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TITLE

RELEASE OF RADIOACTIVITY TO THE
COLUMBIA RIVER FROM IRRADIATED
FUEL ELEMENT RUPTURES

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RELEASE OF RADIOACTIVITY TO THE COLUMBIA RIVER
FROM IRRADIATED FUEL ELEMENT RUPTURES

May 27, 1965

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W. N. Koop
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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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RELEASE OF RADIOACTIVITY TO THE COLUMBIA RIVER
FROM IRRADIATED FUEL ELEMENT RUPTURES

INTRODUCTION

McCormack and Schwendiman issued a report in 1959⁽¹⁾ which estimated the amount of fission products from fuel element failures entering the Columbia River during the period 1952-1958 inclusive. Since that time there have been no attempts made to publish similar information for the subsequent years.

It is the purpose of this report to review the fuel element rupture experience of 1964 as somewhat typical of the last few years, and to estimate the amount of significant fission products which entered the Columbia River. The routine measurements of fission products both in the reactor effluent streams and in the Columbia River will be reviewed for comparison purposes.

DISCUSSION

1964 Ruptured Slug Experience

There were 97 fuel element failures during calendar year 1964. Seventeen of these ruptured elements could not be found and identified in the routine search made after discharge. The other 80 were examined, classified, photographed and/or sketches drawn⁽²⁾. The "side hot spot" failure classification was predominant, accounting for 64 of the 80 identified. Although none were weighed, an estimate of weight loss for each was made from the photographs, sketches and written descriptions made upon examination.

Table I lists the number of ruptured fuel elements experienced from 1955 through 1964.

TABLE I

NUMBER OF RUPTURED FUEL ELEMENTS
1955 through 1964

1955	188
1956	167
1957	200
1958	173
1959	74
1960	138
1961	88
1962	93
1963	69
1964	97

The ruptured element frequency and weight losses sustained in 1964 were compared to similar data from 1955 through 1958, the period studied by Schwendiman and McCormack⁽¹⁾. There were almost twice as many fuel element failures during each

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of those earlier years as there were in 1964. About half of the ruptured elements of the earlier period were classified then as "severe" because of large penetrations of clad sides and fragmenting of the metal. The estimated weight loss on that kind of fuel element failure was 150 grams, while the others were estimated to have lost 9 grams each. The average weight loss during these four years was about 12 kg or almost five times the total weight loss of 2.4 kg in 1964. The reduction in the number of ruptured elements per year and the loss of irradiated rupture debris per failure can be largely attributed to the improvements in design and fabrication of the fuel elements. As a result of three reactors being discontinued, the number of failures in the future can be expected to be further reduced.

Significant Fission Product Discharge to the Columbia River

There are two sources of fission products which contribute to the total fission product activity in the Columbia River. Uranium is naturally present in the river water used to cool the reactors. This uranium is subject to the reactor neutron flux and the U-235 fission while passing through. A small but significant portion, somewhat greater than 5%(3)(4), is absorbed on the aluminum surfaces for extended periods of time, developing a retention - release equilibrium with the cooling water. The other source of fission product is the irradiated uranium released from fuel elements when they rupture in the reactors.

Of the fission products which enter the river, iodine-131 is the most significant and is the only one which will be of concern in this report. It is the only one of all radionuclides which concentrates in the thyroid gland of humans, and its maximum permissible concentration in water is lower than for any of the other fission product radionuclides.

An estimated 2400 grams of irradiated uranium containing 920 curies of I-131 were lost from fuel elements as a result of the 97 cladding failures during 1964. The uranium lost from any single fuel element ranged from a few grams to a maximum of about 100 grams with an average of 25 grams per failed element.

It is assumed that all of the I-131 contained in the uranium lost was delivered directly to the river. This will over-estimate the I-131 release because a portion of it will remain occluded in the uranium to be leached at a later date after a significant reduction by radioactive decay. The distribution of the estimated 920 curies released over the year 1964 is shown in Figure 1.

During 1964 about 500 kg of natural uranium in the cooling water passed through the eight reactors creating about 800 curies of I-131. Most of the fissioning occurs while uranium is retained on the tube and fuel element surfaces, releasing the I-131 into the effluent water. The amount of I-131 varies throughout the year, which can be seen in Figure 1.

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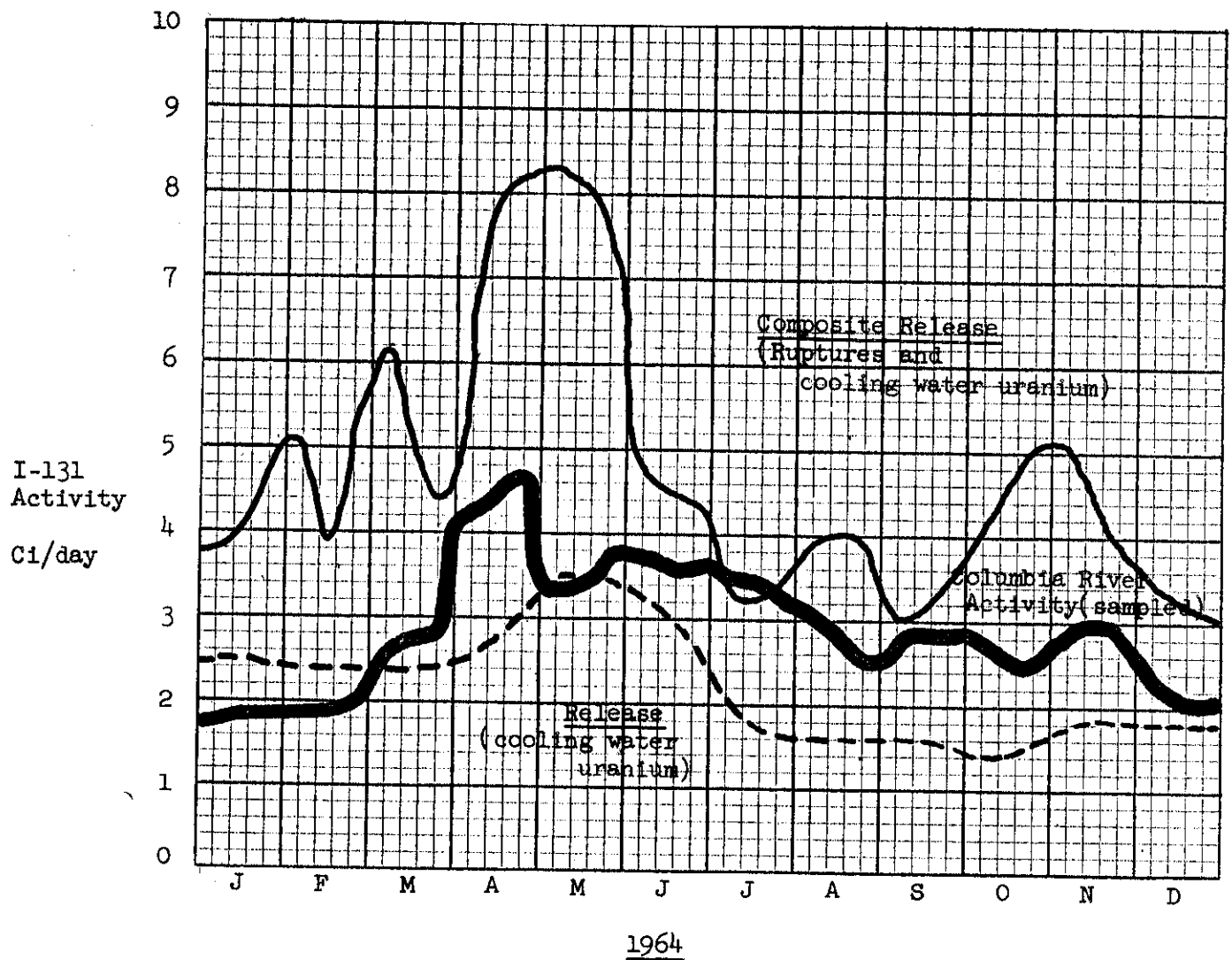
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Figure 1

EFFLUENT COOLING WATER I-131 ACTIVITY
COLUMBIA RIVER I-131 ACTIVITY



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River sampling devices, installed and operated by Hanford Laboratories personnel, composited water samples during all of 1964. These weekly integrated samples were collected off-shore near the 300 Area from prior to the first of the year until mid-August. Starting in mid-June and continuing after the end of 1964, samples were also collected with a similar device at the Richland Water Plant intake. These samples were analyzed for I-131 as well as several other radionuclides.

The amount of I-131 in the river was determined by the Hanford Laboratories to be about 1100 curies for 1964⁽⁵⁾. The rate of transport is shown in curies per day during the year 1964 in Figure 1.

Contribution from Ruptured Fuel

The amount of I-131 released into the coolant stream from the fuel element ruptures was determined from the weight of irradiated uranium lost and the I-131 concentration in the uranium.

The weight loss was determined by reviewing records of the 97 individual ruptures. Most of the ruptures appeared to have lost about 25 grams, two appear to have lost as much as 100 grams, fourteen were classified in the 50 to 75 gram range and twenty-two in the 1 to 10 gram range, for a total of 2.4 kg and 25 grams each on the average.

The fuel element most susceptible to cladding failure is located in the core tubes just downstream of the maximum flux. In the old reactors this is a "Wescott" flux of 4.4×10^{13} n/cm²/sec corresponding to a power level of 6.3×10^3 watts/lb. or 13.8 watts/gram.

The equilibrium amount of I-131 created in 2.4 kg of irradiated uranium at this power level is:

$$\begin{aligned} \text{I-131} &= (2.4)(10^3)(13.8)(84.2)(330)(10^{-6}) = 920 \text{ Ci} \\ &= \frac{920}{2400} = 0.383 \text{ Ci/g} \end{aligned}$$

This result is probably conservative because all of the failures do not occur within this high a power level region.

The average daily release to the river, assuming all of the I-131 is released, was determined from the I-131 in the uranium lost from each failed element. The results are plotted in Figure 1, based on the average for a 28-day period so they can be compared with other results.

Severe Failure, 1963

The failure of a fuel element in the KE Reactor on May 12, 1963, resulted in the largest single release of fission products to the river experienced at Hanford⁽⁶⁾. About 450 grams of uranium was lost from the element.

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The automatic Columbia River monitor at 300 Area showed a rise in river activity which lasted 12 to 16 hours, peaking about four hours after arrival of the leading edge of the activity. The peak activity was about 60% above the background level. No ruptured fuel element during 1964 showed up so distinctly at the 300 Area river monitoring station.

The pound of irradiated uranium which was lost from the element is indicative of release of 170 curies of I-131. The cumulative sample taken at the 300 Area during the period May 6 to May 13, 1963, showed a concentration of 44 pCi I-131/l and a transport of 100 curies of I-131 past the point during the period.

Contribution from Uranium in Cooling Water

The amount of I-131 generated from the uranium present in the cooling water, was determined from the Np-239 water analysis⁽⁷⁾⁽⁸⁾⁽⁹⁾⁽¹⁰⁾ by direct ratio. During 1964, 2×10^5 curies of Np-239 was present in the reactor cooling water effluent. Assuming an average holdup time in the reactor of about 400 hours, the I-131/Np-239 ratio is 0.004. This ratio is based on the build up of Np-239 from neutron capture by U-238 and the I-131 from the fission of U-235 in the neutron flux present outside the fuel elements in the coolant tube⁽¹¹⁾. The assumed holdup time is based on work by both Perkins⁽⁴⁾ and Silker⁽³⁾⁽¹²⁾ and confirmed by the ratio of I-131 to Np-239 found by reactor effluent sample analysis⁽⁷⁾⁽¹³⁾⁽¹⁴⁾⁽¹⁵⁾ in 1963 and early 1964.

The amount of I-131 from coolant water uranium during 1964 is:

$$\text{I-131} = (2 \times 10^5)(0.004) = 800 \text{ Ci.}$$

The amount of Np-239 present in the reactor cooling water effluent varied throughout the year, and it is assumed the I-131 varies in direct proportion. The monthly contribution of I-131 based on the Np-239 was calculated and the results plotted as Ci/day in Figure 1.

Significance of the 1964 Fission Product Release

The total activity release from the reactors to the river was in the order of 6×10^6 curies corrected to a four hour decay. The total release of fission products was about 3.9×10^4 curies, using the same decay correction, or about 0.7% of the total activity.

It is estimated that ruptured fuel elements contributed 920 curies of I-131 to the river. In addition the natural uranium present in river water used for cooling the reactors, upon irradiation contributed about 800 curies of I-131 making a total contribution of about 1700 curies.

The average river flow rate during 1964 was 131,500 cu ft/sec⁽⁵⁾. With the above releases of I-131 to the river, the average concentration of I-131 in the river water totaled 14.5 pCi/l, 7.7 pCi/l from fuel element ruptures and 6.8 pCi/l from the irradiation of the uranium in the cooling water. The 1100 curies measured

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in the transport at Richland transposes to a concentration of 93 pCi/l⁽⁵⁾.

Federal Radiation Council Report No. 2 states that an average daily intake of 80 picocuries of I-131 meets the RPG for the thyroid for averages of suitable samples of an exposed population group of 0.5 rem per year. Considering a liter and a half the average daily fluid intake for the standard man, the average daily intake rate from river water containing the I-131 estimated to be released to the river was 22 pCi. The average daily intake rate for measured values in the river water at Richland was 14 pCi.

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