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ANOMALOUS CONCENTRATIONS OF $^{58}$Co AND $^{137}$Cs IN DARDANELLE RESERVOIR

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ABSTRACT

The unforeseen occurrence of significant concentrations of Co-58 and Cs-137 in a quiescent backwater of Dardanelle Reservoir provided an opportunity to observe the equilibrium distribution of Cs-137 between solution and suspended solids. This equilibrium has not been observed in other areas of the reservoir because small amounts of this nuclide are regularly injected into the lake by the AP&L Nuclear I facility. The systematics of the Co-58 concentration lead to the conclusion that significant amounts of these two nuclides had been injected into the sampling area from a source unknown.

INTRODUCTION

The distribution of Cs-137 between solution and the surface of clay minerals in the Dardanelle Reservoir has been studied by Chittenden (1983). A statistical study of the data showed that, in those parts of the reservoir accessible to the cooling water released from the Arkansas Nuclear I facility, the concentration of Cs-137 in the water is simply a function of the amount of activity of the nuclide released from the reactor and that, consequently, ion exchange of Cs-137 between solution and the surface of suspended particles was not a significant factor in controlling the nuclide's concentration in solution. The residence time of water in the reservoir was too short for equilibrium to be attained and thus no significant correlation was observed between the concentrations of Na(I) and Cs-137 at the stations sampled continuously through the four years of the study.

Upon examining the data (Chittenden, 1978) from an infrequently sampled station (6), the possibility of a correlation between the aqueous Cs-137 activity and the total dissolved solids (TDS) present in the water can be seen. TDS is used as a measure of Na(I) concentration because the concentrations of individual ionic species were not determined during the 1975-1977 portion of the study. This station is unusual among the stations sampled in that it is outside of the normal circulation pattern which is induced in the reservoir by the pumping of cooling water for the AP&L facility. The station is located in a "bay" on the western bank of the Illinois Bayou where Highway US 64 crosses the stream. This area is marked by quiescent surface and subsurface waters (Bechtel Corporation, 1969).

Attention was first brought to this backwater by the appearance of high concentrations of Co-58, summarized in Table 1. The monotonic decrease of this nuclide's concentration over a period of fifteen months was quite unusual. Once plotted (Figure 1), the data resembled closely a radioactive decay curve with a half-life of 72.6 days. The half-life of Co-58 is 71.3 days. It seems that the concentration of aqueous Co-58 depends only on the half-life and that the nuclide is not being washed out of the sampling area by turnover that would be expected even in quiescent areas. It can thus be concluded that the source of this aqueous Co-58 is one fixed in position, i.e. absorbed on the bottom sediment with small amounts being released to the water through ion exchange with aqueous ions.

The concentrations of a number of divalent transition metal ions have been measured in the Arkansas River at Van Buren and the combined concentration was fairly constant (United States Geological Survey, 1978). It can then be assumed that the elution rate of Co-58 remained constant over the period of this study. The distribution coefficient of trace quantities of Co(II) between clay and solution in the presence of di- and trivalent aqueous ions can be estimated from Erickson (1979) to be greater than 10^4 L/kg.

Figure 1. Variation of the Aqueous Co-58 Concentration with Time (t_0 = December 1, 1975)
CONCLUSIONS

It can be concluded that a significant amount of Co-58 found its way to this quiescent backwater area and was adsorbed by the bottom sediment where it slowly decayed with only a minute and constant fraction of the activity being released from the clay by ion exchange. It was observed, in June, 1976, that when significant amounts of Co-58 were found in water samples, Cs-137 was also present in relatively large quantities. If Cs-137 was injected into the area of station #6, it is possible that an ion exchange equilibrium could have been established between aqueous and adsorbed Cs-137 in this area of the reservoir where injections of newly formed radionuclides would be rare due to its isolation from the lake’s main circulation patterns (Chittenden, 1978; 1981; 1983). The variation in the aqueous concentration of Cs-137 should be predictable by an equation derived from ion exchange theory,

\[ \frac{1}{Cs} = C_1 + \frac{C_2}{TDS} \]

where

- \(Cs\) = the activity of aqueous Cs-137 in pCi/L
- TDS = total dissolved solids in g/L
- \(C_1, C_2\) = constants

The variation of Cs-137 with TDS is summarized in Table 1 and Figure 2. The points in Figure 2 were fit to the best straight line by linear regression analysis.

\[ \frac{1}{Cs} = -14.7 + \frac{7.172}{TDS} \]

Table 1. TDS and Aqueous Concentrations of \(^{137}\)Cs and \(^{58}\)Co

<table>
<thead>
<tr>
<th>Collection Date</th>
<th>TDS(g/L)</th>
<th>(^{137})Cs(pCi/L)</th>
<th>(^{58})Co(pCi/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/14/75</td>
<td>---</td>
<td>---</td>
<td>0.08 ± 0.13</td>
</tr>
<tr>
<td>3/27/76</td>
<td>0.159</td>
<td>0.053 ± 0.020</td>
<td>0.41 ± 0.05</td>
</tr>
<tr>
<td>6/21/76</td>
<td>0.511</td>
<td>1.30 ± 0.10</td>
<td>0.20 ± 0.02</td>
</tr>
<tr>
<td>8/19/76</td>
<td>0.312</td>
<td>0.24 ± 0.03</td>
<td>0.17 ± 0.04</td>
</tr>
<tr>
<td>10/29/76</td>
<td>0.375</td>
<td>0.081 ± 0.007</td>
<td>0.047 ± 0.019</td>
</tr>
<tr>
<td>1/25/77</td>
<td>---</td>
<td>---</td>
<td>0.018 ± 0.022</td>
</tr>
<tr>
<td>3/26/77</td>
<td>0.191</td>
<td>0.016 ± 0.003</td>
<td>0.000 ± 0.023</td>
</tr>
</tbody>
</table>

Although the standard deviation of the observed values of 1/Cs from those predicted by the regression line was large (0.2 L/g), the value of \(R^2\) was 86.4\%, indicating that the equation is an adequate predictor of the aqueous Cs-137 activity. Ion exchange is the major factor determining the concentration of this nuclide at station #6 where there is little occasion for upsetting the equilibrium.

LITERATURE CITED


