	1.72	11.7
P-29B	4/8/2003	5.7	349	7.68	27	0.6	<1.3	0.69	NA	NA	NA	NA	NA	NA	NA	NA
P-29TA	4/8/2003	5.7	251	4.87	25	11.5	1.4	0.52	0.46	0.117	0.689	0.411	3.53	4.33	2.16	1.45
P-29TD	4/9/2003	5.43	116	0.84	34	3.4	3.3	1.36	2.25	0.149	0.583	0.352	3.07	4.81	1.85	8.72
P-30TA	4/9/2003	5.56	298	5.9	20	26	<1.5	<0.42	0.401	0.026	0.679	0.36	2.15	3.9	2	1.17
P-30TC	4/10/2003	7.36	286	8.67	51	2.2	<0.36	0.25	9.01	0.014	0.802	0.349	0.071	4.18	1.78	1.28

NA -- Not Analyzed

Table 2: Properties of sediment cored near groundwater wells.

Well	Sample Depth	Wt.% Heavy Minerals	Sieve Analyses wt%							
			coarse sand >0.6 mm	medium sand 0.6-0.425    0.425-0.25		fine sand 0.25-0.18    0.18-0.15		very fine sand 0.15-0.106    0.106-0.075		silt+clay <0.075
P-29B	179	0.33	14.81	20.47	40.55	12.28	3.12	2.92	1.75	4.09
	186	0.61	54.37	19.20	10.84	4.37	1.90	2.47	1.52	5.32
	195	3.91	13.29	6.36	5.78	6.74	10.02	32.76	17.34	7.71
P-29TA Acid	656	2.28	55.41	7.53	7.34	3.28	1.74	3.28	5.02	16.41
	662	0.35	61.52	10.55	13.87	5.08	1.76	2.34	1.56	3.32
	676	3.08	63.15	9.96	11.75	3.39	1.20	1.59	1.39	7.57
P-29TD	403	0.63	32.88	39.81	14.81	2.12	0.96	1.73	1.54	6.15
	416	8.45	68.55	12.50	6.45	2.62	1.01	1.61	1.21	6.05
	425	5.50	76.08	4.90	3.92	2.35	1.18	1.76	1.96	7.84
P-23TC	575	0.86	37.52	7.54	9.48	7.54	6.58	17.41	5.80	8.12
	579	0.45	80.27	6.51	3.64	2.11	1.34	3.26	0.96	1.92
	587	0.22	50.63	12.87	10.76	5.49	2.53	3.80	3.16	10.76
P-16A	125	1.44	63.54	23.83	7.33	1.83	1.02	1.22	0.41	0.81
	136	0.15	51.44	32.10	9.88	2.26	1.44	1.65	0.41	0.82
P-16B	76	0.68	22.43	26.34	27.37	7.61	5.97	7.61	1.23	1.44
P-16TA	616	0.34	25.68	24.32	40.08	4.86	0.78	1.17	0.78	2.33
P-24TA	955	1.76	72.06	10.60	5.59	1.93	0.77	1.35	1.35	6.36
	968	0.27	12.87	17.82	43.96	15.45	3.96	2.18	0.99	2.77
P-24TC	588	1.78	42.75	27.85	17.02	3.87	1.35	1.74	1.35	4.06



**Figure 1**



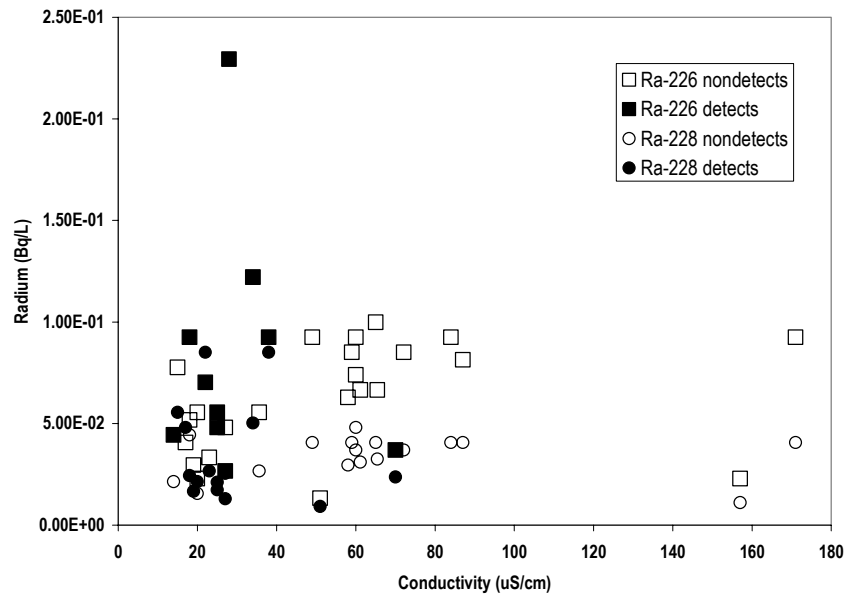


Figure 3

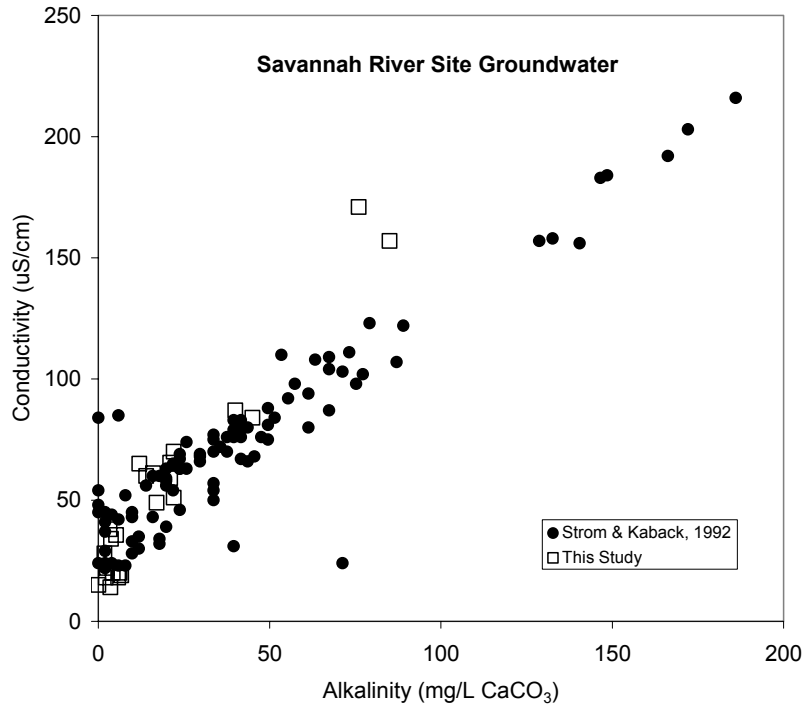
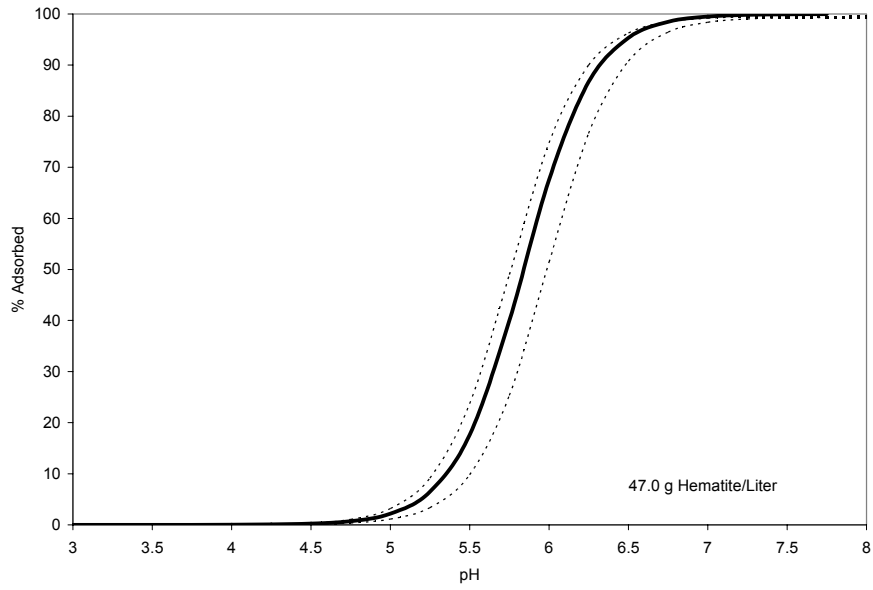


Figure 4



**Figure 5**



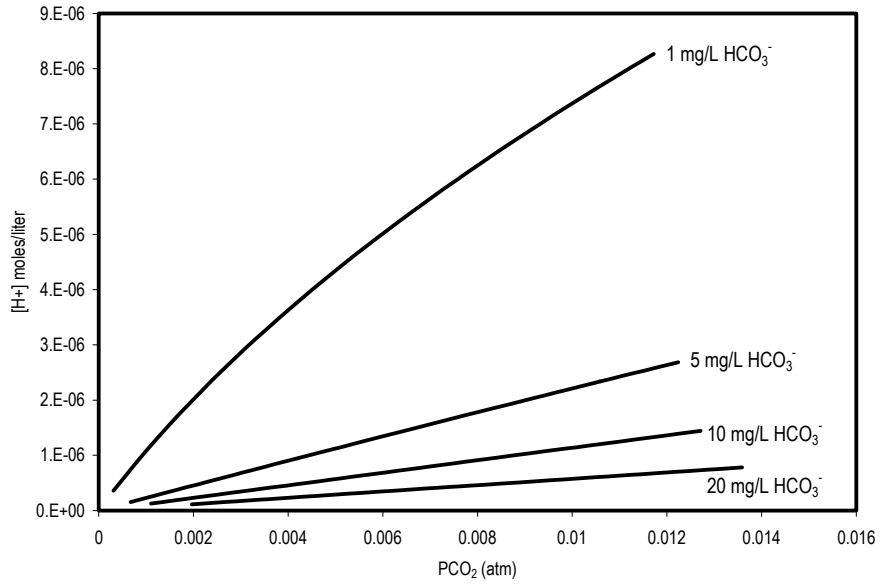
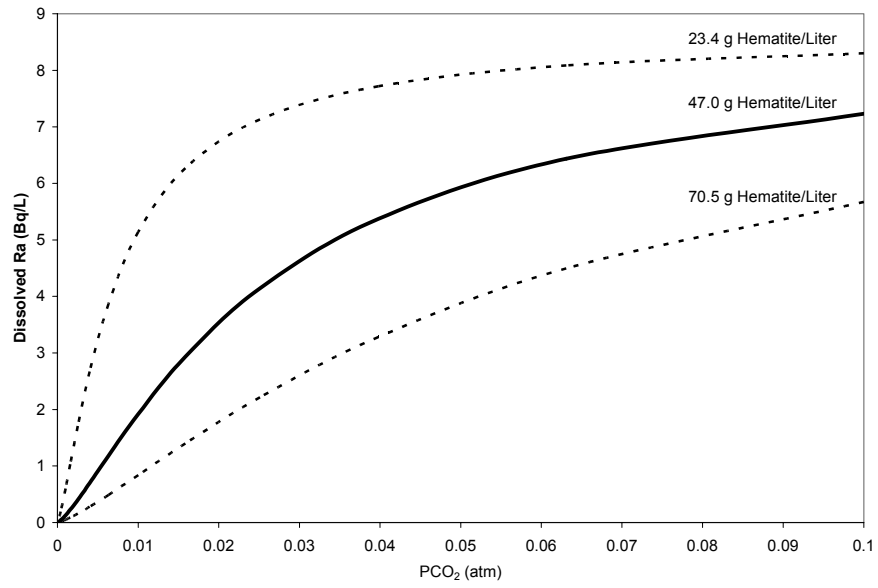


Figure 6



**Figure 7**