



UNITED STATES ATOMIC ENERGY COMMISSION
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May 23, 1973

Mr. R. Jerrel Everett
Directorate of Regulatory
Operations - Region I
U. S. Atomic Energy Commission
970 Broad Street
Newark, New Jersey 07102

Dear Jerry:

Enclosed is a report of the special study carried out on the Oyster Creek February rad waste composite. Similar problems were identified in the Cubitainer and glass bottles used on May 2, 1973, for collection of the special samples from Oyster Creek's Waste Tank B. Additional studies on various samples from other nuclear power plants indicate that fractionation of radionuclides between filtrate, filterable material, and container walls is a serious common problem.

As you may already know, the information will be presented by Claude Sill at the Albany meeting, May 22 thru 24, 1973. A summary report on all the studies carried out to date will be made available soon. We are giving this problem top priority and will keep you informed of our progress.

Very truly yours,

Donald R. Percival
Acting Chief, Chemistry Section
Analytical Chemistry Branch
Health Services Laboratory

Enclosure:
Report

B/232

DISTRIBUTIONS OF MANGANESE-54 AND COBALT-60
IN THE HEALTH SERVICES LABORATORY SPLIT OF AN
OYSTER CREEK RAD WASTE COMPOSITE SAMPLE

Health Services Laboratory
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April 1973

On April 13, 1973, Dr. David McCurdy, State of New Jersey Department of Environmental Protection, advised the Health Services Laboratory (HSL) that his manganese-54 and cobalt-60 results on the February 1973 rad waste composite from Oyster Creek were considerably higher than the HSL results. The two laboratories' results for strontium-90, cesium-134, and cesium-137, however, agreed quite well. Dr. McCurdy suggested that the HSL check for losses of manganese-54 and cobalt-60 on the walls of the polyethylene Cubitainer in which the sample was sent. Dr. McCurdy stated that his splits were collected in glass bottles and analyzed within two days after collection.

The sample had been made 10% volume/volume with concentrated HNO_3 on or before the collection date of March 2, 1973. On March 9, the HSL performed a gamma spectral analysis on an unfiltered 400-ml aliquot. On April 13, the 210 ml of sample remaining in the Cubitainer was filtered through a 0.45-micron membrane filter, and the activity remaining in the empty container was subsequently removed quantitatively with successive acid washes. Each fraction was analyzed by gamma spectrometry. The radionuclides identified and their distributions on April 13, 1973, are presented in Table I.

The HSL results for manganese-54 and cobalt-60 on March 9 and April 13, 1973, are presented in Table II. The March 9 results were obtained on a 400-ml aliquot of unfiltered sample. Unfortunately, the aliquot had been discarded and was not available for reexamination on April 13. The April 13 results were obtained on the filtrate from the 210 ml of sample remaining in the Cubitainer on that date. The results on the filtered and unfiltered samples agree surprisingly well. Apparently, the insoluble material in the March 9 aliquot was not representative of nor proportional to the insoluble material present in the Cubitainer on April 13.

Estimates of the manganese-54 and cobalt-60 concentrations attributable to filtrate, filtered material, and walls of the Cubitainer are shown in Table III. The estimates were based on the following assumptions:

- (1) The filtrate of April 13, 1973 was identical to the unfiltered 400-ml aliquot analyzed on March 9, 1973.
- (2) The original volume of the sample, although not precisely known, was assumed to have been 900 ml.

- (3) No significant losses of manganese-54 and cobalt-60 activities associated with filterable material occurred in the aliquots taken for various analyses between March 9 and April 13, 1973. The concentrations of manganese-54 and cobalt-60 attributable to filterable material, therefore, are simply 1/900 of the total activities determined on April 13, 1973.
- (4) The manganese-54 and cobalt-60 activities adhering to the walls of the Cubitainer had reached a maximum by March 9 and remained constant thereafter. Therefore, the equivalent concentrations are 1/900 of the total activities determined on April 13, 1973.

Correction of the totals in Table III for dilution of the sample with HNO_3 and for decay of manganese-54 gives $(11.6 \pm 1.3) \times 10^{-6}$ $\mu\text{Ci/ml}$ for manganese-54 and $(25.7 \pm 1.3) \times 10^{-6}$ $\mu\text{Ci/ml}$ for cobalt-60. The New Jersey results were $(11.2 \pm 0.5) \times 10^{-6}$ $\mu\text{Ci/ml}$ and $(26.7 \pm 1.3) \times 10^{-6}$ $\mu\text{Ci/ml}$. The ratios, HSL/New Jersey, are 1.04 ± 0.13 and 0.96 ± 0.07 for manganese-54 and cobalt-60, respectively. The HSL cobalt-60/manganese-54 ratio is 2.22 ± 0.27 and New Jersey's ratio is 2.38 ± 0.16 .

The agreement between the original New Jersey results and the HSL results of April 13, 1973, is probably fortuitous since the latter were obtained using rather tenuous assumptions with data on the remaining 23% of a sample that was 42 days old. However, the important point is the identification of a serious problem affecting the entire laboratory intercomparison program. Preliminary studies indicate that a similar and equally serious problem exists with samples collected in Cubitainers at other reactor facilities using the procedure recommended by the HSL. That procedure involves the addition of 20 ml of concentrated hydrochloric acid and 100 mg of NaHSO_3 per liter of sample. Dr. McCurdy suggested the use of glass rather than plastic containers as a possible solution to the problem. Accordingly, the HSL has requested the special collection of two samples of the April rad waste composite from Oyster Creek, one in glass and one in a Cubitainer. Additional studies are being made by the HSL to solve this problem. Results and recommendations will be made available as soon as possible.

TABLE I. DISTRIBUTION OF ACTIVITIES ON APRIL 13, 1973

Fraction	Net Cpm								
	^{54}Mn	^{60}Co	^{58}Co	^{51}Cr	^{134}Cs	^{137}Cs	^{95}Tb	^{53}Fe	^{124}Sb
Filtrate ¹	10.1±1.2	26.8±1.5		4.9±2.1	98±3	121±3			
10-min 70-ml aqua regia wash ²	29.8±1.0	38.5±1.1	3.1±0.6	3.7±0.7		3.7±0.6	2.0±0.5	0.9±0.2	0.2±0.1
10-min 70-ml concentrated HCl + H ₂ O ₂ wash	13.8±0.6	15.9±0.6	1.2±0.3			0.9±0.3		0.4±0.1	
70-ml dilute HCl-HF Over weekend	26.0±0.8	39.6±0.9				2.5±0.5	2.0±0.4		
Overnight	2.2±0.3	3.9±0.3							
SUBTOTAL WASHES	71.8±1.4	97.9±1.6				7.1±0.8			
Filter ³	14.0±1.0	22.8±1.1				2.1±0.6			
Washes + Filter	85.8±1.7	120.7±1.9	4.3±0.7	3.7±0.7		9.2±1.0	4.0±0.6		
TOTAL CPM	95.9±2.1	147.5±2.4	4.3±0.7	8.6±4.9	98±3	130.2±3.2	4.0±0.6	1.3±0.2	0.2±0.1
Σ in Filtrate	10.5±1.3	18.2±1.1							
Σ on Filter	14.6±1.1	15.5±0.8							
Σ on Bottle	74.9±2.2	66.4±1.5							

¹Activity in total 210 ml of filtrate based on analysis of a 70-ml aliquot.

²All acid washes were evaporated to 70 ml for gamma spectrometry.

³The membrane filter was wet-ashed and diluted to 70 ml for gamma spectrometry.

TABLE II. MANGANESE-54 AND COBALT-60
CONCENTRATIONS, $\mu\text{Ci/ml}$

	<u>March 9, 1973</u>	<u>April 13, 1973</u>
^{54}Mn	$(3.8 \pm 0.2) \times 10^{-6}$	$(3.2 \pm 0.3) \times 10^{-6}$
^{60}Co	$(1.24 \pm 0.03) \times 10^{-6}$	$(1.13 \pm 0.07) \times 10^{-6}$

TABLE III. ESTIMATES OF MANGANESE-54 AND COBALT-60 CONCENTRATIONS
IN THE ORIGINAL SAMPLE ASSUMING HOMOGENEITY

<u>Fraction</u>	<u>Activity Attributable to Each Fraction</u>			
	<u>^{54}Mn</u>		<u>^{60}Co</u>	
	<u>$\mu\text{Ci/ml} \times 10^6$</u>	<u>%</u>	<u>$\mu\text{Ci/ml} \times 10^6$</u>	<u>%</u>
Filtrate	3.2 ± 0.3	34 ± 5	11.3 ± 0.7	49 ± 4
Filter	1.0 ± 0.2	10 ± 2	2.2 ± 0.2	10 ± 1
Cubitainer	<u>5.3 ± 1.0</u>	56 ± 12	<u>9.6 ± 1.0</u>	41 ± 5
TOTAL	9.5 ± 1.1		23.1 ± 1.2	