UPDATE SUMMARY OF THE 1991 DISCHARGE OF ENRICHED URANIUM TO THE SANITARY SEWER (DEUSS) REPORT

April 1995

Prepared by
H & R Technical
151 Lafayette Drive, Suite 220
Oak Ridge, Tennessee 37831

for the
Water Compliance Section
Environmental Management Department
Health, Safety, Environment, and Accountability Organization

Oak Ridge Y-12 Plant
Oak Ridge, Tennessee 37831
managed by
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
Under Contract DE-AC05-84OR21400

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1.0 INTRODUCTION

In 1991, a team of staff members from various organizations at the Y-12 Plant conducted and published a review (Duke et al, 1991) of the historical background and regulatory and technical issues associated with release of uranium to the Oak Ridge sewer system.

The study provided a basis to conclude that the Y-12 Plant is in compliance with respect to all regulatory issues, including the possibility of criticality. Uranium concentrations in the Oak Ridge sewer system are generally less than drinking water quality criteria and are on a decreasing trend.

Since the 1991 Y-12 Plant report, a flow study of the Y-12 Plant sanitary sewer collection system has been completed by the Y-12 Plant staff (Hanzelka and Maguire, 1993). Additional data has been obtained by the Y-12 Plant and the City of Oak Ridge (COR, 1994). COR developed limits on radionuclide concentrations in sludges used for land application (Stetar, 1993). Martin Marietta Energy Systems has provided recommendations to the Department of Energy (DOE) regarding the impacts of sludge land farming operations on the ORR (Frye, 1992). The DOE Office of Nuclear Safety (USDOE, 1993) completed an audit of activities related to radiological contamination of the COR sewer system due to DOE operations. In addition COR is currently developing limits on radionuclide releases for all industrial customers to be applied through the permitting process. In 1994, a new sanitary sewer monitoring station was installed and began operation at the Y-12 Plant to determine releases specifically from the Y-12 Plant. Previously, estimates were based on mass balance calculations using data from the City Monitoring Station which monitors Union Valley and Y-12 Plant releases.

The purpose of this report is to update the 1991 Y-12 Plant study taking into account current data and information.

2.0 Y-12 PLANT INPUT TO THE OAK RIDGE SEWER SYSTEM

Sources of uranium in the Oak Ridge sewer system include the Clinch River which is the water supply, the Y-12 Plant, Union Valley Industrial Park, and balance of the industrial and domestic users of the COR sewer system before the flow reaches the COR publicly owned treatment works (POTW). Other natural radioisotopes (e.g. Th-230, Ra-226, Rn-222 and short-lived daughters, Pb-210 and Po-210) are present in the water supply and will be present in the sewer system. Other radioisotopes (e.g. cesium and cobalt) are added from other industrial sources in Oak Ridge. Medical radioisotopes are released from the medical complex associated with Methodist Medical Center.

The uranium input from the Y-12 Plant may be enriched in U-235 which raises special nuclear material (SNM) issues as discussed in Duke et al, 1991. The U-235 release in the sewer system was estimated to be less than 1 gram per day (g/d). Total amounts of U-235 accumulated in the POTW were less than 40 grams (g) at concentrations less than $1 \times 10^{7}$ grams per liter (g/L) which is over 8 orders of magnitude less than the maximum subcritical concentration of 11.6 g/L (see Appendix M in Duke et al, 1991).
No direct data were available for 1991, 1992 or 1993 on the radioisotope content and flow for the Y-12 Plant wastewater. Flow and uranium content were measured at the city station which includes both Union Valley and the Y-12 Plant. Uranium content was measured for Union Valley. Starting in late 1993, new flow measurements gave results for Union Valley of about 0.2 x 10^6 liters per day (L/d) (50,000 gallons per day). The city station flow has been measured to be about 2.8 x 10^6 L/d. So the average Y-12 Plant flow is given by the difference (2.6 x 10^6 L/d). A new station at the Y-12 Plant began operation in 1994 to monitor flow and radioisotope content so that the Y-12 Plant contribution can be determined on the basis of direct data. Before data were available, the contribution of the Y-12 Plant alone was based on mass balance calculations. When the new station began operation, monitoring at the Union Valley and City Stations were discontinued. The result is that data were collected for all three sites for one weekly time period. Data were collected for the City Station and Union Valley for the period 1/6/94 - 7/28/94 while data for the Y-12 Plant were collected for the period 7/23/94 - 11/22/94. Rainfall during the 1/6/94 - 7/28/94 period was unusually high - more than 20 inches above normal rainfall for the period. Therefore, the data for Union Valley and City Station are atypical and are not used for mass balance analysis.

2.1 Y-12 Plant Water Supply and Wastewater

Flow and concentration data for the years 1992, 1993 and 1994 at the City Station, Union Valley and the Y-12 Plant are given in Table 1. Estimates based on mass balance calculations are shown in parenthesis. The 1994 values are based on a relatively complete data set made possible by operation of the new East End Sanitary Sewer Monitoring Station (EESMS) which integrates the total contribution of the Y-12 Plant to the City Station. The consistency of the Y-12 Plant flow estimates for 1992 and 1993 with actual data for 1994 indicates that previous estimates based on mass balance calculations are reasonable. Therefore, the decreasing uranium release trend for the Y-12 Plant shown by the data in Table 1 may be accepted as reflecting actual conditions. The uranium releases from the Y-12 Plant given in Table 1 are substantially less than estimated in the Duke et al study (40 g/d) and provide documentation for the decreasing trend noted in the Duke et al study.

The Y-12 Plant water supply (Clinch River) contains background concentrations of natural radionuclides. Surface water in the U.S. contains, on average, about 2 parts per billion (ppb) total uranium. Using this value, the uranium release for the Y-12 Plant sewer system from background uranium is estimated to be about 5.2 g/d. Excess uranium release estimates are given in the last column of Table 1. The Y-12 Plant contribution to uranium release in the Oak Ridge Sewer System is estimated to be about 1.2 g/d in 1994.
### Table 1 - Uranium in the Y-12 Plant sewer system wastewater.

<table>
<thead>
<tr>
<th>Location</th>
<th>Year</th>
<th>Flow (10^3 L/d)</th>
<th>Total Uranium Concentration (ppb)</th>
<th>Total Uranium Release (g/d)</th>
<th>Excess Uranium Release (g/d)¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>City Station</td>
<td>1992</td>
<td>2.8</td>
<td>7</td>
<td>19.6</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>1993</td>
<td>2.8</td>
<td>6</td>
<td>16.8</td>
<td>11.2</td>
</tr>
<tr>
<td></td>
<td>1994²</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Union Valley</td>
<td>1992</td>
<td>0.2</td>
<td>29</td>
<td>5.8</td>
<td>5.4</td>
</tr>
<tr>
<td></td>
<td>1993</td>
<td>0.2</td>
<td>25</td>
<td>5.0</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td>1994</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Y-12 EESSMS</td>
<td>1992</td>
<td>(2.6)³</td>
<td>(5.3)</td>
<td>(13.8)</td>
<td>(8.6)</td>
</tr>
<tr>
<td></td>
<td>1993</td>
<td>(2.6)</td>
<td>(4.5)</td>
<td>(11.8)</td>
<td>(6.6)</td>
</tr>
<tr>
<td></td>
<td>1994</td>
<td>2.6</td>
<td>2.5</td>
<td>6.4</td>
<td>1.2</td>
</tr>
</tbody>
</table>

¹ Excess Uranium = total uranium - background uranium. Background uranium concentration is taken to be the U. S. average background surface water concentration of about 2 ppb.

² Data were atypical for the City Station and Union Valley during 1994 (see text).

³ Numbers in parentheses are based on mass balance calculations.

The excess uranium release from the Y-12 Plant contains relatively low concentrations of other radionuclides in the natural decay series compared to their relative levels in the water supply because the uranium processed at the Y-12 Plant was separated from daughter products in the milling component of the fuel cycle. The total dose from daughter products at equilibrium far exceeds that due to uranium isotopes alone. Thus, the Y-12 Plant contribution to the overall risks associated with Oak Ridge Sewer system is expected to be small compared to the contribution of background concentrations of natural radionuclides in the system.

The decreasing trend and low total release amounts for total uranium apply for the uranium isotopes U-238, U-234 and U-235. The less than 1 g/d estimate given in the
Duke et al study for U-235 is replaced by the 1994 value based on actual data of less than 0.2 g/d. This value was determined using the following equation:

\[
\text{Conc. } U_{235} \times \text{Flow} \times \frac{\text{Specific Activity } U_{235}}{2 \times 10^6 \text{ pCi/g}} = 0.17 \text{g/d}
\]

3.0 EVALUATION OF URANIUM RELEASES FROM THE Y-12 PLANT

Concentrations of uranium isotopes in the Y-12 Plant wastewater to the Oak Ridge Sewer System are compared to guidelines and standards in Table 2. While no direct regulations exist for uranium concentrations in sewers, the data in Table 2 provides a basis for concluding that the concentrations in the Y-12 Plant releases to the Oak Ridge sewer system are well below even drinking water criteria. The proposed sanitary sewage limits in 10 CFR Part 834 which codifies DOE Orders 5400.5 "Radiation Protection of the Public and the Environment," are concentration limits at sewer release points of five times the derived concentration guides, annual total releases of 5 curies (Ci) for H-3, 1 Ci for C-14 and 1 Ci for others combined and potential doses must be a small fraction of the primary EDE limit of 100 millirems per year (mrem/yr). The Y-12 Plant sanitary sewage system clearly meets these limits by a wide margin.

Table 2. Concentrations of uranium in the Y-12 Plant wastewater to the Oak Ridge Sewer System compared to guidelines and standards.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Y-12 Plant Wastewater Concentration (pCi/L)</th>
<th>Drinking Water Criteria (pCi/L)(^1)</th>
<th>Ratio(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>1.22</td>
<td>22</td>
<td>0.055</td>
</tr>
<tr>
<td>U-234</td>
<td>2.61</td>
<td>19</td>
<td>0.14</td>
</tr>
<tr>
<td>U-235</td>
<td>0.13</td>
<td>21</td>
<td>0.0062</td>
</tr>
<tr>
<td>Totals</td>
<td></td>
<td></td>
<td>0.20</td>
</tr>
</tbody>
</table>

\(^1\) These limits are based on the general EPA dose limit of 4 mrem/yr for the drinking water pathway with 2 L/d intake. The EPA MCL is 30 pCi/L for uranium (Federal Register, Vol. 56. No. 138, Thursday, July 18, 1991).

\(^2\) The ratio of wastewater concentration to drinking water criteria. The ratios for DOE derived concentration guides which are equivalent to 100 mrem/yr assuming a 2 L/d intake rate would be about 25 times lower.
Concentrations in the sewer line even at locations near possible sources (see Section 4) are generally less than drinking water criteria (4 mrem/yr or approximately 1/25 x DCGs). Total release of uranium isotopes above background from Y-12 Plant to the Oak Ridge Sewer System is less than 5 millicuries per year. Potential doses to members of the public due to releases from the Y-12 Plant sewer system are less than 1 mrem/yr including the potential doses from land application of POTW sludges on the Oak Ridge Reservation. A pathways analysis would be necessary to more quantitatively assess the significance of the Y-12 Plant uranium releases. However, the pathways analysis would show further dilution of the concentrations below drinking water criteria. Other potential pathways such as land application of Oak Ridge sewage sludge are being analyzed in other studies (see references).

Based on 1993 data a total uranium input to the Oak Ridge POTW of about 50 g/d resulted in a dry sludge total uranium concentration of about 40 picocuries per gram (pCi/g). According to calculations by Stetar (1993), the sludge concentration corresponding to 4 mrem/yr based on worst case assumptions for land application plans on the ORR is about 440 pCi/g. Therefore, the input to the Oak Ridge POTW to produce 4 mrem/yr from land application would be about 500 g/d. At current total uranium release levels due to the Y-12 Plant operations (~1 g/d), it does not appear that release limits to be developed by the City of Oak Ridge will have any impact on the Y-12 Plant operations.

The Y-12 Plant effluent measurements of total uranium and %U-235 are made using mass spectrometry. The minimum detectable amount for total uranium is 1 ppb under typical measurement conditions. The average value reported (2.5 ppb) is well above the minimum detectable value. Contributions of elements other than uranium with 238, 235, and 234 mass numbers should be very low.

Measurements of U-238, U-235, and U-234 isotopes are made using alpha spectrometry. The minimum detectable activities (MDAs) are about 0.5 picocuries per liter (pCi/L) compared to the measured values given in Table 2. Alpha activity can contribute to the uranium isotope peaks. Measured alpha activity was highly variable averaging about 6.3 pCi/L against an MDA of 15 pCi/L.

Overall, the total uranium values based on mass spectrometry should be more accurate than the alpha spectrometry values for the low levels of uranium measured. However, in order to obtain activity concentrations, it is necessary to determine either total uranium and %U-235 mass concentrations or isotopic composition. For high specific activities, total activity concentrations could be significant even though total uranium is less than 1 ppb. Both methods indicate that the uranium release concentrations in the Y-12 Plant sanitary sewer effluent have shown a decreasing trend and are well below DOE guidelines and EPA regulations.

Calculations for the total uranium release estimates based on specific isotope levels are:

\[
U-238: \frac{1.22 \text{ pCi/L} \times (2.6 \times 10^6 \text{ L/d})}{0.33 \times 10^6 \text{ pCi/g}} = 9.6 \text{ g/d}
\]

\[
U-235: \frac{0.13 \text{ pCi/L} \times (2.6 \times 10^6 \text{ L/d})}{2 \times 10^6 \text{ pCi/g}} = 0.17 \text{ g/d}
\]

\[
U-234: \frac{2.61 \text{ pCi/L} \times (2.6 \times 10^6 \text{ L/d})}{6200 \times 10^6 \text{ pCi/g}} = 0.001 \text{ g/d}
\]
Therefore, the total uranium release based on isotopic composition is about 9.8 g/d and the excess release based on the background release estimate of 5.2 g/d is about 4.6 g/d. The results for total uranium and %U-235 using alpha spectrometry are significantly higher than the results obtained using mass spectrometry. This incongruity is not unusual, considering the majority of alpha spectrometry results were less than the MDAs. As stated above, the total uranium values based on mass spectrometry should be more accurate. It is possible to get better sample result agreement by increasing sample volumes or sample count times for the alpha spectrometry, thereby lowering the MDAs. The purpose of this study, however, is to determine compliance with current regulatory requirements and lower MDAs would not affect the conclusions herein.

4.0 ON-SITE SOURCES OF URANIUM IN THE Y-12 PLANT SEWER SYSTEM

Although the Y-12 Plant contribution of uranium to the Oak Ridge Sewer System is very low, the U.S. DOE, Office of Nuclear Safety (U.S. DOE, 1993) commented that DOE/ORO had not fully quantified the concentration of uranium entering the sewer system from known sources such as hand wash basins and dishwashers at the Y-12 Plant. Additional sampling has been conducted at several locations within the Y-12 Plant sanitary sewer collection system. Sampling included the sewer lines from a change house and a labware dishwasher. Releases to the sewer system from a hand washing station used to remove low level uranium contamination were estimated by Y-12 Plant health physicists to be less than 0.2 microcurie per year. Such releases would result in concentrations of less than 2 x 10^4 pCi/L at the City Station. Sampling also included sewer mains which serve several different areas of the Y-12 Plant.

Total uranium mass values and isotope uranium values are given in Table 3 for sewer lines at the west end of the Y-12 Plant. Caveats which apply to the data are listed in the footnotes to Table 3. Total uranium values range from less than 2 to 54 ppb. Taking into account the caveats, total activity concentrations ranged from 5 to 30 pCi/L with percent mass of U-235 varying from about 0.4% to 5%. While these concentrations are less than drinking water criteria (see Table 2), they are higher than expected background concentrations. Concentrations in the wastewater from the labware dishwasher reflected by station SS-600 were in the low end of the range. Total uranium concentrations in wastewater from the change houses were consistently less than 1 ppb. Specific sources are not identifiable from the data.
Table 3  Concentrations of uranium in Y-12 Plant sewer samples.

<table>
<thead>
<tr>
<th>Sample Location</th>
<th>Date</th>
<th>U-238 (pCi/L)</th>
<th>U-234</th>
<th>U-235 (Mass % U-235)</th>
<th>Total uranium (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SS-710</td>
<td>4/22/94</td>
<td></td>
<td></td>
<td></td>
<td>0.021</td>
</tr>
<tr>
<td>9401-3</td>
<td>4/29/94</td>
<td></td>
<td></td>
<td>0.42</td>
<td>0.018</td>
</tr>
<tr>
<td></td>
<td>4/14/94</td>
<td></td>
<td></td>
<td>0.4</td>
<td>0.05</td>
</tr>
<tr>
<td>SS-720</td>
<td>4/22/94</td>
<td></td>
<td></td>
<td>0.38</td>
<td>0.021</td>
</tr>
<tr>
<td>9767-13</td>
<td>4/29/94</td>
<td></td>
<td></td>
<td>0.42</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>4/14/94</td>
<td></td>
<td></td>
<td></td>
<td>0.054</td>
</tr>
<tr>
<td>SS-700</td>
<td>2/01/94</td>
<td>14</td>
<td>6.9</td>
<td>0.038</td>
<td>0.42</td>
</tr>
<tr>
<td>9204-3</td>
<td>2/02/94</td>
<td>13</td>
<td>6.1</td>
<td>0.072</td>
<td>0.39</td>
</tr>
<tr>
<td></td>
<td>2/03/94</td>
<td>12</td>
<td>5.6</td>
<td>0.37</td>
<td>0.44</td>
</tr>
<tr>
<td>SS-800</td>
<td>2/01/94</td>
<td>2.6</td>
<td>4.5</td>
<td>0.28</td>
<td>0.93</td>
</tr>
<tr>
<td>9204-2</td>
<td>2/02/94</td>
<td>3.8</td>
<td>6.4</td>
<td>0.43</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>2/03/94</td>
<td>2.1</td>
<td>4.1</td>
<td>0.5</td>
<td>1.2</td>
</tr>
<tr>
<td>SS-600</td>
<td>2/01/94</td>
<td>0.41</td>
<td>4.5</td>
<td>0.26</td>
<td>NA</td>
</tr>
<tr>
<td>9710-2</td>
<td>2/02/94</td>
<td>0.36</td>
<td>4.6</td>
<td>0.11</td>
<td>263</td>
</tr>
<tr>
<td></td>
<td>2/03/94</td>
<td>0.41</td>
<td>724</td>
<td>3.4</td>
<td>NA</td>
</tr>
</tbody>
</table>

1 Stations SS-710 and SS-720 should be similar to SS-700 as they are in the same line with no known sources between.

2 This is a suspect value since for the listed isotopic values the % U-235 value should be 0.09%.

3 This is a suspect value since for the listed isotopic values the % U-235 should be 4.8%.

4 This is a suspect value since for the calculated % U-235 of 58%, the total uranium value (pCi/L) should be about 31 pCi/L even if the total uranium value were 1 ppb.
Since portions of the sewer line are old and probably leak, they could receive surface water and groundwater. Concentrations of uranium isotopes in the surface water in the vicinity of the sewer line samples are in the same range as the sewer line samples. The highest groundwater concentrations at the west end of the Y-12 Plant are associated with the S-3 plume. Although the S-3 site is west of the hydrologic divide that separates the Bear Creek Regime from the East Fork Poplar Creek Regime, the S-3 plume has crossed the divide because of a mound in the water table created by the disposal of large volumes of liquid wastes during operation of the S-3 site. The sewer samples were taken in the western end of the East Fork Poplar Creek regime. There the S-3 plume mixes with other plumes from sites in the East Fork Poplar Creek regime.

No isotope specific data is available for groundwater in the areas near the sewer line sampling points. Gross alpha activity exceeding 15 pCi/L occurs in the unconsolidated zone at the west end of the East Fork Poplar Creek Regime. Gross beta activity exceeded 50 pCi/L in the same area. Gross alpha and gross beta were not measured in the sewer line samples. Even if they were, no quantitative statements could be made about the sources of uranium in the sewer lines. A detailed (and costly) study would be necessary to identify sources. Given plume mixing, quantitative results would be difficult to achieve.

A study of the Y-12 Plant sewer pipelines is underway to determine replacement needs. As the pipelines are replaced or repaired, external sources of contamination will be eliminated. Since contamination levels are low, it does not appear necessary to expend a lot of questionably productive effort towards quantitative definition of sources. Monitoring of surface water, groundwater and soils will continue and will be directed for specific studies as necessary.
5.0 REFERENCES


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